THE HONG KONG POLYTECHNIC UNIVERSITY
香港理工大學

DEPARTMENT OF
CIVIL AND ENVIRONMENTAL ENGINEERING
土木及環境工程學系

RESEARCH CENTRE FOR
RESOURCES ENGINEERING TOWARDS CARBON NEUTRALITY
±RCRE 碳中和資源工程研究中心

Co-organiser:

Joint Laboratory on
Solid Waste Science
固體廢棄物科學聯合實驗室

Supporting Organisation:

rilem

ReACT 2024

The 1st International Conference on Research and Application of Carbonation Technology for Wastes and Concrete

11-14 December 2024

Conference Proceedings

CONTENTS

Appreciation to Sponsorship

NELD

耐 尔 得 智 能 科 技

Beijing NELD Intelligent Technology Co., Ltd

Founded in 2006, NELD is a Chinese national high-tech enterprise focusing on the research, development and service of automated equipment for concrete material testing, customized equipment for scientific research, and intelligent laboratory system.

Plenary Sessions

Prof. Changwen Miao

Academician of Chinese Academy of Engineering
Professor
School of Materials Science and Engineering
Southeast University
China

Green and Low-carbon Construction Materials

Civil engineering materials constitute the foundational basis for significant national infrastructure projects in sectors including transportation, construction, water resources, energy, and defense. The advancement of the "dual carbon" strategy and the emergence of new engineering structure construction have put forward higher requirements for civil engineering materials. This report will introduce the low carbon cementitious materials, the low carbon concrete products and the high performance concrete. Aligned with the national dual-carbon strategy and the exceptional demands of future major engineering projects on the performance of civil engineering materials, we proposed a new paradigm based on deconstruction, reconstruction, and upgrade-construction research approaches. This paradigm introduces the innovative concept of creating cement-based biomimetic metamaterials (CBMs) through biomimetic sequential fabrication, anchored in the cementitious gene (C-S-H). It advocates cross-scale architectural techniques and significant performance leap methodologies for the biomimetic microstructure of CBMs. Incorporating data-driven and high-throughput screening methodologies for intelligent design, this approach aims to surmount the scientific and technological bottlenecks associated with integrating multiple materials, multi-scale sequential architectures, multi-functionality, and multi-scenario applications in CBMs. The development of this new class of biomimetic CBMs, exemplified by biomimetic superhydrophobic concrete, biomimetic high-strength and toughness concrete, cement-based biomimetic aerogels/hydrogels, cement-based biomimetic photothermal metamaterials, promises to revolutionize the mechanical and functional capabilities of civil engineering materials. The report provides reference for meeting new demands of the construction industry and coping with the challenges of carbon neutrality.

Biography

Miao Changwen is full professor of Southeast University in P.R.China, and present board chairman of Jiangsu Research Institute of Building Science. In 2011, He was selected as the member of Academician of the Chinese Academy of Engineering which is the highest honour for the Chinese scientists and engineers who have made great contributions to and excellent achievements in the field of engineering. He owns authorized 18 Chinese patents, and is author of 101 academic papers and 3 monographs.

Over the past nearly 20 years, Prof. Miao has dedicated his life to R&D of concrete admixture and high performance cementitious material. By means of diligent and pioneering works, he made outstanding contributions to the industrial development of cement and concrete in China. He made the 100 percentage of converting rate from achievements in scientific research to industry application. Besides, he created the total output value of approximately 313 million

Euros and the profit tax of 62.5 million Euros. His achievements in science and technology have been successfully used in more than 100 national key engineering projects of China, such as Three Gorges Project, Su-Tong Yangtze River Bridge, Tianwan nuclear power station, Nanjing subway and Qinghai-Tibet Railway.

Prof. Jos Brouwers

Full Professor and Chair of Building Materials
Department of the Built Environment
Eindhoven University of Technology
The Netherlands

Circularity and Carbonation of Building Materials

In this presentation the smart recycling of concrete and the positive role of carbonation is addressed. Smart recycling means new crushing and separating techniques to obtain flows of relatively pure aggregates and cement stone, whereby the latter is tried to separate further in reacted and unreacted cement. Treatment of the flows, such as their accelerated carbonation, are introduced. Besides the material and processing techniques, also new characterization methods are presented to assess the phase composition of generated flows.

Biography

Prof. Jos Brouwers is a Full Professor and Chair of Building Materials at Eindhoven University of Technology (TU/e), The Netherlands. He obtained his MSc in Mechanical Engineering and his PhD in Applied Physics at TU/e. His research interests include physics of particle packing (granular building materials, and amorphisation of metals and metalloids); inorganic chemistry and mineralogy of cementitious materials; physics of suspension rheology; chemical reactions (heterogeneous, dissolution and precipitation); synthesis and application of nano-engineered materials (e.g. silica, aerogel and titania); biomaterial-cementitious composites; computational modelling of particle packing, cementitious hydration and carbonation reactions; and recycling and minerals processing. Prof. Brouwers is also a Visiting Professor at State Key Lab of Silicate Building Materials at Wuhan University of Technology, China, where he focuses on construction materials and sustainable building. So far, he co-authored 290 publications in peer-reviewed journals, of which 32 single-authored, that have been cited more than 25,000 times with h-index of 82.

Prof. Fazhou Wang

Vice President & Professor
School of Materials Science and Engineering
Wuhan University of Technology
China

Innovative Pathways to Transform CO_2 into Carbon Mineralized Materials

Carbon mineralized materials have emerged as a promising approach to combat climate change by sequestering flue gas contained CO_2 in stable mineral form. The process of carbon mineralization involves the reaction of CO_2 with metal-bearing minerals or industrial waste materials to form stable carbonate minerals. This approach offers several advantages, including permanent CO_2 storage, potential for large-scale implementation, and the production of valuable materials for various applications. In this presentation, the speaker will share some thoughts on the development and application of carbon mineralized materials in China and aboard, with a particular focus on the innovative WUT pathways to create and use carbon mineralized materials from two unique perspectives.

Biography

Prof. Fazhou Wang has been engaged in the academic research and engineering application of high-performance cement and concrete materials. He has undertaken over 40 national and provincial projects, including National Key Research & Development Project, National 863 Project, and National Natural Science Foundation. Prof. Fazhou has published 3 books (1 of which is a co-author) and more than 100 SCI papers. He has received 2 National Awards for Science and Technology Progress and 6 provincial and ministerial 1st prizes. He serves as vice president of Wuhan University of Technology, director of State Key Laboratory of Silicate Materials for Architectures, vice president of “China Concrete and Cement-based Products Association” and chairman of “Hubei Province Ceramic Society”. He has won the Youth Science and Technology Award of China Silicate Society, Youth May Fourth Medal of Hubei Province and was financially supported by the “Outstanding Youth Fund of National Natural Science Foundation”.

Dr Maciej Zajac

Principal Scientist
Global Research and Development Department
Heidelberg Materials AG
Germany

From Idea to Industrial Application: Enforced Carbonation of Recycled Concrete Paste

Circularity and decarbonization are crucial for achieving a sustainable future, particularly in the building materials industry. Achieving a circular and carbon-neutral concrete production is of utmost importance as it is the most used construction material worldwide.

This presentation focuses on a carbon capture, storage and utilization (CCSU) approach for CO_2 mineralization using recycled concrete. The technology encompasses three key elements: the selective separation of old concrete, the enforced carbonation of the resulting fine fraction known as recycled concrete paste (RCP), and the development of new composite cements incorporating carbonated RCP.

Selective separation aims at efficiently breaking down old concrete and separating it into its original constituents - gravel, sand, and hardened cement paste. This mechanical process utilizes attrition-driven soft crushing, favouring cracking of the cement paste and separating it from the surfaces of gravel and sand. Notably, this process incurs minimal energy consumption resulting in low environmental impacts and processing costs. Trials have proved the technology, with first commercial applications now emerging.

Enforced carbonation leverages the spontaneous reaction between cement hydrates and CO_2 known as concrete carbonation. Contrary to concrete, the small particle size of RCP and the higher CO_2 concentration in flue gases greatly accelerates the process. In less than thirty minutes, more than 80% of process emissions from the original clinker production can be captured under ambient conditions, transforming RCP carbonation into an efficient carbon capture technique. During carbonation, calcium from cement hydrates reacts with CO_2 forming stable calcium carbonate, similar to limestone and hence directly storing the captured CO_2. Alumina and silica from hydrates transform into an alumina-silica gel, a highly reactive pozzolan suitable for use in composite cements. After laboratory-scale development and investigation, the process is well-understood and industrial optimization is underway, with the first dedicated carbonation reactors being constructed.

Carbonated recycled concrete paste has demonstrated rapid pozzolanic reactivity in composite cements during research projects. Unlike typical pozzolanic materials that require weeks of curing time, the alumina-silica gel fully reacts within the first few days of hydration. This enables higher clinker replacement rates while achieving satisfactory early and final compressive strengths. These findings have facilitated the development and industrial testing of these innovative cement formulations, with tests conducted in concrete plants validating their viability and advantages without requiring modifications to existing concrete technologies.

Enforced carbonation has the potential to be a cost-competitive CCSU technology for the cement industry, which is fully circular on top. Its energy and cost efficiency, combined with the generation of value-added carbon-negative materials from waste, make it highly attractive. This technology, initiated in the laboratory, has now reached a mature stage, enabling its first commercial applications.

Biography

Maciej Zajac serves as a Principal Scientist in the Global Research and Development Department at Heidelberg Materials, one of the world's largest building materials companies, based in Heidelberg, Germany. He spearheads research initiatives to innovate sustainable products, contributing towards climate protection and yielding added value for both customers and the company.

In 2007, Maciej Zajac earned his Ph.D. in Physical Chemistry from Burgundy University, Dijon, France. Subsequently, he joined HeidelbergCement as a Junior Scientist. In 2021, he successfully completed his habilitation procedure in the field of Chemical Engineering at AGH University, Cracow. Serving as a Principal Scientist, Maciej guides and oversees several pivotal R&D projects, including collaborations with external partners. His current research emphasizes circular economy and CO_2 utilization via mineral carbonation in the construction industry, development of the modern composite cements and alternative binders.

To his credit, Maciej Zajac has co-authored over 70 scientific articles in peer-reviewed journals and holds more than 40 patents.

Prof. Takafumi Noguchi

Professor
Department of Architecture
The University of Tokyo
Japan

Ideal Carbon-neutral Concrete with Permanent Circulation of Calcium and Carbon Dioxide

Concrete will play a major role in establishing a carbon-neutral society in 2050. Even if the cement, the main material of concrete, can be produced using renewable energy, the amount of carbon dioxide generated by the calcination of limestone will not change. In addition, concrete without cement clinker is being developed, but it cannot be completely carbon neutral because the raw materials for the binder are often industrial by-products, or the production of the raw materials requires a large amount of energy. On the other hand, concrete is the second most used substance after water, and a large amount of concrete has been produced to date, with further increases in production expected in the future. Concrete contains a large amount of calcium, and the carbon dioxide separated from the calcium is present in large quantities in the atmosphere. If these can be combined to produce new concrete, it will not only contribute to reducing the concentration of carbon dioxide in the atmosphere, but it can become a permanently recyclable concrete. This keynote speech will provide an overview of the "Research and Development of Calcium Carbonate Circulation System in Construction," a moonshot R&D project funded by the Japanese government. The project aims to construct structures using calcium carbonate concrete produced solely from waste concrete and carbon dioxide. Currently, high-speed carbonation of waste concrete by direct air capture has been achieved, calcium carbonate concrete of 30 MPa can be produced, and structural members made from it have been confirmed to have sufficient toughness. In addition, calcium carbonate concrete is a carbon-negative construction material and is expected to achieve resource recycling that is not problematic from an LCA perspective, making calcium carbonate concrete an ideal CCUS technology.

Biography

Takafumi Noguchi graduated from the University of Tokyo in 1985 and received his Ph.D. degree at the same university. He became an assistant professor at the University of Tokyo in 1988, then an associate professor in 1998 before becoming a full professor at the same university in 2014 until now. He has served as Vice President of Architectural Institute of Japan for two years starting in 2021, as Vice President of Japan Concrete Institute since June 2024, and as Chair of ISO/TC71/SC8 (Environmental Management for Concrete and Concrete Structures) since 2018. He is a six-time recipient of the Japan Cement Association's Paper Award, as well as a recipient of paper awards from Japan Concrete Institute and Society of Materials Science, Japan. He also received Best Work Award from Japan Concrete Institute in 2016, from ACI in 2017, and from fib in 2018, respectively, for the realization of building using environmentally friendly concrete. Fifty-six researchers received their Ph.D. degrees under his supervision. His current recent research interests include development of carbon-neutral concrete, sustainable recycling of concrete structures, optimum supply-chain of resources and wastes in concrete, and performance assessment and conservation of historical concrete structures.

Prof. Chi-sun Poon

Michael Anson Professor in Civil Engineering, Chair Professor of Sustainable Construction Materials, Head of Department of Civil and Environmental Engineering and Director of Research Centre for Resources Engineering towards Carbon Neutrality
The Hong Kong Polytechnic University
Hong Kong

Achieving Carbon Neutrality through Sustainable Construction Materials and Technologies

Biography

Prof. Chi-sun Poon specializes in the research and development of environmental-friendly construction materials, waste management, waste recycling technologies, concrete technologies and sustainable construction. He is an Editor of Construction and Building Materials since 2014. He is also an Editorial Board Member of Cement and Concrete Composites since 2017. Prof. Poon has published over 800 papers in international journals and conferences (including 650 international journal papers, 160 referred conference papers, and 7 book chapters). His current h-index is 122 with the total number of over 50,000 citations in Scopus. He has obtained more than 20 patents related to concrete and waste recycling technologies. He has been listed among the World’s Top 2% Scientists released by Stanford University. He was awarded the State Technological Innovation Award 2017 (2nd Class).

Prof. Liwu Mo

Professor
State Key Laboratory of Materials-oriented Chemical Engineering
College of Materials Science and Engineering
Nanjing Tech University
China

Accelerated Carbonation of Ca/Mg-rich Solid Wastes: Reaction Mechanism, Products Design and Industrial Application

Accelerated carbonation of Ca/Mg-rich solid wastes in terms of steel slag, magnesium slag, etc, could yield various carbonates and other hydraulic products, which provides a potential approach to toward efficient waste valorization coupled with economical CO_2 sequestration. The carbonation mechanism of steel slag, in particular the carbonation behaviours of various minerals phases, under the semi-dry state and in suspension solution as well as the microstructures of relevant products are introduced. Various Ca/Mg carbonate products in terms of precast blocks, artificial aggregates and carbonated supplementary cementitious materials can be designed based on the carbonation reaction mechanism. The microstructures and properties of the carbonate products are summarized. In addition, some typical case studies on the industrial applications of accelerated carbonation technology are also presented.

Biography

Liwu Mo is a full professor at State Key Laboratory of Materials-oriented Chemical Engineering, and College of Materials Science and Engineering, Nanjing Tech University. He is a director of concrete disease research center at Nanjing Tech, and leads a research group on the developing and efficiently using low-carbon/high-performance cement and concrete materials. His research interest also covers wastes valorization, special and functional engineering materials, CO_2 sequestration and utilization. He served more than 500 key concrete projects including dams, airports, bridges, tunnels, high-speed railways, energy infrastructures, etc. in China. He has published more than 100 peer-reviewed papers, 2 book chapters, and owned 15 authorized patents, of which 7 patented technologies/products have been successfully commercialized.

Prof. Thomas Matschei

Chair of Building Materials
Institute of Building Materials
RWTH Aachen University
Germany

CO_2 Beneficiation of German Lignite Fly Ash under Different Reaction Conditions and its Reactivity as SCM

Matschei, T.[1], Felten, C.[1], Schwedt, A.[2], and Vollpracht, A.[1]

[1]RWTH Aachen University, Institute of Building Materials
[2]RWTH Aachen University, Central Facility for Electron Microscopy
Email: matschei@ibac.rwth-aachen.de

Reducing direct CO_2 emissions in cement production is a primary goal of the cement industry. Replacing clinker with industrial by-products as supplementary cementitious materials (SCMs) is an effective and widely adopted strategy to achieve this. Traditionally, hard-coal fly ash and blast furnace slag have been the most common SCMs. However, due to industrial transformations in Europe, their availability has decreased, necessitating the exploration of alternative SCMs.

So far lignite fly ash (LFA) has been utilized only to a limited extent of < 5 % in Germany as a building material or in road construction due to its highly variable composition, high contents of quicklime and periclase, and fluctuating sulfate levels [1]. This study aims to transform potentially harmful oxides within LFA into stable carbonates through carbonation, thereby sequestering CO_2. Additionally, the carbonation of Ca-Si-Al phases is targeted, as the resulting Si-Al-containing gel exhibits pozzolanic reactivity. This process has the potential to enhance reactivity of the beneficiated fly ash. Finally a use as reactive SCM replacing CO_2 intense Portland clinker [2] in the final binder is envisaged.

Within the frame of the project CO2Treat a comprehensive parameter study was conducted on four different LFAs from western and eastern Germany using both wet and moist reactors. Detailed chemical and mineralogical analyses were performed on selected samples. Figure 1 shows the complex microstructure of an LFA determined with the aid of electron backscatter diffraction (EBSD). Multiphase particles with inclusions of quicklime, periclase and brownmillerite, among others, can be seen. The parameter study revealed significant variations in the carbonation behavior of the LFAs. Chemical-mineralogical analyses indicated the carbonation of a sulfur-containing, amorphous glass phase and the concurrent influence of hydration reactions. Subsequent R^3 tests on carbonated samples demonstrated significantly enhanced reactivity.

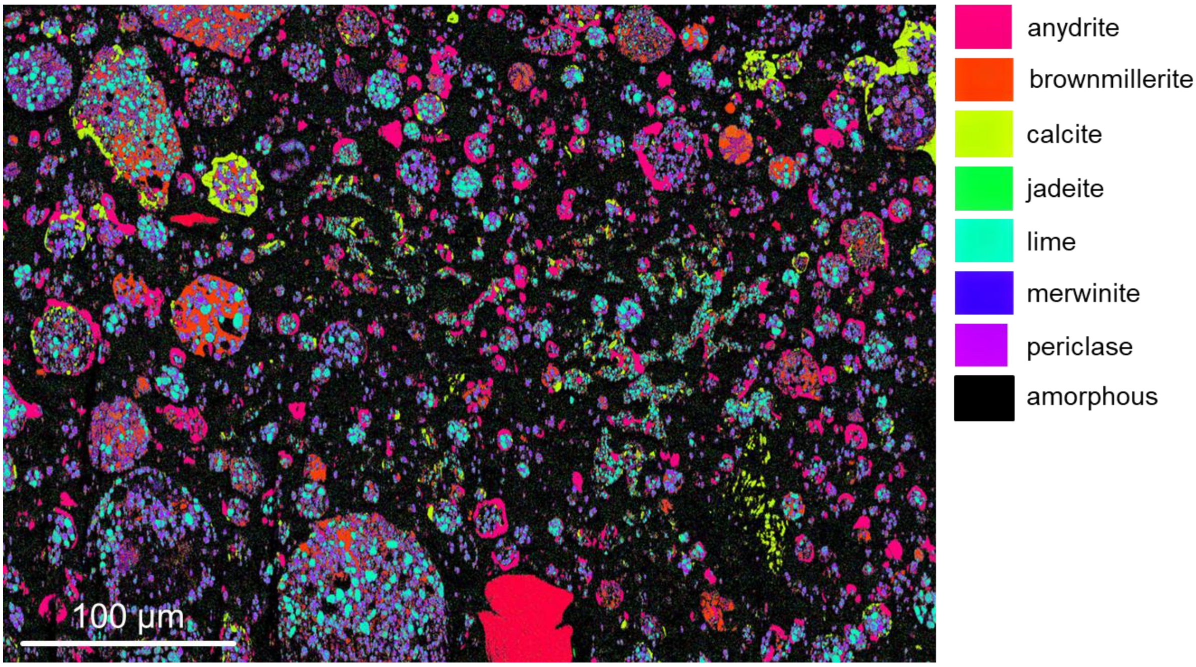


Figure 1: EBSD image of an LFA before carbonation

Keywords

Mineral carbonation; lignite fly ash; CO_2-negative binder; wet carbonation; moist carbonation

Acknowledgements

C.F. would like to acknowledge the support by BMBF (FKZ: 033RU015A; ERA-MIN3 CO2Treat JTC-2021_196)

References

1. Sören Steger, M.R., Winfried Bulach, Doris Schüler, Izabela Kosinska, Stefanie Degreif, Günter Dehoust, Thomas Bergmann, Peter Krause, Rüdiger Oetjen-Dehne, *Stoffstromorientierte Ermittlung des Beitrags der Sekundärrohstoffwirtschaft zur Schonung von Primärrohstoffen und Steigerung der Ressourcenproduktivität*. 2019, Umweltbundesamt: https://www.umweltbundesamt.de/kraftwerksnebenprodukte-steinkohleflugaschen-rea#sekundarproduktion.
2. He, Z., et al., *Comparison of CO2 emissions from OPC and recycled cement production.* Construction and Building Materials, 2019. **211**: p. 965-973.

Session A1

Advanced Technology in Carbonation

NEXT GENERATION “GREEN CONSTRUCTION MATERIALS” DEVELOPED BY MINERAL CARBONATION OF STEEL SLAGS

Quaghebeur, M.[1], **Nielsen, P.**[1], **Kamyab, H.**[1], **Peys, A.**[1], **and Horckmans, L.**[1]

[1] Vlaamse Instelling voor Technologisch Onderzoek (VITO), Mol 2400, Belgium
Email: mieke.quaghebeur@vito.be

Abstract

Mineral carbonation is recognized as a prominent carbon sequestration technique, characterized by the transformation of Ca/Mg-rich feedstocks, notably those containing calcium (Ca) and magnesium (Mg), into stable carbonates through reaction with CO_2. This process converts CO_2 - a molecule with low energy - into solid mineral carbonates, a reaction that is thermodynamically advantageous, and can occur at temperatures close to ambient. Given the vast scale of the construction materials market, which produces more than 50 billion tons of concrete and mineral aggregates each year, mineral carbonation is deemed one of the most substantial and energy-efficient methods for CO_2 utilization. The technology harbors the potential to create a variety of “green construction materials” such as pre-cast products, aggregates, and reactive fillers, while also offering the chance to valorize a broad spectrum of alkaline residues, including mine tailings, steel slags, fly ashes, and waste cement (figure 1).

Figure 1: Smart value chains are needed to make mineralization plants successful.

Despite this, the integration of CO_2 into construction materials and related technologies remains nascent. This can be explained by the fact that the exploitation of industrial residues as resources presents inherent challenges due to their fluctuating composition, which can vary over time and across different production facilities. Typically, these materials require processes such as comminution and beneficiation to render them suitable for use as secondary raw materials.

The steel industry is currently confronted with the imperative to mitigate CO_2 emissions. An innovative approach to address this challenge involves the sequestration of CO_2, coupled with the valorization of by-products. This dual strategy not only aims to reduce the environmental footprint of steel production but also enhances the value of industrial waste. In Europe, the steel industry produces various types of slags, which are by-products of the steel manufacturing process. The types of steel slags commonly produced include:

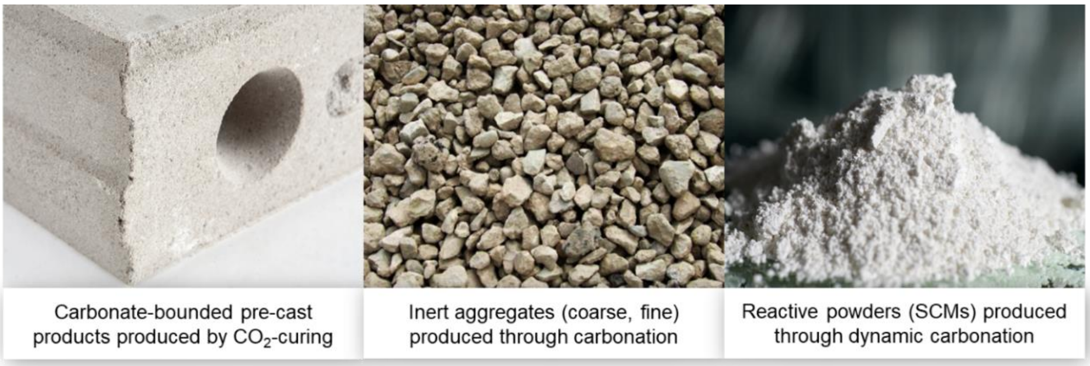
- Granulated Blast Furnace Slag (GBFS): This material is produced during the iron-making process when iron ore, coke, and limestone are melted in a blast furnace. The slag contains mainly calcium oxide, silicon dioxide and aluminum oxide and is typically rapidly cooled, or "quenched," with water to prevent the formation of crystalline phases.

- Electric Arc Furnace (EAF) Slag. This type of slag is produced when scrap metal (steel or stainless steel) and fluxes are melted using an electric arc furnace. The primary constituents of EAF slag are silicates (predominantly calcium and silicon). Additionally, it contains significant amounts of magnesium and might contain iron, chromium, molybdenum, and nickel depending on its origin.
- Basic Oxygen Furnace (BOF) Slag. Generated during the process of converting pig iron into steel with the basic oxygen process. This material is notably abundant in iron content. It also comprises a variety of other elements, including calcium and minor amounts of silicon and magnesium, which are derived from the lime-based fluxes used in the steel-making process and the oxidation of impurities from the pig iron.

The European steel industry is undergoing a transformation process, aiming to reduce CO_2 emissions by substituting the traditional blast furnace/BOF route with more sustainable methods like Direct Reduced Iron (DRI) combined with EAF or Submerged Arc Furnace (SAF), heated with renewable energy. This shift is expected to introduce new types of slags with differences in chemical and mineralogical composition compared to the slags currently produced.

Steel slags are increasingly recognized as a valuable resource for CO_2 sequestration and the production of green construction materials. The inherent chemical properties of steel slags make them suitable for reactions with CO_2 to form stable carbonates. Different types of steel slags are distinguished by their unique chemical and mineralogical compositions, as well as the presence of various trace elements. These distinctions necessitate the development of dedicated pre-treatment methodologies to adequately prepare the slags for valorization. The inherent properties of each slag type dictate the most appropriate valorization strategies and the potential construction products that can be synthesized.

This paper examines the foremost mineral carbonation strategies for the valorization of distinct steel slag types into construction materials. This paper will explore the research findings and commercial potential related to the most promising mineral carbonation valorization pathways for different types of steel slags, with a particular emphasis on how the characteristics of the steel slags influence the chosen valorization route. The examination of mineral carbonation will be restricted to semi-dry carbonation processes that have been scaled to industrial levels or have undergone pilot scale trials (Figure 2).



Carbonate-bounded pre-cast products produced by CO_2-curing

Inert aggregates (coarse, fine) produced through carbonation

Reactive powders (SCMs) produced through dynamic carbonation

Figure 2: Next generation "green construction products" based on mineral carbonation

Keywords
Mineral carbonation; steel slags; smart value chains; green construction products

ACCELERATED CARBONATION THROUGH HIGH TEMPERATURE OF CEMENT-BASED MATERIALS TOWARDS LOW CARBON CONSTRUCTION

Dianchao Wang[1], **Takafumi Noguchi**[1], **Takayuki Hayakawa**[2]

[1]Department of Architecture, Graduate School of Engineering, The University of Tokyo
[2]Central Research Laboratory, Taiheiyo Cement Corporation
Email: wdc@g.ecc.u-tokyo.ac.jp

Abstract

With increasing CO_2 emissions from heavy industries posing a growing threat to the environment, there is an urgent need to develop CO_2 sequestration technologies, especially those that can fix CO_2 rapidly. In this study, the CO_2 uptake performances of hydrated cement paste under various environments parameters coupling with high temperature were investigated through accelerated carbonation experiments. The results show that high temperature is an effective way to improve CO_2 sequestration efficiency, in which the waste heat accompanied by the emitted CO_2 can be used as the source of high temperature. The mechanisms of carbonation at varying vapor levels have been studied with a focus on the transportation of carbonate substances at around 100ºC. The proportion of hydrated products that contribute to calcium carbonate varies based on the level of vapor present, and higher rates of carbonation typically result in the formation of meta-stable calcium carbonate, like aragonite. Additionally, various operating parameters, such as carbonation time, sample size, vapor content, and water-to-cement ratio, have been examined to determine their impact on the carbonation reaction. These parameters were compared to their effects at ambient temperature. It was found that these factors play a crucial role in the carbonation reaction. The study concluded that carbon dioxide sequestration was highly efficient at 600ºC in samples, even with a large size and a low water-to-cement ratio.

Keywords
Accelerated carbonation; high temperature; CO_2 sequestration; cement-based materials; parameter investigation

1. Introduction

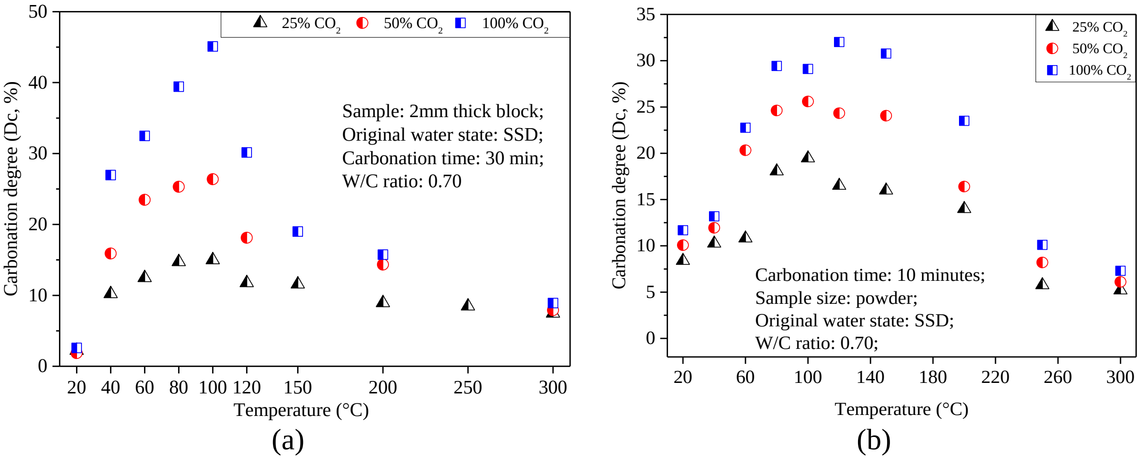
Reducing greenhouse gas (GHG) emissions, especially CO_2, is the core component of containing global temperature growth. CO_2 removal would be used for extra emissions compensation to achieve net negative emissions and further decrease the global mean temperature. The CO_2 sequestration amount of cement-based materials is significant, whereas the low carbonation speed prohibits further application. Cement production requires both high temperatures and enormous flow rates, entailing the consumption of large amounts of energy. In this study, further investigations of the effect of temperature coupling with other parameters (CO_2 concentration, vapor content etc.) on the carbonation reaction of hydrated cement paste have been conducted. The results and conclusions could provide a meaningful reference for developing CO_2 sequestration technologies.

2. Experimental

In this study, cement pastes were adopted for the carbonation test. The specific gravity of the cement is 3.14 g/cm^3, and the Blaine surface area is 4490 cm^2/g. Three water to cement ratios of 0.40, 0.55, and 0.70 were adopted in this study. Two original water content statuses of the cement paste samples were adjusted before carbonation: saturated surface dry (SSD) state and fully dry (FD) state. The SSD is the condition of the sample in which the surface is dry, but the inter voids are saturated with liquid water. The FD state samples were obtained after drying in a 60°C nitrogen environment until constant mass. For the recycled aggregates, the water content in the original aggregates before carbonation is around 1.7%. Thermogravimetric analysis (TG) was adopted for the measurement of CO_2 content of the samples. The device type is TAPS300S.

3. Results and discussion

The CO_2 absorption results of the block samples under different temperatures and CO_2 concentrations are shown in Figure 1, and the carbonation time is 30 minutes for block samples and 10 minutes for powder samples. The results in Figure 1(a) show that the temperature influence laws on different CO_2 concentrations are the same: the carbonation degree in a specific CO_2 concentration environment increases with temperature and peaks at around 100°C, then decreases with the growing carbonation temperature. The maximum carbonation degrees under different CO_2 concentrations occur at 100°C. At 20°C, the carbonation degrees are close to 2.0% under different CO_2 concentrations, and similar characteristics can be found at 300°C, in which the carbonation degrees at around 7.5~8.0%. These phenomena indicate that CO_2 concentration has a limited influence on the carbonation performance at 20 °C and 300 °C. When comparing the results in Figire 1(a) with that in Figure 1(b), the temperature shows similar influence law on the carbonation performance of powder and blocks. The carbonation degree increases with temperature, and reaches the peak value at around 100°C~120 °C. After reaching the peak value, the carbonation degree experiences a sharp falling with growing temperature for block samples, whereas the reduction rate of powder samples is much slower until 150 °C.



(a) (b)

Figure 1. Carbonation performance of cement paste at different CO_2 concentrations and temperatures: (a) block samples; (b) powder samples.

4. Conclusion

In this research, the CO_2 sequestration performance of hydrated cement paste was tested and evaluated, and the carbonated samples were further evaluated by different methods. The increased carbonation temperature could accelerate the carbonation reaction of cement paste to a significant extent, and the optimum temperature is 100°C. Increasing CO_2 concentration, reducing sample size, and adjusting the water content through vapor content and appropriate liquid water addition under high temperatures could enhance the CO_2 sequestration efficiencies.

EFFECTS OF POLYMORPHS OF CALCIUM CARBONATE ON COMPRESSIVE STRENGTH OF CALCIUM CARBONATE CONCRETE

Masahito Tada[1], Yuji Mitani[1], Hikotsugu Hyodo[1], and Ippei Maruyama[2]

[1] Taiheiyo Cement Corporation, Japan
[2] The University of Tokyo, Japan
Email: Masahito_Tada@taiheiyo-cement.co.jp

Abstract

Reducing greenhouse gas emissions is a global priority and cement production contributes 5-8% of the world's anthropogenic CO_2 emissions. These emissions are mainly due to energy consumption during firing and CO_2 release from limestone decarbonization. Therefore, the developing of CO_2 reduction technologies in the cement and concrete sector is crucial. Maruyama et al. (2021) proposed calcium carbonate concrete (CCC) as a new construction material. It uses demolished concrete, water, and CO_2. This material hardens by supplying a calcium bicarbonate solution between particles of demolished concrete powder and precipitating $CaCO_3$ from the solution by temperature changes. $CaCO_3$ exists in three crystal polymorphs: Calcite, Vaterite, and Aragonite, each with different densities and morphologies. The presence of these polymorphs in different proportions may affect the bonding strength between particles in CCC, and consequently, the compressive strength. Therefore, this study aims to reveal the influence of different ratios of $CaCO_3$ polymorphs in cement paste powders on the compressive strength of CCC.

The carbonated cement paste powder to simulate demolished concrete was used as the material for the CCC in this study. The cement paste cured in water at 20 °C for 28 days was dried, crushed and sieved to a maximum particle size of 0.6mm. The obtained powder was subjected to carbonation to adjust the ratio of $CaCO_3$ polymorphs. The carbonation conditions were set at 5% of CO_2 with 20, 45 and 60°C (hereafter, named as 20°C-5%, 45°C-5%, and 65°C-5%, respectively), and 80% of CO_2 with 45°C (hereafter, named as 45°C-80%). The carbonated powder was mixed with 10% water, then filled into cylindrical molds with a diameter of 1cm and a height of 2cm and compacted by compression at 10MPa. After demolding, the specimens were cured using a three-cycle immersion-drying process. The process involved immersing the cylinders in a calcium bicarbonate solution with magnesium sulfate for 2 hours, followed by drying them at 105°C for at least 12 hours per cycle, to produce the CCC. The compressive strength of CCC was measured, a content of each $CaCO_3$ polymorph was calculated by TG-DTA and XRD/Rietveld method, and the morphologies of the fracture surface of CCC were observed by SEM.

The results showed that the compressive strength of CCC after immersion-drying curing was the highest at 37.9 MPa for 45°C-5%, followed by 31.1 MPa for 60°C-5% and 21.9 MPa for 20°C-5%. The compressive strength of CCC using 45°C-80% was the lowest at 12.4 MPa. However, after immersion-drying curing, the increase in $CaCO_3$ content was highest at 45°C-80% and lowest at 45°C-5%.

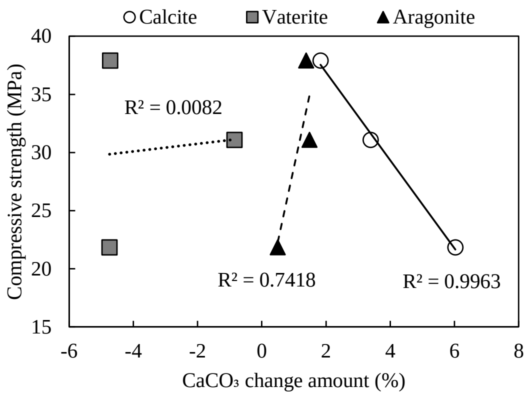


Figure 1 Relationship between the compressive strength of CCC and the amount of change in $CaCO_3$

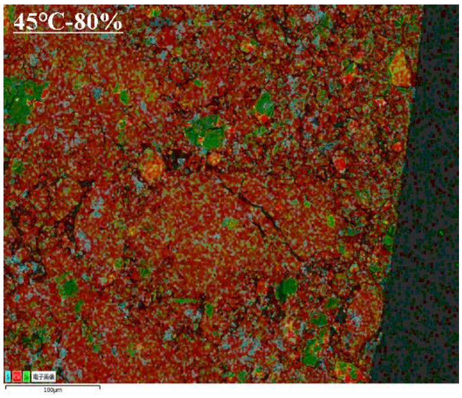


Figure 2 EDS mapping images of polished surface of CCC
(Red: Ca, Green: Si, Blue: S)

Figure 1 shows the relationship between the compressive strength of CCC with 20°C-5%, 45°C-5%, and 65°C-5% and the amount of change in the $CaCO_3$ polymorphs after immersion-drying curing. There is a high correlation between the increase in aragonite content and compressive strength. In addition, according to the SEM observations, the needle-shaped aragonite on the particle surface was entangled with aragonite on the surrounding particles. Therefore, it is believed the correlation is due to an increase in the contact area and the stress transfer between the aragonite generated on the particles.

The amount of aragonite increased the most in the CCC using 40°C-80%. However, no aragonite generated on the particle surface of CCC using 40°C -80% from the SEM observation. In addition, according to Figure 2, a large amount of Ca was distributed inside the particles of the 45°C-80% than that of 45°C -5%. These suggest the possibility of aragonite precipitation inside the particles. This precipitation did not contribute to particle bonding and may have resulted in the lowest compressive strength of 40°C-80%.

In addition, a strong liner correlation was found between the amount of hemihydrate gypsum produced during immersion-drying curing and the compressive strength. This is probably due to the addition of magnesium sulfate to the calcium bicarbonate solution. The hemihydrate gypsum was probably formed by the reaction of magnesium sulfate with calcium hydroxide or C-S-H.

Keywords

Calcium carbonate concrete; CO_2; compressive strength; $CaCO_3$ polymorphs; pressure molding; recycle concrete

Acknowledgements

The authors acknowledge the financial support from the NEDO Moonshot Research and Development Program, ”C^4S Research and Development Program, Calcium Carbonate Circulation System for Construction" (Project Manager: Prof. Takafumi Noguchi, the University of Tokyo) The authors would like to acknowledge the support and valuable insights provided by Prof. Takafumi Noguchi University of Tokyo, Prof. Manabu Kanematsu from Tokyo University of Science, and all members of both research laboratories.

Reference

I. Maruyama et al (2021): A New Concept of Calcium Carbonate Concrete using Demolished Concrete and CO_2 , Journal of Advanced Concrete Technology, Vol.19, pp.1052-1060

EX-SITU MONITORING OF CARBONATION OF CEMENT PASTE USING RAMAN MICROSPECTROSCOPY

Park, J.[1] **and Moon, J.**[1,2]

[1]Department of Civil and Environmental Engineering, Seoul National University, Korea
[2]Institute of Construction and Environmental Engineering, Seoul National University, Korea
Email: pjs2sl@snu.ac.kr

Abstract

Accelerated carbonation of cement is getting more attention as a solution to sequestrate atmospheric CO_2 into minerals. Conventional characterization methods, such as thermogravimetry, powder X-ray diffraction, and pH measurement, have been used, but they may not be ideal methods for investigating the carbonation kinetics because they rely on bulk measurements. This study aims to monitor the carbonation of cement paste using ex-situ Raman microspectroscopy with submicron spatial resolution. The spectral features of the hydration and carbonation product were analyzed during the process. The phase transformations of clinker, C-S-H, $Ca(OH)_2$, as well as other types of hydrates (e.g., AFt and AFm), are spatially and temporally resolved to compare carbonation rates between phases in the cement system. Finally, the carbonation kinetics are studied by analyzing the spatial relationships of these phases.

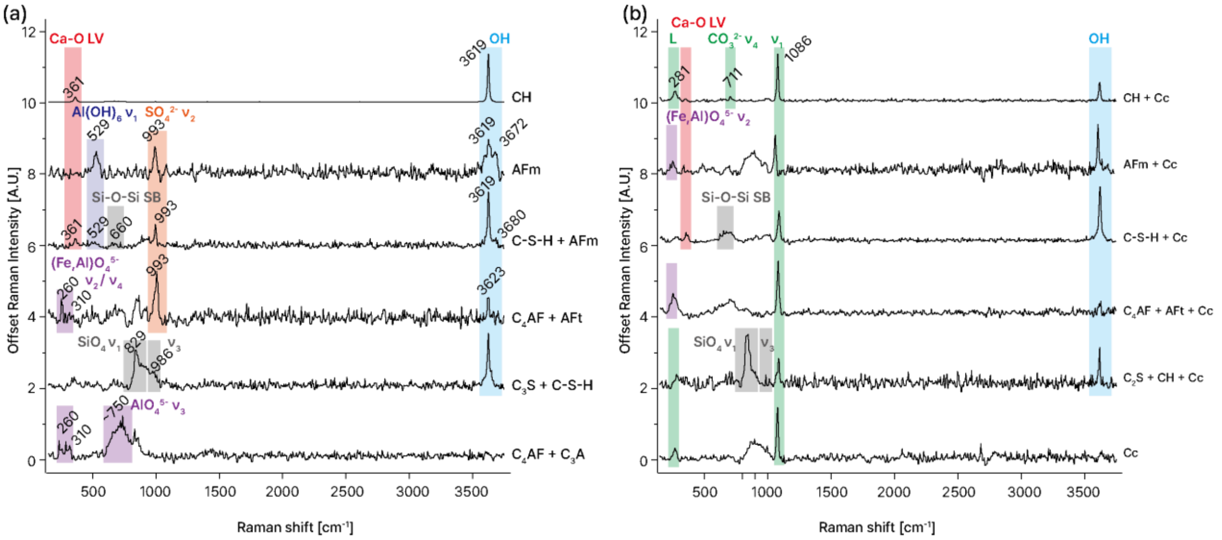


Figure 1. Representative spectra with peak assignments at specific measurement locations of (a) cement paste hydrated for 7 days and (b) cement paste carbonated for > 1 day. The peak position and vibrational modes are annotated.

Keywords

Cement carbonation; ex-situ monitoring; raman microspectroscopy; carbonation kinetics

Acknowledgments

This research was supported by the Basic Science Research Program through the National Research Foundation of Korea (NRF), funded by the Ministry of Education (RS-202300248910).

A MILD STRATEGY TO IN-SITU SYNTHESIZE STRONG BULK CO_2 MINERALIZED MATERIAL CONTROLLED BY ORGANIC TEMPLATE

Chen, J.[1, 2], **Zhao, M.**[1, 2], **Liu, Z.**[1, 2, 3], **Wang, F.**[2, 3] **and Hu, S.**[2]

[1] School of Materials Science and Engineering, Wuhan University of Technology
[2] State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology
Email: liuzc9@whut.edu.cn

Abstract

Traditional high temperature fabrication method for ceramics suffers from significant energy consumption, and limits the development of novel ceramic composites with the integration of temperature-sensitive material. Although bioinspired mineralization provides an effective strategy to realize room temperature preparation of ceramics, large-scale fabrication is still a challenge. Herein, we demonstrate that a low temperature procedure for the fabrication of large size ceramics by using the carbonation reaction of sodium alginate (SA) doped γ-dicalcium silicate (γ-C_2S) compacts. Mesocrystals with specific orientation were created by the precisely control of the nucleation, growth and assembly of calcium carbonate, through the in-situ carbonation reaction of CO_2 using SA as an organic template. Molecular interactions and microscopic crystal alignment were regulated by this bioinspired in-situ mineralization process, which results in the formation of a CO_2 mineralized material with exceptional mechanical properties rivalling that of biological materials. Furthermore, we show that the advanced ceramic composites with compressive strength approaching 300 MPa can be fabricated in large size by using this in-situ carbonation strategy, with the added benefits of fixing 197.2 kg CO_2 per ton of CO_2 mineralized material during its manufacturing. Our method provides an alternative to traditional sintering methods that should be attractive for the efficient and economical fabrication of temperature-sensitive, high-performance biomaterials.

Keywords

Bioinspired materials; biomineralization; carbon mineralization material; mechanical properties; mesocrystals

PROLONGED IN-SITU CO_2 MIXING FOR ENHANCING DIRECT CO_2 ABSORPTION AND COMPRESSIVE STRENGTH OF CEMENTITIOUS COMPOSITE

Won Kyung Kim[1], **Junboum Park**[1], **and Juhyuk Moon**[1]

[1]Department of Civil and Environmental Engineering, Seoul National University, Seoul
08826, Republic of Korea
Email: semgua@snu.ac.kr

Abstract

The release of substantial quantities of greenhouse gases has resulted in unusual climate patterns, garnering worldwide environmental concerns. Consequently, there is a growing necessity to utilize carbon dioxide (CO_2) generated by various industries. This research adopts in-situ CO_2 mixing technology as a method for incorporating gaseous CO_2 into freshly mixed cement paste. The in-situ CO_2 mixing is a representative mineral carbonation technique of carbon capture, and utilization (CCU), which actively promotes the calcium carbonate ($CaCO_3$) precipitation in cementitious mixtures [1].

The phenomenon is attributed to the formation of a direct reaction between calcium ions (Ca^{2+}) released from the dissolution of cement phases and dissociated carbonate ions (CO_3^{2-}). The strong alkaline condition of the cementitious mixture facilitates the chemical reaction [2]. The continuous ionization of CO_2 and subsequent precipitation of $CaCO_3$ seeds result in a denser matrix and accelerated early hydration. Consequently, the early compressive strength of the paste could be improved, alongside active CO_2 absorption during the fresh stage, enabling it as a potential CCU technique.

This study tried to maximize the CCU capacity of in-situ CO_2 mixing during the fresh stage. Various mixing environments have been tested with different mixing time and potential additives. As a result, the CO_2 absorption was significantly increased from 1.6 vol.% to 4.6 vol.%. Stable CO_2 sequestration was confirmed through a quantitative evaluation of uptaken CO_2 content by thermogravimetric analysis (TGA) and thermodynamic modeling.

Additionally, measurements of paste fluidity, surface temperature, and compressive strength at 1, 3, 7, and 28 days were conducted to assess the changes in the mechanical performance due to the in-situ CO_2 mixing. The results revealed a 79% enhancement in early compressive strength and a 23% recovery of strength after 28 days by extending CO_2 mixing time. Furthermore, comprehensive X-ray diffraction (XRD) and TGA analyses were performed to investigate the impact of CO_2-rich system on the development of hydration products and mechanical performance during the later curing stages.

Keywords

In-situ CO_2 mixing; carbon dioxide; mineral carbonation; CCUS; calcite

Acknowledgements
This work was supported by Korea Institute of Energy Technology Evaluation and Planning (KETEP) grant funded by the Korea government (MOTIE) (20212010200080, Development of In-situ carbonation technology using CO_2 related from cement industry).

References

[1] Hepburn, C., Adlen, E., Beddington, J., Carter, E. A., Fuss, S., Mac Dowell, N., ... & Williams, C. K. (2019). The technological and economic prospects for CO_2 utilization and removal. *Nature*, *575*(7781), 87-97.

[2] König, M., Vaes, J., Klemm, E., & Pant, D. (2019). Solvents and supporting electrolytes in the electrocatalytic reduction of CO_2. *Iscience*, *19*, 135-160.

STUDY ON THE MECHANISM OF $NaHCO_3$-PROMOTED $CaSiO_3$ CARBONATION KINETICS AND MECHANICAL PROPERTY ENHANCEMENT

Chengbo Wei[1], **Zengmei Wang**[1]

[1]School of Materials Science and Engineering, Jiangsu Key Lab. of Construction Materials,
Southeast University, Nanjing 211189, PR China
Email: 18764296519@163.com (C. Wei); 101011338@seu.edu.cn (Z. Wang)

Abstract

Silicate cement stands as a pivotal raw material for national economic development. Nevertheless, the cement production process has long grappled with two significant challenges: high emissions of waste gases and excessive energy consumption [1,2]. With the national strategic goals of "carbon peak" and "carbon neutrality" in mind, addressing these issues becomes increasingly urgent.

To reduce the energy consumption in cement manufacturing and minimize CO_2 emissions, the development of low-calcium silicate carbonation-hardened materials as alternatives to Ordinary Portland Cement (OPC) is imperative. $CaSiO_3$ (CS) possesses lower hydration activity and higher carbonation reactivity, serving as an ultra-low-calcium CO_2 curing adhesive that not only exhibits favorable mechanical properties but also enables CO_2 sequestration. In this study, $NaHCO_3$ was introduced into the CS carbonation system to enhance the carbonation reactivity. Compressive strength was tested, and various analytical methods including XRD, TGA, carbonation reaction heat, FT-IR, SEM, BSE-EDS, pH, electrical conductivity, Zeta potential were used to investigate the impact of $NaHCO_3$ on carbonation hardening performance, reactivity, carbonation products, and microstructural characteristics. The results indicated that the introduction of $NaHCO_3$ significantly increased CS compressive strength (up to 101.58 MPa) and carbonation degree, evident in the early peak of the carbonation heat curve and the overall heat release. After carbonization under water, unreacted particles were enveloped by silica gel. The continuous layer of $CaCO_3$ formed on the outer surface of the gel constituted the framework of the carbonated calcium silicate product, with calcite as the exclusive crystalline phase of $CaCO_3$. Conversely, in the presence of $NaHCO_3$ solution, both calcite and spindle-shaped aragonite crystals were produced. Aragonite crystals were interwoven within the crevices of calcite, forming a denser matrix structure, imparting mechanical reinforcement. Additionally, by calculating the reaction activation energy, it was found that in the $NaHCO_3$ solution medium, the reaction activation energy of CS was 3.752 kJ/mol, significantly lower than 8.3669 kJ/mol in the water medium, suggesting the catalytic role of $NaHCO_3$ in enhancing carbonation reactivity. These findings hold the potential to provide additional insights for the enhancement of sustainable binders based on calcium silicate, which offer reduced carbon emissions.

Keywords
$CaSiO_3$; CO_2 sequestration; Carbonation; $NaHCO_3$

References

[1] E. Benhelal, E. Shamsaei, M.I. Rashid, Challenges against CO_2 abatement strategies in cement industry: A review, Journal of Environmental Sciences (China). 104 (2021) 84, 10.1016/j.jes.2020.11.020.

[2] M. Zajac, I. Maruyama, A. Iizuka, J. Skibsted, Enforced carbonation of cementitious materials, Cem. Concr. Res. 174 (2023) 107285, 10.1016/j.cemconres.2023.107285.

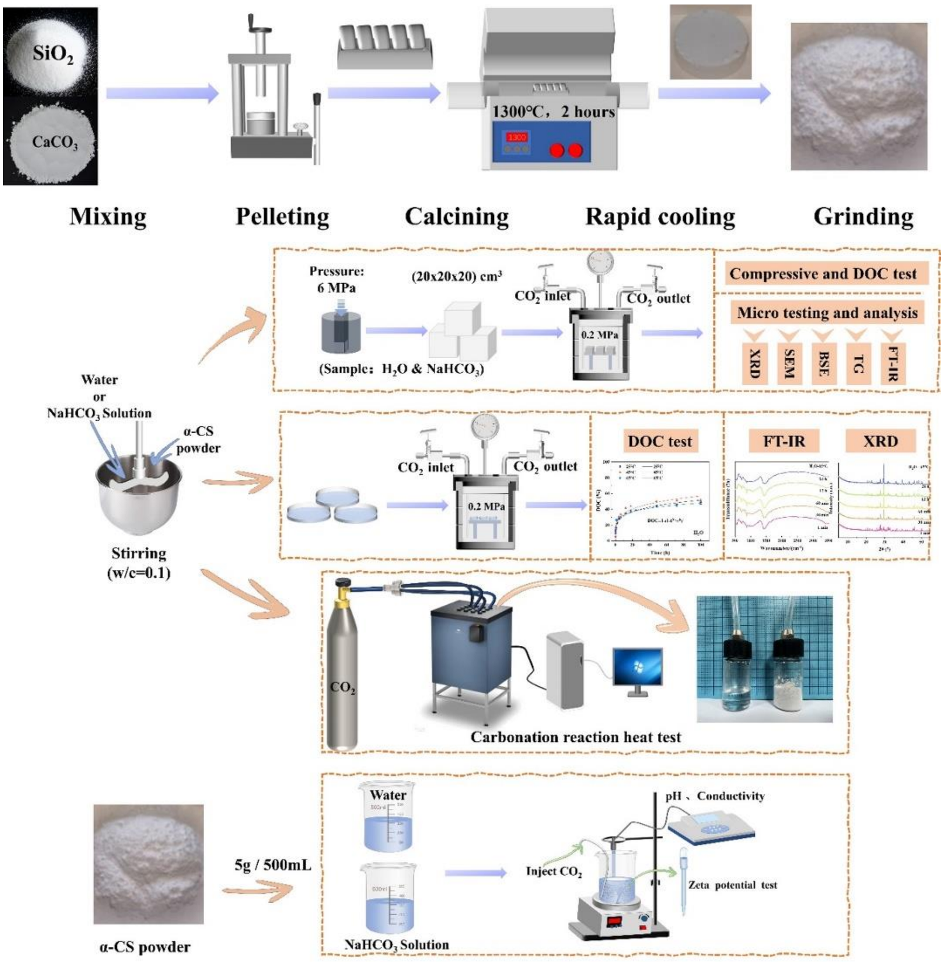


Fig. 1. Flow chart of the synthesis and main carbonization process of α-CS

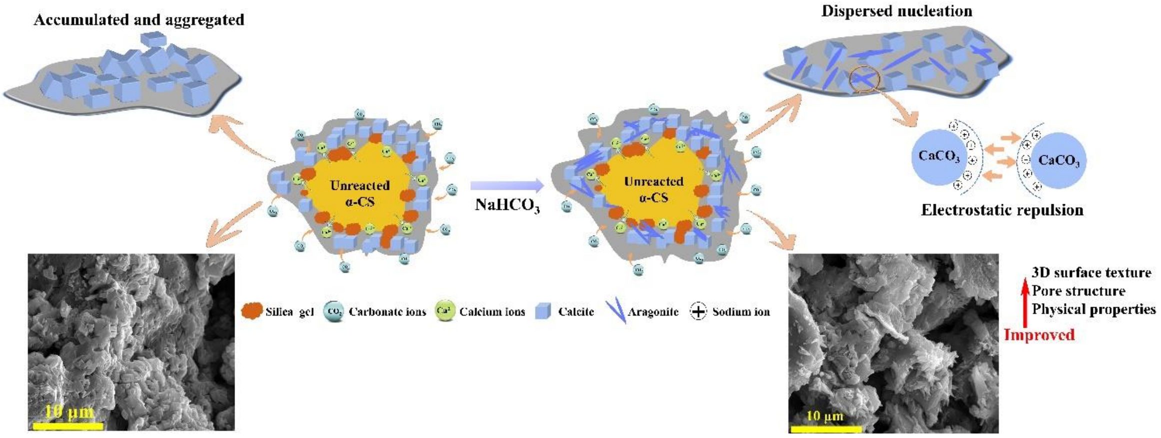


Fig. 2. Schematic diagrams of carbonation mechanism of CS and enhancement mechanism of CS by $NaHCO_3$

CARBONATION MECHANISM OF BOFS UNDER HIGH-GRAVITY CONDITIONS AND OPTIMIZATION OF THE CARBON REMOVAL PROCESS

Qifeng Song[1], Tung-Chai Ling[1]

[1] College of Civil Engineering, Hunan University, Changsha, China
Email: tcling611@yahoo.com

Abstract

China, as one of the largest steel producers, emits over 2 billion tons of CO_2 annually and generates substantial amounts of solid waste of basic oxygen furnace slag (BOFS). Fortunately, the high Ca and Mg ion content of BOFS can be a highly reactive precursor for carbon sequestration. Hence, achieving a balance between high CO_2 removal efficiency, energy consumption, and costs- effectiveness is of paramount important.

A rotating packed bed (RPB) can create a high-gravity (higee) field via centrifugal acceleration, thereby promoting physical reaction efficiency. The gas-liquid micro-mixing efficiency and mass transfer performance in an RPB are significantly improved. This study explores the phase assemblage evolution mechanism of BOFS under higee carbonation conditions. The results shown in Figure 1 indicates that the thermodynamic advantages of $Ca(OH)_2$ (CH)-rich BOFS are amplified under higee conditions, achieving the same level of carbonation in 3 minutes compared to conventional aqueous carbonation, which typically requires about 2 hours. This is mainly due to the fact that higee field promotes ex-situ precipitation of $CaCO_3$, increases Ca leaching rates, and enhances gas-liquid mass transfer efficiency. Additionally, this process facilitates the partial carbonation of inert Ca-Fe phases (srebrodolskite), increasing the total Ca conversion rate of BOFS by 25%. In contrast, C_2S-rich BOFS rapidly forms highly polymerized silica gel with a lower Ca/Si ratio, which continuously accumulates on the surface, impeding further carbonation and resulting in a lower carbonation limit compared to conventional aqueous carbonation.

Furthermore, this study optimized the higee carbonation process to achieve high mass transfer efficiency, energy savings, and cost effectiveness, providing recommendations for parameters such as RPB centrifugal acceleration, gas-liquid velocity ratio, and gas flow rate for scale-up application in steel plants (see Figure 2). Systematic graphical presentation identified an acceleration of 430 m/s^2, a gas-liquid velocity ratio of 25-30, and a Q_G/RPB volume range of 4.7-6.6 min^{-1} as the optimal operating conditions for high mass transfer coefficient ($>0.3\ s^{-1}$) and low energy consumption (144-151 kWh/t-CO_2). These parameters serve as a reference for RPB equipment of any scale. Finally, a 1.5-fold reduction in mass transfer performance with prolonged RPB operation was established, and reducing the liquid-to-solid ratio was found to be a cost-effective measure to compensate for efficiency losses. The estimated cost for the higee carbonation process ranged from 106-111 CNY/t-CO_2.

Keywords
Higee carbonation; basic oxygen furnace slag; phase evolution; process optimization

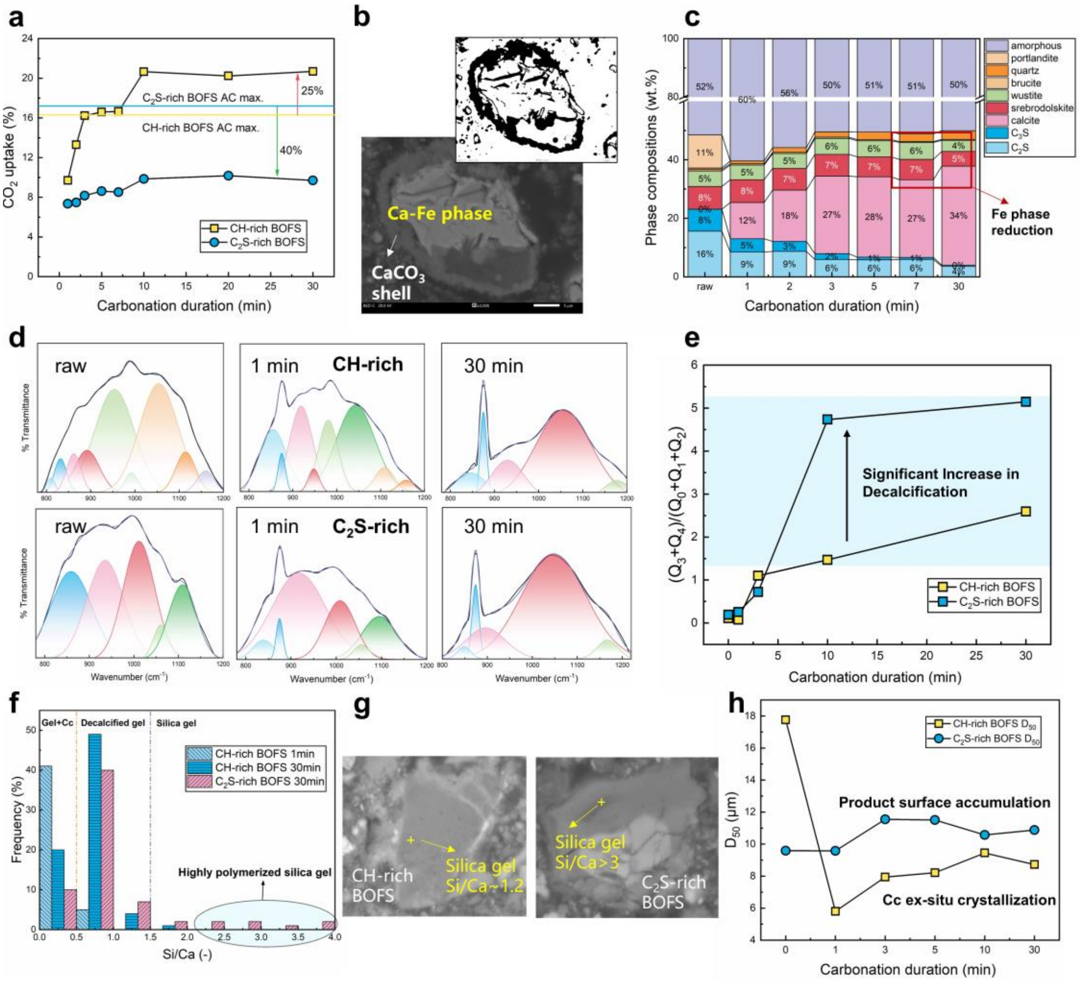


Figure 1. Phase evolution of CH-rich and C_2S-rich BOFS under higee carbonation. **a.** CO_2 uptake; **b.** BSE image of Ca-Fe phase in CH-rich BOFS after 30 min of carbonation; **c.** QXRD of CH-rich BOFS, showing a significant reduction of Fe-containing phases after 30 min of carbonation; d. FTIR partial deconvolution spectra of both BOFS types; **e.** Semi-quantitative results of silica gel based on FTIR deconvolution results; **f.** Si/Ca ratio of carbonated products from both BOFS types; **g.** BSE-EDS images of both BOFS types after 30 min of carbonation; **h.** Particle size variation of both BOFS; **Note: AC in (a) refers to traditional aqueous carbonation.*

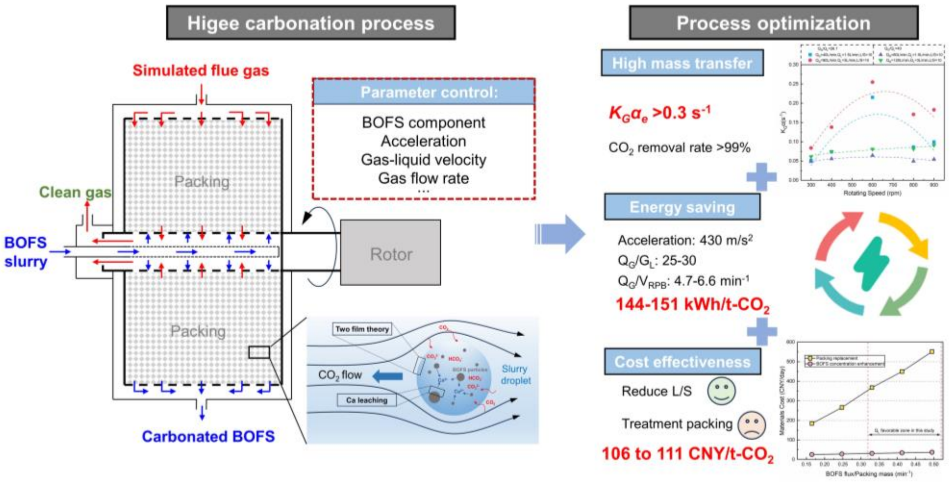


Figure 2. Recommended parameters for industrial application optimization of the higee CO_2 removal process using BOFS as a carbon sink material.

A NOVEL APPROACH FOR IMPROVING AQUEOUS CARBONATION KINETICS WITH CO_2 MICRO- AND NANO- BUBBLES

Yi Jiang[1], Zihan Ma[1], Zhenjiang Gu[1], Faqian Liu[2], Peiliang Shen[1], Chi Sun Poon[1]

[1] Department of Civil and Environmental Engineering & Research Centre for Resources Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hong Kong 999077, China
[2] School of Chemical Engineering and Technology, Sun Yat-sen University, Zhuhai 519082, China
Email: peiliang.shen@polyu.edu.hk; cecspoon@polyu.edu.hk

Abstract

The kinetics of carbonation change dramatically with the mode of gas-liquid-solid interactions. Although prior studies have indicated the significance of enhancing liquid-solid interaction by transferring carbonation from a gaseous environment to an aqueous environment to improve carbonation rate, addressing the rate-limiting gas-liquid mass transfer is becoming vital for further accelerating carbonation.

Therefore, in this study, micro- and nano-bubbles (MNBs) were used as a novel accelerator for recycled cement paste (RCP) carbonation. The rate of calcium carbonate (Cc) precipitation, the CO_2 utilization efficiency, the composition and morphology of the carbonated RCP and the changes in solution chemistry were examined and compared to carbonation using normal bubbles.

The results indicate that the high surface-to-volume ratio and long lifetime of MNBs contributed to the rapid mass transfer of gaseous CO_2 to aqueous carbonate species. It created stronger attractions between the negatively charged MNBs and the calcium ions, which remarkably enhanced RCP dissolution and Cc precipitation. Meanwhile, the huge amount of MNBs and the significant inter-particle collision changed Cc nucleation behaviour and resulted in RCP breakage. These small changes in CO_2-water-RCP interaction induced complete differences in the carbonation kinetics and RCP microstructures, where it was found that (i) carbonation efficiency was improved by ~5 times, (ii) the CO_2 utilization efficiency improved by ~4 times and reached a maximum of 89.2%, (iii) carbonation products were characterized by a more open microstructure with 72.2% higher porosity, and an ultra-fine RCP with the specific surface area of 135 m^2/g was produced. The use of CO_2 MNBs breaks through the rate limitation of aqueous carbonation from a CO_2 dissolution perspective. It contributes to the fundamental understanding of the control over carbonation kinetics and carbonation products.

Keywords
Carbonation kinetics; recycled cement paste; microstructure; micro- and nano-bubbles

ROLE OF PARTIAL LIMESTONE CALCINATION IN CARBONATED LIME-BASED BINDERS

Xiong Qian[1], Peiliang Shen[1], Chuanlin Hu[2], and Chi Sun Poon[1]

[1]Department of Civil and Environmental Engineering, Research Centre for Resources Engineering toward Carbon Neutrality, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong
[2] State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan, China
Email: Xiong.qian@polyu.edu.hk

Abstract

Most carbonation reactions are characterized by products encasing unreacted particles, leading to incomplete reactions and consequently lower material utilization efficiency. This study introduces a novel approach for preparing gradient carbonated materials by partially calcined limestone (PCL), which is calcined below full calcination temperatures to maintain some original limestone while partially transforming into lime. The mechanical strength, phases evolution and microstructure were investigated. The results indicated that the mechanical properties of the materials improve continuously and porosity markedly decreases as the calcination degree of PCL increases, peaking at around 35 %. However, excessive calcination degree impedes the formation of a compact structure. Simultaneously, the carbonation process yields $CaCO_3$ with a lower decomposition temperature, exhibiting two distinctive microstructural features: an encapsulating layer on the surface of unreacted limestone and the tiny particles (<5 μm) scattered between the layers. This study presents a promising approach to carbonated material design, demonstrating that through controlled partial calcination of limestone, opening avenues for more efficient material utilization.

Keywords

Partial calcination; limestone; mechanical properties; core-shell structure

Session A2

Theory of Carbonation

CRITICAL PHENOMENA ON THE ACCELERATION OF CARBONATION OF CEMENTITIOUS MATERIALS

Maruyama, I.[1], Cheng, L.[1], Zhenli, Y.[1], Igami, R[2], Kurihara, R.[1]

[1] Department of Architecture, The University of Tokyo
[2] Department of Environmental Engineering and Architecture, Ngoya University
Email: i.maruyama@bme.arch.t.u-toyo.ac.jp

Abstract

The concrete sector is heavily used and has a large amount of CO2 emission associated with cement production. In recent years, there has been a lot of research investment and development into reducing CO2 emissions from materials. Concrete is considered an essential material for civil infrastructure and buildings. Still, due to these CO2 emissions, technological development is being promoted regarding significant changes in terms of material selection and design in structures, so carbon neutrality in the concrete sector is an urgent issue.

Currently, to reduce the clinker ratio during production, consideration is being given to pulverizing cement waste and industrial by-products containing CaO, carbonating them, and using them as part of the cement, as well as substituting industrial by-products and minerals containing CaO for cement and subjecting them to carbonation curing to create a solidified body.

In many cases, not only CaO but also SiO_2 and Al_2O_3 coexist, making it crucial to understand the behavior of systems including calcium (alumino-) silicate hydrate. This understanding is important for the rapid carbonation process of cement-based materials, optimization of particle size distribution, and effective fundamental technology for selecting curing methods.

This contribution discusses the behavior of C-A-S-H and C-S-H under drying and carbonation, especially microstructural change, moisture content during carbonation, matrix texture, and calcium carbonate polymorphs, and how the colloidal nature of C-A-S-H / CH is important. We also elucidate that the carbonation rate can be governed by moisture transport in the matrix when several conditions are satisfied, which is crucial for the acceleration of semi-dry carbonation.

Keywords

Calcium (alumino-) silicate hydrate; carbonation; moisture content; microstructure

PHASE EVOLUTION AND MICROSTRUCTURE CHANGES INDUCED BY ACCELERATED CARBONATION IN NATURAL HYDRAULIC LIME PASTE WITH GGBFS ADDITION

Dongmin Wang[1], Ze Liu[1], Dong Xu[1], and Guodong Qi[1]

[1]School of Chemical and Environmental Engineering, China University of Mining and
Technology (Beijing), Beijing, 100083, China
Email: wangdongmin@cumtb.edu.cn

Abstract

Natural Hydraulic Lime (NHL) blended with appropriate amounts of ground granulated blast furnace slag (GGBFS) can be applied to more demanding conditions in the restoration of ancient buildings. This study systematically investigates the carbonation depth, phase composition, microstructure, and compressive strength over accelerated carbonation (~3% CO_2) aging periods for NHL compounded with different proportions of GGBFS (S-NHL). The research demonstrates that the densification of the S-NHL microstructure due to the formation of C-S-H, C_3AH_{10}, and $C_4A\hat{C}H_{11}$ from GGBFS pozzolanic reaction reduces the carbonation depth. In S-NHL, the carbonation of CH occurs more readily than that of pozzolanic reaction products like C-S-H, and accelerated carbonation (AC) facilitates the swift conversion of a substantial amount of CH into calcite. During the AC process, NHL only contains calcite type calcium carbonate, while after AC 28d, S-NHL generates a small amount of aragonite and vaterite due to the carbonation of C-S-H, C_3AH_{10}, etc; The total pore volume and porosity of S-NHL continue to decrease, but its average and most probable pore sizes first decrease and then increase. AC significantly reduces large pores in S-NHL, resulting in predominantly capillary pores (50~1000 nm) in hardened pastes. AC facilitates the development of compressive strength in S-NHL pastes, with strength gains positively correlating with CH content.

Keywords

Natural hydraulic lime; ground granulated blast furnace slag; accelerated carbonation; microstructure; mechanical properties

CARBONATION DEGREE OF C-S-H IN WET-DRY CYCLE

Oh, D.[1], **Zhang, J.**[1], **Zheng, T.**[1] **and Kitagaki, R.**[1]

[1] Faculty of Engineering, Hokkaido University
Email: ekdud333@eng.hokudai.ac.jp

Abstract

As part of the global warming countermeasures, many studies are being conducted to absorb CO_2 using cement hydrates efficiently$^{1-2)}$. Calcium silicate hydrate (C-S-H), which is charged with the largest composition in waste concrete, can reabsorb CO_2. Therefore, it is necessary to improve the carbonation efficiency of C-S-H. The wet-dry cycle promotes the carbonation of hardened cement paste: the rearrangement of the C-S-H sheets due to drying creates macropores, which increase their continuity, and then, as the humidity increases, more Ca^{2+} ions are leached from the C-S-H sheets$^{1)}$. Previous studies have shown that the carbonation efficiency of synthetic C-S-H increased under the wet-dry cycle; however, the synthetic C-S-H used in the previous experiment had a small particle size and, therefore, a large surface area, so the effect of the wet-dry cycle on improving CO_2 absorption was limited$^{2)}$.

In this study, the effect of the wet-dry cycle on the carbonation of C-S-H sized of about 1mm was analyzed. The C-S-H with a Ca/Si ratio of 1.5 was prepared by mixing C_3S with the stoichiometric amount of nano silica. The water-to-binder ratio was 0.875, and the superplasticizer added 3 wt% of the binder. C_3S is manufactured in a laboratory, and the raw materials and calcination temperature program are shown in Tables 1 and 2, respectively. The humidity conditions for carbonation were divided into two types: wet-dry cycle (60-80% RH and 50-90% RH) and constant relative humidity (40% RH, 70% RH, 100% RH). Tables 3 and 4 list the mixing proportion for C-S-H and the experiment condition, respectively. To quantify the CO_2 absorption rate in each sample, thermogravimetric analysis (TGA) was conducted. The measurement was performed under an N_2 atmosphere at 20 ℃ to 950 ℃ with a heating rate of 10 ℃/min, and the samples were maintained at 105 ℃ for 30 min for drying.

Table 1. Raw Materials of C_3S (per 100g of C_3S)

$CaCO_3$	SiO_2	Al_2O_3	Fe_2O_3
131.50 g	26.32 g	1.5 g	1.5 g

Table 2. Calcination Temperature Program

Temperature	30-1600 (℃)	1600 (℃)	1600-1300 (℃)
Time	90 min	180 min	10 min

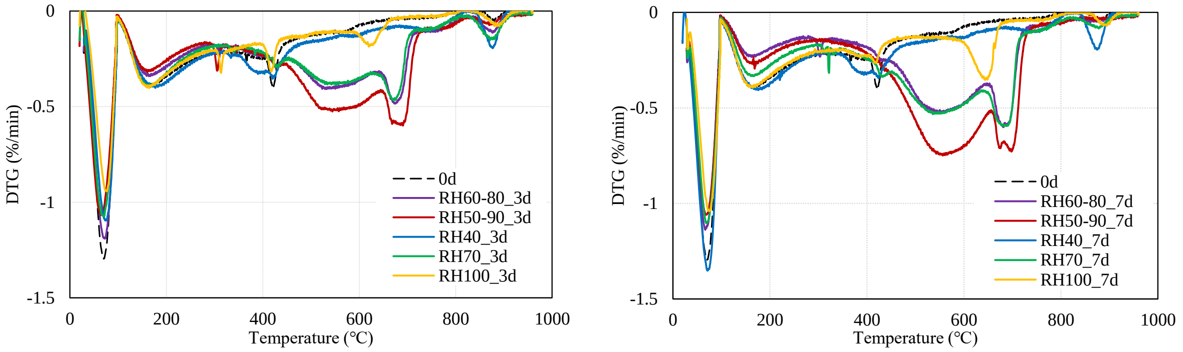
Table 3. Mixing Proportion for C-S-H

Materials	C_3S	Nano silica	Water	Superplasticizer
Weight	158.34 g	41.67 g	175.0 g	6.0 g

Table 4. Experiment Conditions

C/S	C-S-H particle size	Temperature	CO_2 concentration
1.5	0.6-1.18 mm	20 ℃	400-500 ppm
Relative humidity			Carbonation periods
60-80 %RH, 50-90 %RH, 40 %RH, 70 %RH, 100 %RH			3, 7 days

Figure 1 shows the TGA results. The graph of uncarbonated C-S-H (0d) shows that a small amount of $Ca(OH)_2$ was contained, but the amount can be negligible. The weight loss from 100 to 400°C, owing to the dehydration of C-S-H, decreased in the carbonated samples under the wet-dry cycle compared to that of under constant RH, and the difference of this was larger over time. This indicates that the carbonation of C-S-H was promoted by the wet-dry cycle. In the temperature range from 350 to 450°C, the broad peak, considered amorphous calcium carbonate (ACC), was observed at RH 40 after 3 and 14 days of carbonation. This is because the polymorphic transformation from the metastable to the stable state is suppressed at low RH due to the lack of water in the pores3). Moreover, for wet-dry cycle samples (RH60-80, RH50-90) and RH 70, a broad peak around 450 to 650°C was observed due to decarbonation of metastable calcium carbonates such as vaterite and aragonite. Except for RH40, calcite peak was observed around 600 to 750°C. The CO_2 absorption rate is shown in Table 5, and it was calculated from weight loss between 450 and 800°C of TGA results. The CO_2 absorption rate of RH70 is almost the same as that of RH60-80. However, it can be seen that the CO_2 absorption rate of RH50-90 was 30% higher than that of RH70. From the result, minimum and maximum RH also affect the promotion of C-S-H carbonation.



(a) After 3 days of carbonation (b) After 7 days of carbonation

Figure 1. TGA results

Table 5. CO_2 absorption rate (%)

	RH60-80	RH50-90	RH40	RH70	RH100
3 days	10.54	13.51	4.23	9.89	3.08
7 days	12.72	17.70	4.29	13.41	4.51

Keywords
C-S-H; carbonation; relative humidity; wet-dry cycle; thermogravimetric analysis

Acknowledgement
This study was partly sponsored by Japan Society for the Promotion of Science KAKENHI Grant Number 24K17386.

References

1) D. Oh, R. Kitagaki, T. Masuo, Z. Li, R. Kurihara, T. Noguchi, I. Maruyama, Natural carbonation boost for hardened cement fines by dripping technique, Cement and Concrete Composites, 153 (2024), 105731.
2) T. Zheng, J. Zhang, D. Oh, R. Kitagaki, The initial carbonation behavior of synthetic C-S-H by relative humidity, International Conference on Concrete Sustainability (ICCS 2024), Guimarães, Portugal, 09/2024.
3) E. Drouet, S. Poyet, P. Bescop, J.M. Torrenti, X. Bourbon, Carbonation of hardened cement pastes: Influence of temperature. Cement and Concrete Research, 115 (2019), 445–459.

STUDY ON THE HEAT CONDITIONS FOR APPROPRIATE CO_2 QUANTIFICATION METHOD IN CEMENTITIOUS MATERIALS

Takahashi, H.[1], Miyamoto, S. [1], Tabara, K.[2], Kameda, H.[2], Yamashita, K.[2], Minagawa, H. [1] and Hisada, M.[1]

[1]Tohoku University
[2]Denka Co., Ltd.
Email: hayato.takahashi.a2@tohoku.ac.jp

Abstract

In recent years, “carbon capture and utilization technology” is the biggest concern all over the world towards carbon neutrality [e.g., 1]. To accurately determine the contribution to carbon neutrality in concrete industry, it is important to establish a uniform method for evaluating the carbon dioxide capture of concrete. TG-DTA is a simple and representative method for measuring the carbon dioxide capture of concrete, which measures the increase or decrease in mass due to the reaction of compounds caused by increasing temperature. Therefore, if only the decarbonation of calcium carbonate can be properly extracted, the amount of carbon dioxide capture by TG-DTA can be evaluated very accurately. However, since several minerals coexist in concrete and thus it can be considered that the temperature range corresponding to thermal decomposition of these minerals might interfere with the temperature range of decarbonation. Therefore, it is difficult to accurately extract only the decarbonation. Based on the above background, in this study, major cement hydrates other than calcium carbonate in concrete were prepared, and the interference of temperature range of these hydrates’ decomposition with that of decarbonation was examined. Then, this study also proposed the alternative heating method to eliminate the interference.

In the experiment, ettringite (Ett), monosulfate (Ms), monocarbonate (Mc), and C-S-H were prepared as the main hydrates to observe the thermal decomposition. Ett was prepared by mixing the aluminate phases with calcium sulfate dihydrate at a molar ratio of 1:3 and decarbonation water and stirring the mixture for 10 days in a room at 20 ℃ using a magnetic stirrer. Ms was prepared by mixing the aluminate phase with calcium sulfate dihydrate at a molar ratio of 1:1 and decarbonation water and stirring the mixture for 12 days at 80 ℃. Mc was prepared by mixing the aluminate phase with calcium carbonate at a molar ratio of 1:1 and decarbonation water and stirring the mixture for 12 days at 20 ℃. C-S-H was prepared by mixing the alite with decarbonation water and stirring the mixture for 28 days at 20 ℃ (hereinafter denoted as hydrated alite). Note that this method also generated Calcium hydroxide (CH), because excess calcium ions precipitate as CH during the precipitation of C-S-H.

The prepared hydrates were heated using TG-DTA at a conventional temperature increase rate of 20 ℃/min, and their thermal decomposition behavior was observed. **Figure 1** shows the DTG curve of the prepared hydrates. For Ett, the strong DTG peak generally converged by 200 ℃, and all DTG peak converged at around 300 ℃. For Ms and Mc, the DTG peaks for Ms and Mc converged by 500 ℃ although several consecutive peaks were observed due to stepwise dehydration. As for hydrated alite, a large peak was observed between 400 ℃ and 500 ℃ and a small peak in the range of 550 ℃ to 700 ℃, which the former was attributed to the thermal decomposition of CH and the latter was C-S-H. It is known that calcium carbonation decarbonation occurs at 600 ℃ or higher. Therefore, there is a risk of that temperature range of

C-S-H decomposition interfere with the that of the calcium carbonate decarbonation whereas the interference by other phases is negligible.

C-S-H has gel pores and interlayer pores in nanoscale, and the water in these pores has high resistance to dehydration. To occur the dehydration in the pores at a lower temperature, decreasing the ramp rate can be considered as a possible method. Based on this assumption, attempt to increase the temperature at a slower rate was carried out to check the change of DTG peak position of C-S-H dehydration. **Figure 2** shows the DTG curves of the hydrated alite at a temperature increase rate of 5, 3 and 1℃/min, respectively. As a result, the DTG peak of C-S-H is clearly shifted to lower temperatures compared to the case with a temperature increase rate of 20 ℃/min shown in **Figure 1**. In case of 1℃/min, the DTG peak of C-S-H was shifted to between 500 ℃ and 600 ℃, indicating the convergence at lower temperatures than 600 ℃, where decarbonation reaction takes place. Therefore, it can be concluded that the temperature increase rate of TG-DTA to 1 ℃/min can eliminate the interference of the mass loss due to the thermal decomposition of C-S-H on the mass loss of decarbonation.

Keywords
TG-DTA; calcium carbonate; calcium silicate hydrate; thermal decomposition

Acknowledgement
This research is based on results of "Development of Materials, Manufacturing Methods and Quality Control System on Innovative Carbon Negative Concrete", JPNP21014, commissioned by the New Energy and Industrial Technology Development Organization (NEDO).

Reference

1) CUCO HP: https://www.cuco-2030.jp/english/ (July 23, 2024)

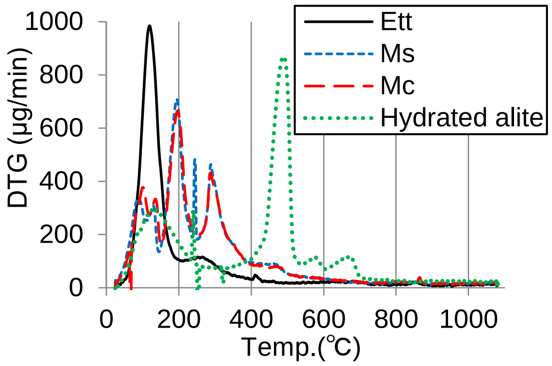


Figure 1. DTG curves for cement hydrates

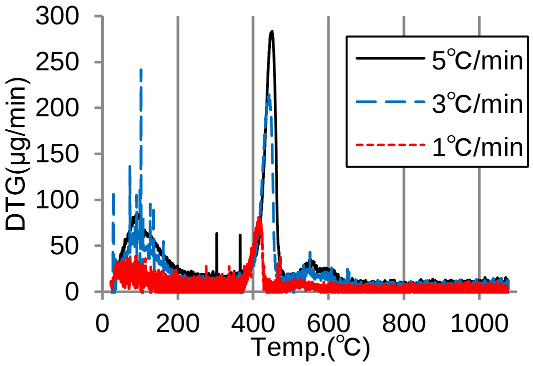


Figure 2. DTG curves for hydrated alite at each temperature increase rate

IMPACT OF Ca/Si AND Al/Si RATIOS ON THE ALUMINA-SILICA GEL FORMED BY WET CARBONATION OF SYNTHESIZED C-S-H PHASES AND ETTRINGITE

Song, J.[1], Zajac, M.[2], Ben Haha, M.[2], and Skibsted, J.[1]

[1] Department of Chemistry and Interdisciplinary Nanoscience Center (iNANO), Aarhus University, Denmark
[2] HeidelbergMaterials, Global R&D, Germany
Email: jiayi_song@chem.au.dk

Abstract

Recent studies have shown that enforced carbonation of recycled concrete fines from end-of-life concrete can bind substantial amounts of CO_2, giving a product with very favorable properties as a supplementary cementitious material in new composite cements [1]. The carbonation products are $CaCO_3$ and an amorphous alumina-silica gel with an alumino-silicate structure that contains different types of Q^3(nAl) and Q^4(mAl) SiO_4 silicate species and aluminum in four-fold coordination as Al(-O-Si)$_4$ sites.

To improve our knowledge on the structure and composition of the amorphous alumina-silica gel, this work investigates four model precursors of synthesized C-S-H in blends with portlandite and/or ettringite (Table 1). The C-S-H phase was prepared by hydration of α-$Ca_2SiO_3(OH)_2$ for 28 days using a water/binder ratio of 0.40. The samples were carbonated in an aqueous solution at room temperature (20 ± 1 °C), using a mixed gas of 25 vol% CO_2 and 75 vol% N_2. The carbonation experiments were interrupted from a few seconds to a maximum of six hours. After drying in a vacuum desiccator, the structure and composition of the carbonated samples were examined by ^{29}Si and ^{27}Al MAS NMR as well as TGA analysis.

Table 1. Bulk compositions (wt%) of the composite C-S-H samples studied in this work.

	CSH	CSH-Ca	CSH-Al-Ca	CSH-Al
Synth. C-S-H	100	80	60	80
Portlandite	-	20	20	-
Ettringite	-	-	20	20

The impact of additional calcium on the carbonation process and kinetics is investigated by the CSH and CSH-Ca samples, which exhibit bulk Ca/Si ratios of 1.39 and 1.98, respectively. Their ^{29}Si NMR spectra show two nearly identical Q^3 and Q^4 peaks after 6 hours of carbonation, reflecting the formation of a pure silica gel. The CSH-Al and CSH-Al-Ca samples have been prepared to study the impact of different Al/Si ratios on the C-S-H carbonation, with bulk Al/Si ratios of 0.30 and 0.22, respectively. The ^{27}Al NMR spectra show a resonance at 56 ppm originating from Al(-O-Si)$_4$ sites incorporated in an alumina-silica gel of similar composition and structure as found in carbonated Portland cements [1]. Additionally, two resonances at ~9.6 ppm and ~6.1 ppm are observed in the region for octahedral Al, which originate from a pure alumina gel with an approximate composition of $Al_2O_3{\cdot}7H_2O$. The corresponding ^{29}Si NMR spectra show resonances from the alumina-silica gel in the Q^3 – Q^4 region, which are broader and significantly different from the distinct Q^3 and Q^4 peaks observed for the pure silica gel

(*i.e.*, samples CSH and CSH-Ca), thereby showing the incorporation of aluminum in the silicate network.

The results from simulations of the ^{29}Si NMR spectra of the four model precursors are summarized in Figure 1. The data shows a slightly delayed carbonation and also a slower carbonation rate of the C-S-H phase in the CSH-Ca sample for the C-S-H blended with portlandite, as compared to the pure C-S-H. This observation is also found by comparing the CSH-Al and CSH-Al-Ca samples and suggests that portlandite carbonates independently and faster than the C-S-H phase. This is in agreement with similar findings for more complex systems such as wet carbonation of pure Portland cement [2]. It is also observed that the decomposition of the C-S-H phase, forming the (alumina-) silica gel, begins slightly earlier for the CSH-Al and CSH-Al-Ca samples, as compared to CSH and CSH-Ca, indicating that the presence of aluminum may promote the carbonation process.

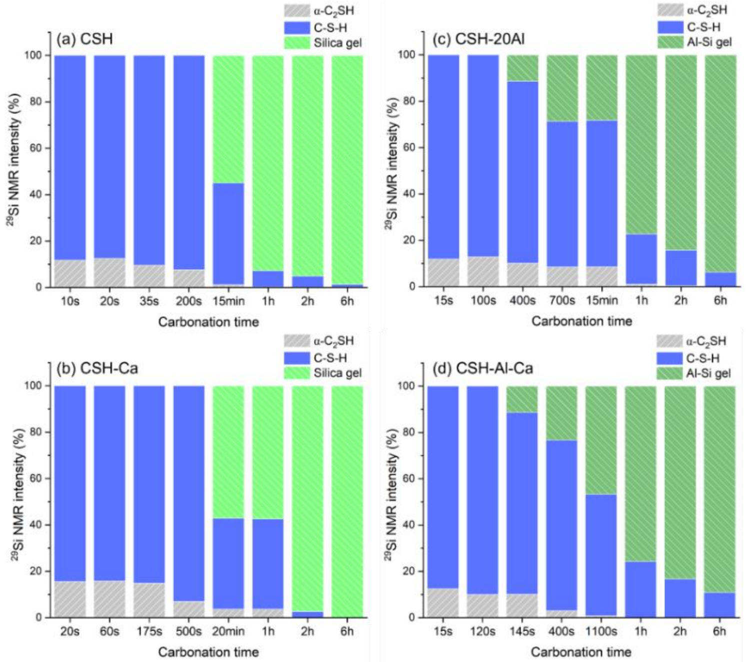


Figure 1. Relative fractions of ^{29}Si in α-C_2SH, the C-S-H phase, and the (alumina-) silica gel in the (a) CSH, (b) CSH-Ca, (c) CSH-Al, and (d) CSH-Al-Ca samples as a function of carbonation time. The intensities are obtained from simulations of the ^{29}Si NMR spectra.

In conclusion, both additional Ca and Al (provided by portlandite and ettringite) have an impact on the carbonation rate for the C-S-H phase. An alumina gel with Al in octahedral coordination is observed as a product, in addition to the alumina-silica gel, for the carbonated C-S-H – ettringite samples.

Keywords

Carbonation; C-S-H; ettringite; alumina-silica gel; NMR spectroscopy

References

[1] M. Zajac, J. Song, P. Ullrich, J. Skocek, M. Ben Haha, J. Skibsted, High early pozzolanic reactivity of alumina-silica gel: A study of the hydration of composite cements with carbonated recycled concrete paste, *Cem. Concr. Res.* 175 (2024) 107345.

[2] M. Zajac, J. Skibsted, P. Durdzinski, F. Bullerjahn, J. Skocek, M. Ben Haha, Kinetics of enforced carbonation of cement paste, *Cem. Concr. Res.* 131 (2020) 106013.

STUDY ON THE HARDENING MECHANISM OF NATURAL HYDRAULIC LIME (NHL) UNDER HYDRATION AND CARBONATION

Guodong Qi[1], **Dongmin Wang**[1], **and Dong Xu**[1]

[1] China University of Mining and Technology (Beijing), Beijing, China
Email: wangdongmin@cumtb.edu.cn

Abstract

Natural hydraulic lime (NHL) possesses both air-hardening components ($Ca(OH)_2$) and hydraulic components (C_2S). Its hardening process is influenced by the hydration of C_2S and the carbonation of $Ca(OH)_2$. This study aims to elucidate the phase composition and microstructural evolution mechanisms during the hardening process of NHL, as well as to clarify the interactions between hydration and carbonation at different hardening stages. The results indicate that in the early stages of hardening (approximately before 28 days), hydration reactions dominate, while carbonation reactions are nearly absent.

The primary hydration products of NHL are calcium silicate hydrate (C-S-H) and calcium hydroxide ($Ca(OH)_2$). As shown in Figure 1, the hydration heat release curve of NHL is similar to that of Portland cement and can be divided into four stages: pre-induction, induction, acceleration, and deceleration. Upon mixing NHL with water, alkaline substances such as $Ca(OH)_2$ and C_2S dissolve rapidly, releasing a large amount of heat, resulting in an intense exothermic peak during the initial hydration phase. As hydration progresses, $Ca(OH)_2$ continues to dissolve, releasing a large quantity of Ca^{2+} and OH^- into the pore solution, creating a highly alkaline environment that promotes the hydration of C_2S. A second exothermic peak, representing the formation of C-S-H, appears between 3 to 6 hours. The rate of heat release during NHL hydration decreases as the water-to-binder ratio increases, particularly in the phase where the second exothermic peak occurs. The peak heat release for NHL0.5 exceeds 0.4 mW/g, indicating that a lower water-to-binder ratio facilitates the hydration of C_2S and the formation of C-S-H.

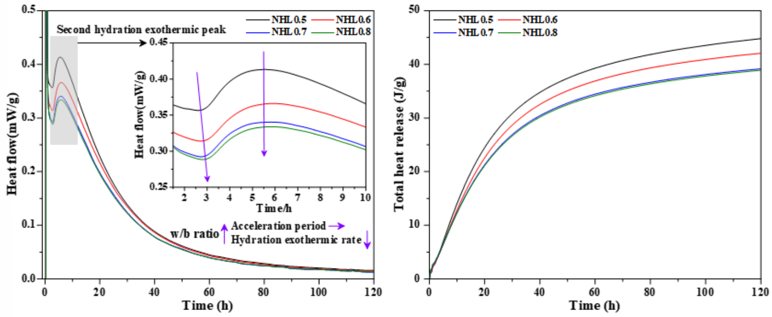


Figure 1. Variation of hydration heat flow and total heat release in NHL pastes with different w/b ratios.

Before the end of the induction period (before 2.6 h), the XRD patterns of NHL0.5 show distinct characteristic diffraction peaks for C_2S, CH, and $CaCO_3$. During the hydration acceleration phase (2.6 h–6 h), sharp diffraction peaks of C_2S can still be detected, although they slightly weaken as hydration progresses. Correspondingly, there is no significant increase in the characteristic diffraction peaks of CH, nor are C-S-H peaks observed, which can be attributed to the relatively low hydration reactivity of C_2S. As hydration continues, the characteristic diffraction peaks of C_2S significantly diminish, while the CH peaks at 18.1° and 34.1° increase notably. Additionally, a pronounced C-S-H characteristic diffraction peak (broad hump) is detected at 32°, indicating that, with the ongoing hydration process, C_2S gradually forms CH and C-S-H. Before 7 days of hydration, the diffraction peaks of $CaCO_3$ remain unchanged, suggesting that carbonation reactions do not occur significantly at this stage. This is because the high pore water content in NHL during this period suppresses CO_2 diffusion inward, thereby limiting carbonation reactions.

For NHL paste with a water-to-binder ratio of 0.5, the primary phases during the hardening process consist of dicalcium silicate (C_2S), calcium hydroxide (CH), calcium carbonate ($CaCO_3$), and calcium silicate hydrate (C-S-H), as shown in Figure 2. In different depth regions, distinct changes in the diffraction peaks of CH (2θ at 18° and 34.1°) and $CaCO_3$ (2θ at 29.5°) in the inner region only become noticeable after 180 days. During this period, CH predominantly undergoes carbonation, transforming into $CaCO_3$. As indicated in Figure 9(a), the diffraction peak intensity of C_2S gradually decreases with curing time, but C_2S peaks remain detectable even at 1 year, indicating that the hydration of C_2S in NHL is a long-term and slow process. The phase evolution in the middle region of the NHL0.5 paste is generally consistent with that of the inner region, whereas the outer region exhibits significantly different phase changes. In the outer region, the diffraction peaks of CH and $CaCO_3$ show notable weakening and enhancement, respectively, after 14 days. In the inner region, there are minimal changes in phase composition and diffraction peak intensity before 14 days of curing, although partial C-S-H formation is visible in Figure 2. As the curing time increases, the CH diffraction peak intensity gradually diminishes, while the $CaCO_3$ peak significantly intensifies. Based on Figure 9, the C_2S diffraction peak intensity continues to weaken with curing time, with only trace amounts of C_2S remaining at 180 days. Furthermore, noticeable differences in the diffraction peak intensity of C_2S are observed across different depth regions and water-to-binder ratios, highlighting the importance of monitoring the impact of carbonation on C_2S over extended curing periods. Additionally, the diffraction peak at 33° indicates that the $CaCO_3$ is present in the vaterite crystalline form, a phenomenon that also appears in the early-age phase composition. Early-age carbonation in the inner region may be attributed to the CO_2 absorbed during the mixing and molding process.

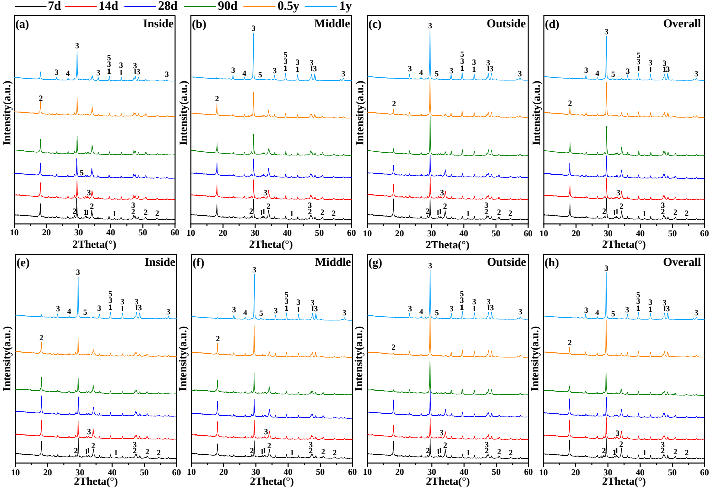


Figure 2. XRD patterns of NHL at different curing ages and zones, NHL0.5~(a-d) and NHL0.6~(e-f). Notation: 1—C_2S; 2—$Ca(OH)_2$; 3—$CaCO_3$; 4—SiO_2; 5—C-S-H.

Keywords
Natural hydraulic lime; hydration; carbonation; hardening mechanism

INSIGHTS INTO THE SIMULTANEOUS FORMATION AND CARBONATION OF C-S-H: THE EFFECT OF INITIAL pH

Yuxi Cai[1], Pan Feng[1,2], and Xin Liu[1]

[1]Jiangsu Key Laboratory of Construction Materials, School of Materials Science and
Engineering, Southeast University, Nanjing 211189, China
[2]State Key Laboratory of High Performance Civil Engineering Materials, Nanjing 210008,
China
Email: pan.feng@seu.edu.cn

Abstract

With the purpose of lowering carbon emissions and controlling high performance, modern concrete evolves with the utilization of substituting clinker with suitable supplementary cementitious materials (SCMs), leading to the reduction in pH of pore solutions and great alterations in the cement chemistry, ultimately impacting the long-term durability, especially the decreased carbonation resistance. The issue is believed to associate closely with the primary hydration product of cement, C-S-H, which assumes a greater role in dominating the performance of concrete.

The initial pH exerts tremendous influence on both the formation and carbonation of C-S-H. This study focuses on the induced dissolution behavior and nano/microstructural alterations of C-S-H under coupled formation and carbonation conditions at varying initial pH levels with the intention to simulate different carbonation scenarios that concrete could be subjected to.

The initial pH has a significant impact on the pH evolution and silicate concentrations of solutions during the simultaneous formation and carbonation of C-S-H, as shown in Figure 1. All samples experience a considerable decrease in pH during carbonation with four typical periods, unchanged period (I), rapid decline period (II), slow ongoing period (III) and steady period (IV). In the period I, large quantities of hydroxide ions are consumed with only a small pH reduction of about 0.5, which is relatively steady compared to the entire process. Afterwards, there exists an abrupt pH decline followed by a more moderate decrease before reaching the stability at a low value. At a low initial pH of 11.65, the initial period tends to be obscured and integrated into the rapid decline phase with an equivalent pH decrease rate, due to the quite low concentrations of hydroxide ions that are easier to be consumed by carbonation.

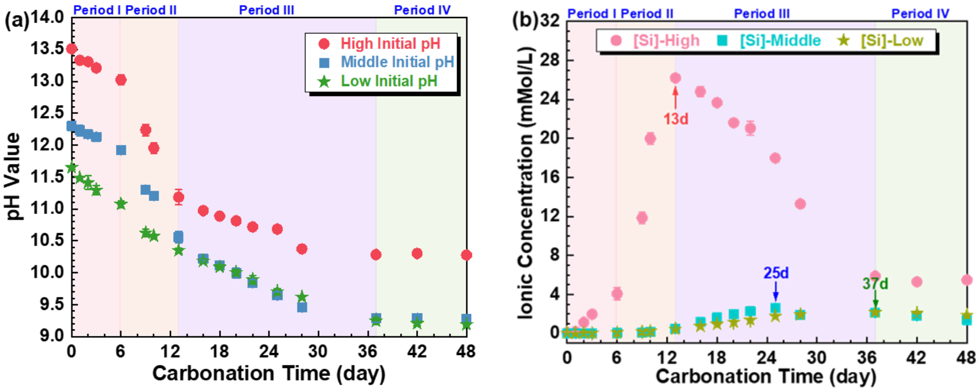


Figure 1. (a) The pH evolution and (b) [Si] of C-S-H suspensions with different initial pH subject to carbonation.

As depicted in Figure 1(b), except the disparities in concentrations, the behavior of calcium and silicon in all solutions exhibits a similar trend, in line with the pH changes. The dissolution of silicate ions is initiated once residual calcium ions with the potential to be carbonated are consumed up, followed by inhibition due to the low pH and low Ca/Si ratio of C-S-H, leading to precipitation of silicate ions. At an initial pH of 13.51, all interlayer calcium and residual calcium ions in the liquid phase are depleted during the unchanged period, with an integrity of silica chains. A decrease in initial pH results in a longer duration for the consumption of interlayer calcium and residual calcium ions, spanning stages I and II, indicating a slower carbonation process.

The weak ingress of carbonation has little influence on the massive formation of C-S-H at initial stages, the composition of which is decided by the decreased pH with lower Ca/Si ratio. During carbonation, the C-S-H is decomposed, with the formation of calcium carbonate and amorphous silica gel. At decreases initial pH, a slower carbonation proceeds as the decomposition of C-S-H was delayed, while the decomposition shows a similarity. The carbonation concentrates on the interlayer calcium of C-S-H and the residual calcium ions in solution when Ca/Si ratio is higher than 0.72. Then the carbonation transfers to the main layer of C-S-H with the dissolution of silicate ions. At Ca/Si decreased to below 0.58, silica gel is formed, and the Ca/Si ratio of C-S-H phase is maintained at 0.58 in further carbonation.

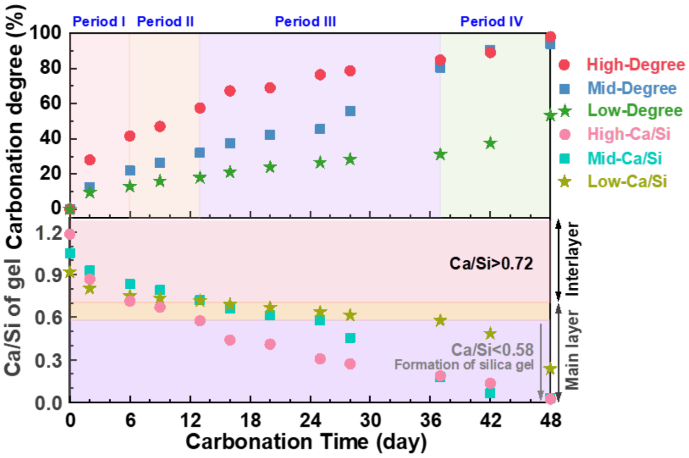


Figure 2. The evolution of carbonation degree and Ca/Si ratio of the solid phase as a function of carbonation time. High: initial pH=13.45, Mid: initial pH=12.30, Low: initial pH=11.65.

The carbonation reduces the hardness of C-S-H pellets, especially at the initial stages, which is a bit compensated by the denser structure formed in interlayer decomposition. The decreased initial pH has a smaller effect on the decline of hardness, which may relate greatly to the changed C-S-H structure.

Keywords

C-S-H; formation; carbonation; alkalinity; decomposition

Acknowledgments

This work was financially supported by Innovandi Core Project 15, the National Natural Science Foundation of China (nos. 52122802 and 52078126), the Jiangsu Provincial Department of Science and Technology Innovation Support Program (nos. BK20222004 and BZ2022036).

COMPARISON OF THE C_3S, C_2S, AND CEMENT DISSOLUTIONS IN WATER AND CO_2 SOLUTIONS

Tang, Wei[1,2], **and Zhan, Baojian**[1,2]

[1]College of Civil and Transportation Engineering, Shenzhen University, Shenzhen, Guangdong, China
[2]Guangdong Provincial Key Laboratory of Durability for Marine Civil Engineering, Shenzhen University, Shenzhen, Guangdong, China
Email: zhanbj@szu.edu.cn

Abstract

The hydration of ordinary Portland cement is accelerated by the CO_2 curing through the dissolution-precipitation process. To explore the mechanism of CO_2 activated cement, we determined the dissolution rates of C_3S, C_2S and cement particles, respectively, over a wide range of calcium and silicates concentrations through the Inductively Coupled Plasma (ICP) analysis. The experimental solubility products (K_{sp}) of C_3S, C_2S, and cement particles were estimated and taken a comparison in the water and CO_2 solutions. Results showed that, compared to cement reacting with water, the free enthalpy ($\Delta_r G$) during the hydrolysis of C_3S, C_2S, and cement are higher in the CO_2 solutions compared with that in the pure water, and this suggested that the CO_2 accelerates the dissolution rate of these minerals. Furthermore, the solid contents in these two systems were tested by the Quantitative X-ray Diffraction (QXRD), Thermogravimetric Analysis (TGA), and Fourier Transform Infrared Spectrometer (FT-IR), which suggested that the conversion rate of the mineral phases in CO_2 solution is up to 17 times higher than that in water in the first 2 hours. Based on the experimental results, A Thermodynamic model was built to give a clear description for the mineral changes of the solid contents during the early hydration of cement in the CO_2 solutions.

Keywords

CO_2 solution; dissolution rate; cement; thermodynamic

COLD SINTERING OF $CaCO_3$ POLYMORPHS: EXPLORING THE STRENGTH ORIGIN IN CALCIUM CARBONATE MATRIX

Jinzewei Nie[1], Wei Wang[1], Yamei Zhang[1,2]

[1]School of Materials Science and Engineering, Jiangsu Key Laboratory of Construction Materials, Southeast University, Nanjing 211189, China
[2]Nanjing Institute for Intelligent Additive Manufacturing Co., Ltd, Nanjing 210000, China
Email: 220222350@seu.edu.cn; wang-wei@seu.edu.cn; ymzhang@seu.edu.cn

Abstract

Carbonation of alkaline solid wastes can significantly lower the CO_2 footprint, and the generating calcium carbonate can refine the pore structure to improve the mechanical properties of concrete. However, the role of different calcium carbonate polymorphs (amorphous calcium carbonate, vaterite, aragonite and calcite) on the densification process of calcium carbonate particles remains unclear. In this study, a novel approach to endow strength integrity to calcium carbonate polymorphs powders via cold sintering process to explore the cementation of different calcium carbonate polymorphs, as well as the effects of applied pressure, temperature and solvent on the strength within calcium carbonate matrix. The results show that the capability for particle rearrangement, dissolution-reprecipitation characteristics, and crystal growth morphology of calcium carbonate polymorphs exert significant influence on the strength of the calcium carbonate matrix. This work largely contributes to the understanding of the strength origin within calcium carbonate matrix and can provide a potential guidance for the development of high strength calcium carbonate binder.

Keywords

Calcium carbonate polymorphs; cold sintering; cementation; mechanical properties

Session A3

CO_2 Activation of Recycled Concrete Fines

REACTIVITY OF AQUEOUS CARBONATED BLENDED PORTLAND CEMENT PASTES

Maia Neto, F.[1], Snellings, R.[2], and Skibsted, J.[1]

[1]Aarhus University, Department of Chemistry and Interdisciplinary Nanoscience Center (iNANO), Aarhus, Denmark
[2]KU Leuven, Earth and Environmental Sciences, Materials, Leuven, Belgium
E-mail: jskib@chem.au.dk

Abstract

Aqueous carbonation of end-of-life concrete fines is a promising method to alleviate greenhouse gas emissions by CO_2 sequestration from point-source emitters, which attracts increasing research attention [1]. Almost all calcium-bearing phases of the recycled cement paste can be carbonated, producing calcium carbonate and an amorphous alumina-silica gel as the principal phases. This also holds for pastes of Portland cement incorporating supplementary cementitious materials (SCMs), where the phase assemblages of the carbonated pastes reflect the original chemical composition of the Portland cement – SCM paste [2]. The product of carbonation exhibits high pozzolanic reactivity, which has been ascribed to the very high specific surface area of the alumina-silica gel and results in a comparatively fast reaction in the early hydration stages, reaching reaction degrees above 50 % in one day [3]. This study investigates effects of the composition of cement pastes and the carbonation conditions on the reactivity and phase assemblage for four aqueous carbonated blended Portland cement pastes.

The samples comprise a neat white Portland cement paste (wPc) and blended pastes of the white Portland cement with three conventional SCMs, *i.e.*, 10MS, 30FA, and 40S8, corresponding to samples with 10 wt.% replacement by microsilica, 30 wt.% by coal combustion fly ash, and 40 wt.% by ground granulated blast furnace slag, respectively. The carbonated cement pastes were produced in an aqueous carbonation setup at ambient temperature, using a glass beaker containing 30 mL solution of 0.1 M NaOH for 1.000 g of cement paste. A gas flow of 10 L/h (10 vol.% CO_2 and 90 vol.% N_2) was continuously released in the solution, which initially led to a constant pH of ~7.6 after equilibration for approx. 10 min.

The estimated phase assemblage is illustrated in Figure 1, which shows that the proportion of the alumina-silica gel varies between 29 and 35 wt% of the phase assemblage, depending on the chemical composition of the pastes. The Al/Si ratio and the number of non-bridging oxygens per SiO_4 tetrahedra (NBO/Si) for the alumina-silica gel depend on the amount of Al in the system [2]. The reactivity of the carbonated cement pastes is evaluated using a modified version of the R^3 test (*e.g.* a 1:1 ratio of carbonated cement paste and calcium hydroxide and a curing temperature of 25 ^oC). These tests reveal that the carbonated blended cement pastes exhibit higher reactivity than the carbonated pure Portland cement paste and it is mainly affected by the proportion and surface area of the alumina-silica gel. Characterization of the reaction products by ^{27}Al and ^{29}Si NMR shows that the pozzolanic reaction of Al-rich alumina-silica gel forms proportionally more C-(A)-S-H phase than Al-poor gels at 8 and 24 hours of reaction. Moreover, the Al/Si ratio of the formed C-(A)-S-H phase is on average 20 % lower than the one observed for the hydrated pastes before carbonation. This is caused by the reaction of calcium carbonates, which leads to the formation of hemi/monocarbonate AFm phases and ettringite. As a consequence, the Al concentration in the pore solution is decreased which results in a lower Al/Si ratio of the C-(A)-S-H phase.

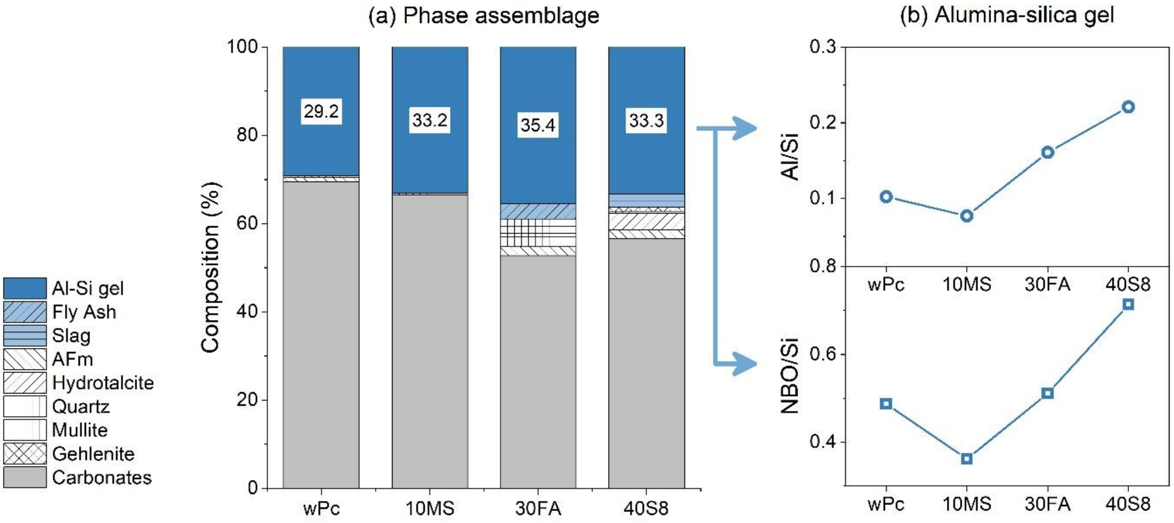


Figure 1. (a) Phase assemblage estimation of carbonated cement pastes. Well crystalline phases were quantified by XRD-Rietveld refinement whereas AFm and glassy SCM phases were estimated by ^{27}Al and ^{29}Si MAS NMR combined with elemental analysis. Carbonates include calcite, vaterite and aragonite. (b) Al/Si and NBO/Si ratios of the alumina-silica gel.

The aqueous carbonated pastes show rapid reactivity, irrespective of the chemical composition of the original paste and carbonation conditions. This is associated with the presence of an alumina-silica gel with a very high surface area (140 – 190 m^2/g), which enhances the dissolution of the alumino-silicate framework and allows full reaction in 2 - 3 days. The very fast reactivity of the alumina-silica gel under reactivity test conditions indicates that the early availability of Ca^{2+} ions is the main limitation for the degree of reaction when carbonated pastes are incorporated in composite cements.

The carbonation conditions (*i.e.*, CO_2 gas concentration, temperature, and solution composition) slightly affect the reactivity of the carbonated cement pastes (cCP's). Pastes carbonated in deionized H_2O and a 0.05 M Na_2SO_4 solution show the highest reactivity as measured by isothermal calorimetry following the R^3 test protocol (118 and 116 J/(g of cCP), respectively). The sample carbonated at 70 °C exhibits the lowest reactivity (96 J/(g of cCP)), which may reflect the very high early reactivity, which is not accounted for in calorimetry experiments.

Keywords
Alumina-silica gel; R^3 test; recycled concrete; SCM - supplementary cementitious materials; solid-state NMR

References

[1] M. Zajac, I. Maruyama, A. Iizuka, J. Skibsted, Enforced carbonation of cementitious materials, *Cem. Concr. Res*. 173 (2023) 107285.
[2] F. Maia Neto, R. Snellings, J. Skibsted, Aqueous carbonation of aged blended Portland cement pastes: Impact of the Al/Si ratio on the structure of the alumina-silica gel, *Cem. Concr. Res*. 177 (2024) 107428.
[3] M. Zajac, J. Song, P. Ullrich, J. Skocek, M. Ben Haha, J. Skibsted, High early pozzolanic reactivity of alumina-silica gel: A study of the hydration of composite cements with carbonated recycled concrete paste, *Cem. Concr. Res*. 175 (2024) 107345.

RE-CEM: APPLICABILITY OF RECYCLED CONCRETE PASTE AS SCM THROUGH CCU – ELUCIDATING THE CORRELATION BETWEEN CHEMICAL COMPOSITION, AMORPHOUS GEL STRUCTURE, AND POZZOLANIC REACTIVITY

Niewöhner, F.[1], **Hilbig, H.**[1], **and Machner, A.**[1]

[1] Technical University of Munich, TUM School of Engineering and Design, Department of Materials Engineering, Professorship of Mineral Construction Materials, Franz-Langinger-Str. 10, 81245 Munich, Germany
Email: fabian.niewoehner@tum.de

Abstract

The need of today's society to reduce emission of greenhouse gasses and resource consumption is currently prevailing in discussions and media more than ever. The construction sector is a massive contributor to global CO_2 emissions, with cement production alone accounting for approximately 7-8% of total anthropogenic CO_2 emissions. The re-CEM project addresses this critical challenge by exploring the potential of carbon capture and utilization (CCU) technologies to convert recycled concrete paste (RCP) into a viable supplementary cementitious material (SCM). This research aims to clarify the complex relationships between RCP's chemical composition, the amorphous Si-Al-gel structure formed during carbonation, and the pozzolanic reactivity of the resulting carbonated RCP (cRCP).

Cement production traditionally relies on high-energy processes and natural resources, leading to substantial CO_2 emissions. The integration of SCMs into cement can significantly reduce these emissions. However, conventional SCMs like fly ash and slag are becoming scarce, necessitating the development of alternative materials. RCP, a by-product of concrete recycling, presents a promising solution due to its high availability and potential for CO_2 sequestration when subjected to carbonation. Despite this potential, RCP is underutilized, with much of it ending up in landfills. The wet carbonation of RCP can enhance its reactivity by forming a reactive Si-Al-gel, transforming it into a highly pozzolanic SCM.

The re-CEM project is organized into four interrelated work packages (WPs), each targeting key aspects of this transformation process:

> **WP1: Preparation and characterization of lab-scale RCP and cRCP** This work package focuses on preparing well-hydrated cement pastes that simulate the composition of RCP. These pastes will undergo wet carbonation, a process where gaseous CO_2 is introduced into an alkaline solution containing the RCP, leading to the formation of carbonates and a Si-Al-gel. The resulting cRCP will be characterized by QXRD, TGA, FTIR, ^{29}Si and ^{27}Al NMR to determine its chemical and mineralogical composition, with particular emphasis on understanding how the initial composition of RCP affects the structure and properties of the carbonated material.
>
> **WP2: Reactivity testing of cRCP and its impact on cement hydration** WP2 is dedicated to assessing the pozzolanic reactivity of cRCP, which is crucial for its effectiveness as an SCM. This will be done using the R^3 test, a standardized method that

measures the heat of hydration and portlandite consumption to evaluate the reactivity of SCMs. In addition to reactivity tests, the impact of cRCP on the hydration process and microstructure of blended cements will be studied. This work package aims to identify the optimal replacement levels of ordinary Portland cement (OPC) with cRCP, which could range from 20% to as much as 50%.

WP3: Correlation of composition, structure, and reactivity of cRCP
The goal of WP3 is to establish a systematic understanding of how the composition and structure of the amorphous Si-Al-gel in cRCP correlate with its pozzolanic reactivity. By combining data from the characterization studies in WP1 and the reactivity assessments in WP2, this work package seeks to develop predictive models that can forecast the performance of cRCP based on its pre-carbonation composition. These models will be essential for determining the most suitable types of RCP for CCU applications and maximizing the efficiency of the carbonation process.

WP4: Feasibility and upscaling of cRCP application
WP4 explores the practical application and scalability of cRCP in the construction industry. This work package will test cRCP derived from real-world recycled concrete materials, evaluating its performance in mortar samples and comparing it to laboratory-prepared cRCP. The feasibility of integrating cRCP into industrial cement production processes will be assessed, with a focus on validating the laboratory findings from WP1-3.

The findings from the re-CEM project are expected to provide a scientific basis for the broader application of CCU methods in the cement industry. By leveraging recycled materials like RCP, the project aims to contribute to developing sustainable construction practices that reduce CO_2 emissions, promote resource efficiency, and advance the circular economy.

Keywords
cRCP; CCU; SCM; pozzolanic reactivity; alumina-silica gel

EXPLORING THE USE OF CONCRETE WASTE FINES IN LIGHTWEIGHT POROUS CEMENT-BASED BLOCKS THROUGH CARBONATION

Sousa, L. N.[1], Koh, C. H.[1], Schollbach, K.[1], and Brouwers, H.J.H.[1]

[1]Department of the Built Environment, Eindhoven University of Technology, 5600MB,
Eindhoven, the Netherlands
Email: l.nobrega.sousa@tue.nl

Abstract

The reuse and recycling of concrete fine waste from construction and demolition wastes (CDW) is pertinent to minimize the negative impact of their environmental footprint, following the circular economy principle. The concrete waste is collected from demolition sites, crushed, and subsequently sieved to separate them into various aggregate fractions for different applications. One of the waste products generated during the crushing process is fine fraction aggregates, which consist of a high content of cement paste and silica-rich aggregates. They exhibit high water absorption capacity and low reactivity and are often detrimental to performance and durability when incorporated directly into new cement-based recipes

In this study, an experimental study was carried out to evaluate the feasibility of preparing lightweight porous and carbonated cement-based blocks using concrete fine wastes. The carbonated cement-based porous blocks were produced by using concrete waste fines. The production aims to optimize CO_2 uptake within the porous blocks to enhance the carbonation process. Carbonation involves the reaction of CO_2 with the hydration products of cement, particularly calcium hydroxide, to form calcium carbonate. This collaborates with the CO_2 sequestration mechanism, thereby reducing greenhouse gas emissions. The porous structure was produced by utilizing a foaming agent, creating air voids and pores that distributed throughout a matrix, making it versatile for various applications, such as insulating materials in building envelopes, wastewater treatment, and pavements.

Some parameters were investigated, such as the water content ratio, as it significantly influences carbonation, affecting the permeability of the mix, the diffusion of CO_2 through the porous structure, and the mechanical strength. Then, the feasibility of producing carbonated blocks was investigated by flexural and compressive strength, density, microstructural characteristics (SEM), thermogravimetric analysis, and thermal performance. The findings provide interesting insights into the potential of porous carbonated blocks as a sustainable route for CO_2 uptake and concrete waste management.

Keywords

Concrete waste fines; lightweight porous blocks; carbonation process; CO_2 sequestration

CARBONATION OF RCF WITH ADDITIONAL ADDITIVE: THE EFFECT ON CRYSTALLINTY OF CARBONATES

Y. Chen[1], Y. Luo[1], H.J.H. Brouwers[1]

[1]Department of the Built Environment, Eindhoven University of Technology, P.O. Box 513,
5600MB, Eindhoven, The Netherlands
Email: y.chen7@tue.nl

Abstract

The increasing demand for sustainable construction practices has driven interest in utilizing Recycled Concrete Fines (RCF) as a valuable resource. Using carbonation to reactivate the RCF is the more efficient method in current research. Additionally, carbonation technology not only reactivates RCF but also sequesters CO_2, reducing greenhouse gas emissions. This process fixes CO_2 in stable carbonates, thereby lowering atmospheric carbon levels. Consequently, it enhances the reactivity of RCF while mitigating CO_2 emission, creating a win-win for sustainable construction and environmental protection.

This study develops novel methods to improve RCF reactivity through wet carbonation processes. Sodium tripolyphosphate (STPP) is applied to tailor the carbonation process of RCF. To determine the effect of STPP on the carbonation of RCF and evaluate its effectiveness, various analytical techniques, including X-ray Diffraction (XRD), Thermogravimetric Analysis (TGA), and Fourier Transform Infrared Spectroscopy (FT-IR) are applied for phase analysis. Scanning Electron Microscopy (SEM) equipped with energy-dispersive spectroscopy (EDX) is used to for microstructure analysis.

STPP could form complexes with calcium ions, and thus prolong the transition from amorphous calcium carbonate to stable crystallized calcium carbonate [1,2]. It is believed that wet carbonation with STPP can affect the carbonation process of RCF. More specifically, the transition from amorphous calcium carbonate to crystalline calcium carbonate during carbonation could be hindered or delayed with STPP addition. It is anticipated that with an optimal concentration of STPP, the retention of amorphous calcium carbonate can further promote the performance of treated RCF as a supplementary cementitious material.

Keywords

Recycle concrete fines; wet carbonation; amorphous carbonates; sodium tripolyphosphate

References

[1] L. Cheng, Y. Chen, B. Yuan, and Q. Yu, “The underlying role of sodium tripolyphosphate on the cementitious mechanism of calcium carbonate binder,” *Compos. Part B Eng.*, vol. 247, p. 110362, Dec. 2022, doi: 10.1016/j.compositesb.2022.110362.

[2] L. Cheng, Y. Chen, T. Liu, H. J. H. Brouwers, and Q. Yu, “Understanding the CaCO3 phase transition of carbonated wollastonite composites caused by sodium tripolyphosphate: From amorphous to crystalline,” *Cem. Concr. Compos.*, vol. 148, p. 105477, Apr. 2024, doi: 10.1016/j.cemconcomp.2024.105477.

UNLOCKING THE CARBONATION PROCESS OF RECYCLED CONCRETE: INSIGHTS FROM INDUSTRIAL RECYCLED CONCRETE FINES

Y. Luo[1], P. Perumal[2,3], and H.J.H. Brouwers[1]

[1]Department of the Built Environment, Eindhoven University of Technology, P.O. Box 513, 5600MB, Eindhoven, The Netherlands
[2]C2CA Technology, 3013AK, Rotterdam, The Netherlands
[3]Faculty of Technology, Fiber, and Particle Engineering Research Unit, PO Box 4300, 90014, University of Oulu, Finland
Email: y.luo1@tue.nl

Abstract

Portland cement is one of the most widely used man-made building materials. However, its production consumes vast amounts of natural resources and energy. For the last decade, the carbonation of waste concrete has received keen attention. Firstly, carbonation treatment can effectively re-activate the recycled concrete fines (RCF), enabling its re-utilization in new concrete production. Moreover, the end-of-life concrete can serve as a carbon sink to sequestrate CO_2. Currently, most carbonation research focuses on laboratory-simulated RCF. However, the CO_2 mineralization process can differ for real RCF due to various factors such as age, weathering, and carbonation degree.

In light of the above, this study aims to experimentally clarify the carbonation process of industrial RCF, thereby reducing the knowledge gap between simulated and real RCF. In this work, industrial RCF was provided by C2CA Technology, The Netherlands. The RCF was dry carbonated in a climate chamber with varied relative humidity while maintaining a constant CO_2 concentration of 10% under ambient temperature. The phase composition, carbonation degree, and decalcification efficiency were investigated using XRD, TGA, and FTIR. Besides, the R3 test was carried out on carbonated RCF, following RILEM TC 267-TRM. Based on the above, the carbonation aspects were further correlated to the reaction kinetics of carbonated RCF when used as supplementary cementitious material (SCM).

The results showed that the main reacting phase for the investigated industrial RCF is C-S-H gel, and portlandite is largely retained during carbonation. The presence of residual portlandite is due to the portlandite crystals being covered by $CaCO_3$ during the weathering process, which prevents complete carbonation of the portlandite. It is interesting to note that in the given carbonation condition, calcite content remains unchanged after carbonation. At the same time, amorphous and poorly crystalline $CaCO_3$ including aragonite and vaterite, are observed. And the poorly crystalline $CaCO_3$ is increased as the carbonation duration increases from 15 min to 24 h. Moreover, relative humidity contributes to a higher carbonation degree of RCF. In comparison, with a carbonation duration of 360 minutes, the carbonation degree of industrial RCF increases from 15.2% to 38.3% as the relative humidity rises. Meanwhile, the environmental humidity promotes the thermal stability/crystallinity of the formed carbonates. As evidenced by the R3 test, the observed transition in crystallinity of carbonates further affects the reactivity of RCF when used as SCM.

Keywords
CO_2 sequestration; recycle concrete fine; supplementary cementitious materials; reactivity test

MINERAL CARBONATION OF RECYCLED CONCRETE PASTE

Namgyu Park[1], **Juhyuk Moon**[1,2]

[1]Department of Civil and Environmental Engineering, Seoul National University, Seoul 08826, Republic of Korea
[2]Institute of Construction and Environmental Engineering, Seoul National University, Seoul 08826, Republic of Korea
Email: parknamgyu@snu.ac.kr

Abstract

Portland cement is an effective material for constructing social infrastructure. It is the most widely used building material worldwide because it has been demonstrated to have high performance relative to its low cost. However, the decarbonization of the cement industry, which emits large amounts of carbon dioxide, is essential for sustainable development.[1] Carbon capture utilization and storage (CCUS) technologies are being extensively studied as one of several methods to reduce carbon dioxide emissions in some heavy carbon emitting industries. Among these, the storage relying on mineral carbonation would be accepted if it can prove to permanently store carbonation products.

Recently, recycled concrete paste (RCP) has been studied as a material with significant CO_2 sequestration potential even in ambient conditions.[2] Recycled concrete paste, derived from demolished concrete, consists of hydrated cement paste and fine fractions of coarse and fine aggregate.[3] However, further carbonation of recycled concrete paste is challenging due to the presence of calcium-silicate-hydrate formed over extended curing periods and calcite formed during natural carbonation.

In this study, a chemical admixture during the mixing process to enhance the CO_2 sequestration ability of recycled cement paste. According to Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES), the use of a chemical admixture resulted in two to three times more calcium leaching compared to pure RCP samples (i.e., without chemical admixture). Consequently, samples with varying contents of the chemical admixture were demolded and subjected to CO_2 curing.

To evaluate the effect and mechanical performance of the chemical admixture on CO_2 fixation, X-ray Diffraction (XRD), Thermogravimetric Analysis (TGA), and compressive strength tests were conducted. The XRD results indicated that an optimal amount of the chemical admixture decomposed all portlandite present in the waste concrete and produced a larger amount of calcite. TGA analysis confirmed that the chemical admixture facilitated the formation of calcium carbonate, the most thermodynamically stable form (i.e., calcite), rather than aragonite or vaterite.

Keywords

CO_2 Curing; carbon dioxide; mineral carbonation; recycled concrete paste; calcite

References

[1] M. Schneider, The cement industry on the way to a low-carbon future, Cem Concr Res 124 (2019). https://doi.org/10.1016/j.cemconres.2019.105792.

[2] M. Zajac, J. Skocek, Ł. Gołek, J. Deja, Supplementary cementitious materials based on recycled concrete paste, J Clean Prod 387 (2023). https://doi.org/10.1016/j.jclepro.2022.135743.

[3] M. Zajac, J. Skibsted, F. Bullerjahn, J. Skocek, Semi-dry carbonation of recycled concrete paste, Journal of CO2 Utilization 63 (2022). https://doi.org/10.1016/j.jcou.2022.102111.

RAPID CARBONATION PROCESS OF RECYCLED CONCRETE POWDERS IN A HIGH-GRAVITY ROTATING PACKED BED

Zhenjiang Gu[1], Long Jiang[1], Zihan Ma[1], Yi Jiang[1], Peiliang Shen[1], Chi Sun Poon[1]

[1]Department of Civil and Environmental Engineering & Research Centre for Resources Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hong Kong 999077, China

Email: Yi289.jiang@connect.polyu.hk; cecspoon@polyu.edu.hk

Abstract

Carbonated recycled concrete powders (RCPs) possess the potential to serve as supplementary cementitious materials (SCMs). However, the practical implementation of accelerated carbonation for RCPs is impeded by sluggish carbonation efficiency. In this study, a novel wet carbonation conducted in a rotating packed bed (RPB) was introduced, which achieves high-efficiency carbonation of RCPs. In the RPB carbonation, a noteworthy carbonation degree of 76 % was achieved within 10 min, with the formation of a core-shell-structure in RCPs particle. The formed $CaCO_3$ (shell) is primarily calcite with high crystallinity and the formed silica gel (core) features a high polymerization degree compared with those formed in normal wet carbonation, which is due to the high mass transfer rate in the RPB. Carbonation in RPB can break the limiting steps in different stages, thereby improving carbonation efficiency and enhancing CO_2 emission reduction capabilities. The proposed RPB carbonation method provides a promising way of pushing industrial CO_2 capture and waste concrete utilization.

Keywords

High-gravity rotating packed bed; recycled concrete powders; aqueous carbonation; CO_2 sequestration

CONNECTION BETWEEN CARBONATION REGIMES AND EARLY POZZOLANIC REACTIVITY OF RECYCLED CONCRETE POWDER: IMPACT OF COMPOSITION AND MICROSTRUCTURE

Zihan Ma[1], **Yi Jiang**[1], **Jionghuang He**[1], **Peiliang Shen**[1], **Qinglong Qin**[1], **Zhenjiang Gu**[1], **Jiangshan Li**[2, 3], **Chi-sun Poon**[1]

[1] Department of Civil and Environmental Engineering & Research Centre for Resources Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hong Kong 999077, China
[2] State Key Laboratory of Geomechanics and Geotechnical Engineering, Institute of Rock and Soil Mechanics, Chinese Academy of Sciences, Wuhan 430071, China
[3] IRSM-CAS/HK PolyU Joint Laboratory on Solid Waste Science, Wuhan 430071, China
Email: peiliang.shen@polyu.edu.hk; cecspoon@polyu.edu.hk

Abstract

This study aims to establish relationship between carbonation regimes and the early pozzolanic reactivity of carbonated recycled concrete powder (CRCP) by comparing the composition, structure and surface properties of the carbonation products. The surface of dry CRCP was characterized by a silica-rich layer and contains low-polymerized silica phases along with over 18% of unstable calcium carbonate (Cc). Wet CRCP exhibits a silica-rich surface with the highest degree of silica polymerization and stable calcite in its composition. Semi-dry CRCP has a calcium-rich surface also contains high silica polymerization and over 29% unstable calcium carbonate, and the highest specific surface area, leading to the fastest silicon dissolution and calcium consumption during pozzolanic reactions. The early pozzolanic reaction kinetics in semi-dry and wet carbonation are limited by the dissolution of carbonation products, whereas dry carbonation does not exhibit this limitation. Understanding this connection is crucial for selecting optimal carbonation techniques to enhance waste concrete utilization.

Keywords
Recycled concrete powder; carbonation; carbonation regimes; calcium carbonate

VATERITE $CaCO_3$ BINDER FROM INDIRECT CARBONATION OF WASTE CONCRETE FINES

Tiejun Ding[1], **Hong Wong**[1], **Marcus Yio**[1] **and Christopher Cheeseman**[1]

[1]Centre for Infrastructure Materials, Department of Civil and Environmental Engineering,
Imperial College London, SW7 2AZ, United Kingdom
Email: tiejun.ding18@imperial.ac.uk

Abstract

The management of waste concrete necessitates the development of better recycling to extract value-added products. A two-step process involving acetic acid leaching of hydrated cement paste and carbonation of the Ca-rich leachate is developed to produce vaterite, a metastable polymorph of calcium carbonate. The vaterite is blended with 0.1 mol/L $MgCl_2$ and $SrCl_2$ solution and converted to needle-like aragonite via dissolution-precipitation at 60 °C to produce a viable binder. The effect of different liquid/solid ratios (0.5, 0.6 and 0.7) and curing period (5h, 1, 3, 7 and 28d) on the strength, phase evolution and microstructure of the $CaCO_3$ binder are investigated. The underlying polymorphic transformation and strength development mechanism are reported.

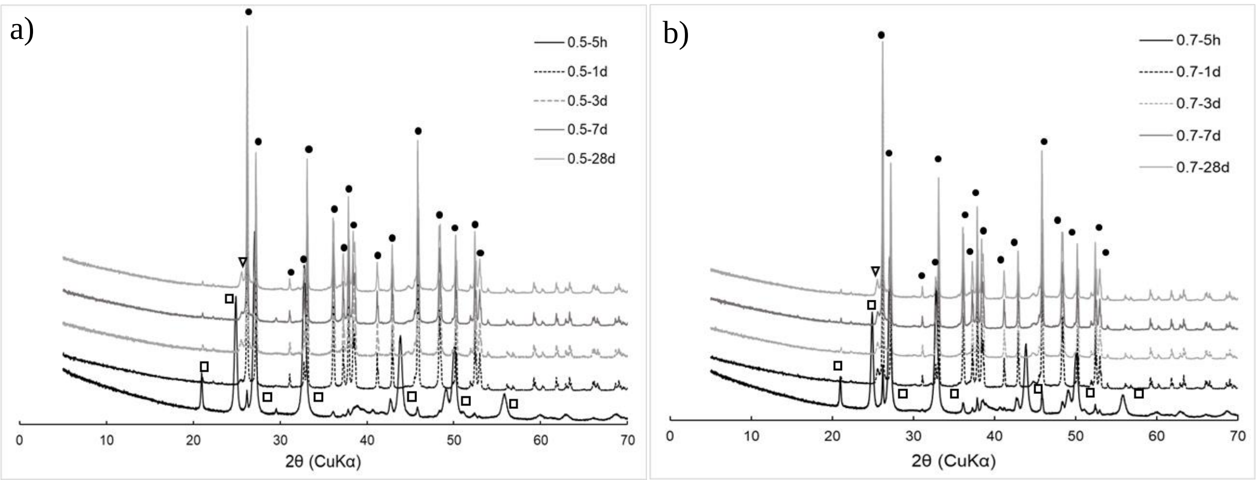


Figure 1. XRD patterns of $CaCO_3$ binder samples prepared at liquid/solid ratio of (a) 0.5 and (b) 0.7 and cured at 60°C for different times (□ – Vaterite; • – aragonite; ▽ – Ca-strontianite)

XRD patterns (Figure 1) show that the transformation from vaterite to aragonite starts within 5 hours and completes in 1 day. Increasing the L/S ratio can promote the dissolution of vaterite and accelerate the aragonite formation. Some Ca-bearing strontianite forms, indicating Sr precipitates with aragonite $CaCO_3$ which explains why this polymorph is stabilised.

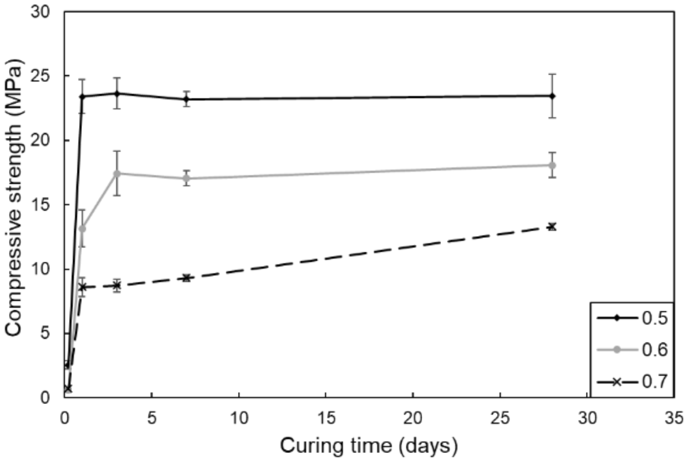


Figure 2. Compressive strength evolution of $CaCO_3$ binder at L/S ratio of 0.5, 0.6 and 0.7.

The compressive strength of 20mm $CaCO_3$ cubes (Figure 2) increases with curing age and lower L/S ratio. This is due to the reduced porosity and characteristic arrangement of the microstructure. The highest compressive strength is 23.6 MPa at L/S ratio 0.5 after 3-day curing.

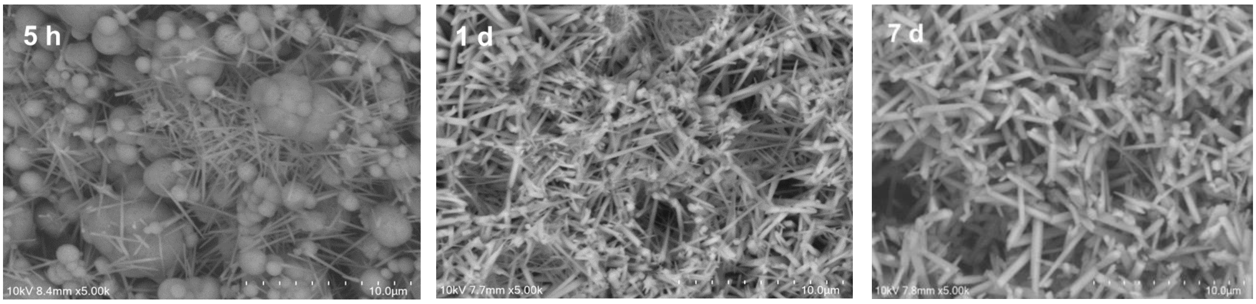


Figure 3. SEM images of hardened $CaCO_3$ binder (L/S ratio 0.7) showing the transformation from spherical vaterite to needle-like aragonite.

SEM images (Figure 3) show the microstructure of hardened $CaCO_3$ binder over time, taking L/S = 0.7 as an example. At 5 hours, clusters of short thin needle-like aragonite start to grow from the edges of dissolving vaterite particles forming bridges between them. As the transformation progresses to 1 day, vaterite has fully dissolved leaving spherical cavities. The aragonite crystals increase in size and form an interlocked structure between the spherical cavities. This micro-truss structure improves the compressive strength of the $CaCO_3$ binder. At 7 days, the aragonite crystals continue to grow in length and diameter while maintaining their dimensional uniformity across the sample. This leads to later-stage strength gains.

In conclusion, vaterite has cementing properties by undergoing a dissolution-precipitation transformation process to aragonite with a strong interlocked structure. Water provides a medium for dissolution. This generates connected porosity which has implications for the application of this material. This process contributes to establishing a circular economy for concrete by recovering low-carbon and high-value $CaCO_3$ binder from waste concrete.

Keywords
Waste concrete fines; circular economy; vaterite; low-carbon technology; calcium carbonate cement

Session A4

Clinkering of Carbonatable Binder

ELUCIDATING THE ROLE OF DIFFERENT PHASES IN CARBONATABLE BINDERS

Liu, Z.[1], Lv, C.[2], and Wang, F.[3]

[1] School of Materials Science and Engineering, Wuhan University of Technology, Wuhan, 430070, China
[2] State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan, 430070, China
Email: liuzc9@whut.edu.cn

Abstract

Accelerated carbonation of carbonatable clinkers into building products is an effective way of CO_2 utilization. However, due to insufficient understanding on the phase characteristics of carbonatable clinkers, there is still a lack of guidance on the selection and design of carbonatable clinkers. In this study, three γ-C_2S based carbonatable clinkers were designed and synthesized, covering the carbonation active phase, the unavoidable C_2AS and amorphous glass phases when using industrial feedstocks. The differences in the carbonation activity, mechanical properties and microstructure were compared. Results show that the uncarbonated phases have a significant impact on the mechanical properties of carbonated matrix. The presence of unreacted γ-C_2S with self-pulverization induced cleavage planes and the amorphous glass phase with poor binding to the adjacent calcium carbonate crystals leads to reduced compressive strength. The carbonation reactivity of γ-C_2S formed in composite system is significantly higher than that of pure γ-C_2S. Benefiting from the higher degree of carbonation, carbonatable clinkers only need to contain more than 40wt. % of γ-C_2S to obtain comparable compressive strength as the pure γ-C_2S system.

Keywords

Carbonatable binders; γ-C_2S; calcium aluminate; C_2AS; accelerated carbonation

TRANSFORMING FERRONICKEL SLAG IN MAGNESIUM SILICATE BINDER AFTER A CARBONATION/DECARBONATION PROCESS

Laniesse, P.[1], Kmeid, M.[1], Patapy, C.[1], Julcour, C.[2], Bourgeois, F.[2], Cyr, M.[1]

[1]LMDC, Université de Toulouse, UPS, INSA, Toulouse, France
[2]LGC, Université de Toulouse, CNRS, INPT, UPS, Toulouse, France
Email: cyr@insa-toulouse.fr

Abstract

Around 2 million tons of ferronickel slags are produced annually in New Caledonia, and more than 90% is actually landfilled. Ferronickel slag is composed of more than 95% of MgO, Fe_2O_3 and SiO_2, but mainly in the form of magnesium-iron silicates of the olivine group, such as ferroan forsterite ($(Mg,Fe)_2SiO_4$), and magnesium silicates of the pyroxene group, such as clinoenstatite ($MgSiO_3$). When quenched with water, the content of amorphous phases could be higher than 50%, but it has been shown that the interest of using them as supplementary cementing materials (SCM) in replacement of Portland cement was limited, as the presence of magnesium silicate decreased the reactivity of the product.

It has been shown that ferronickel slag could be carbonated by a process of attrition-carbonation in a controlled humidity/temperature carbonation device [Bourgeois et al., 2020], at rates between 20% and 80% of the native slag. The resulting material allowed the separation of the magnesium from the silica in two distinct mineral forms: crystallized magnesite ($MgCO_3$) and amorphous silica (SiO_2). This process helped to improve the reactivity of the silica fraction with calcium hydroxide in Portland cement-based materials. A more reactive SCM was thus produced with this process [Cyr et al., 2023], but the free silica in the carbonated slag still needed Portlandite (coming from Portland cement hydration) to react.

Knowing that the carbonated product contains amorphous silica but also magnesium carbonate, which could form magnesium oxide (periclase) after heating, thus magnesium silicate binders could be synthetized (see for instance Walling et al. 2016). The CO_2 used for the initial carbonation process would be released again, but could be recycled for future carbonation of other slag. Moreover, this process could help avoiding the decarbonation of natural $MgCO_3$ that is typically used for the production of MgO in the magnesium-silicate binder synthesis.

The transformed ferronickel slag could then be used as the sole binder that would not need other constituent such as Portland cement to harden. The aim of this project is to study the production of such a binder from ferronickel slag, and to evaluate its reactivity.

Carbonation process
The carbonation reaction here takes place in aqueous phase using a laboratory 300 mL autoclave operated as an attrition-leaching reactor: it contains a bed of millimeter-sized grinding beads stirred by a pin-drive impeller. This device enables to control both pressure and temperature, with a continuous attrition of the slag particles to remove leached layers. More detailed description of the reactor is provided by [Dufourny, 2022]. The temperature and pressure were kept constant during the whole reaction that last 24h. In the investigated conditions, the temperature was maintained at 180°C and CO_2 partial pressure at 20 bars.

Synthesis of the magnesium-silicate binder

Table 1 and 2 report the chemical and mineralogical compositions, respectively, of the native and carbonated slags. The experiments were carried out to determine whether it was possible to synthesize periclase (MgO) and within which temperature range. Results showed that the decarbonation of magnesite started from 500°C, and that between 500 and 600°C, magnesite ($MgCO_3$) was transformed in periclase. However, at higher temperature, only magnesioferrite ($MgFe_2O_4$) was formed. Then it was decided to maintain the processing temperature between 500 and 600°C in order to prepare the silico-magnesium binder.

Table 1: Oxide composition (%) of native and carbonated slags obtained from ICP-AES analysis

	MgO	SiO_2	Al_2O_3	CaO	Fe_2O_3	L.O.I
Ferronickel slag	31.2	52.6	2.7	0.70	12.4	-
Carbonated slag	20.3	35.6	2.8	1.21	10.4	30.3

Table 2: Rietveld refinement results (%) for the native and carbonated slags

	$(Mg,Fe)_2SiO_4$	$MgSiO_3$	$MgCO_3$	$FeCO_3$	amorphous
Ferronickel slag	44.1 ± 0.6	0.4 ± 0.3			55.4 ± 0.6
Carbonated slag	7.0 ± 0.6	-	37.3 ± 0.7	3.6 ± 0.3	52.1 ± 1.0

Reactivity of the magnesium-silicate binder

Paste was cast with the binder prepared after treatment for 1 h at 600°C, using a liquid-to-binder ratio of 1.2. After 7 days, the paste was characterized by XRD (Figure 1), and the presence of M-S-H ($MgO-SiO_2-H_2O$) was confirmed.

Hydration was monitored by isothermal microcalorimetry at 20°C (Figure 2). It can be seen that this binder released more that 200 J/g of heat, i.e. around two thirds of typical a Portland cement containing 95% of clinker.

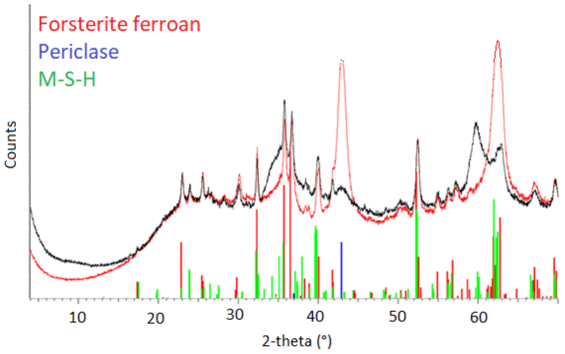


Figure 1: XRD diffractograms of the carbonation slag treated at 600°C before (red spectrum) and after 7 days of hydration (black spectrum)

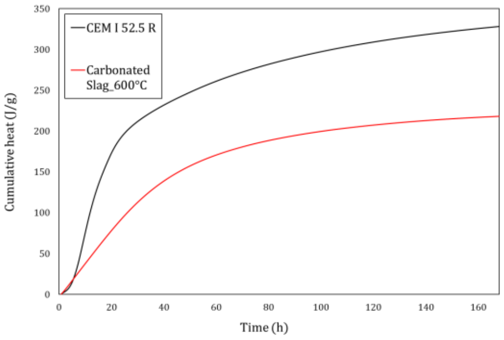


Figure 2: Calorimetry curves for silico-magnesian binder paste with carbonated slag and Portland cement

Keywords

Ferronickel slag; magnesium-silicate binder; mineralization

References

Bourgeois F., Laniesse P., Cyr M., Julcour C., Definition and Exploration of the Integrated CO_2 Mineralization Technological Cycle. Frontiers Energy Research 8 (2020) 113.

Cyr M., Laniesse P., Dufourny A., Julcour C., Bourgeois F., Assessment of carbonated ferronickel slag as supplementary cementing materials with improved carbon impact and reactivity, WASCON 2023, Dec 2023, Hong Kong.

Dufourny A., Julcour C., Esvan J., Cassayre L., Laniesse P., Bourgeois F., Observation of the depassivation effect of attrition on magnesium silicates direct aqueous carbonation products, Sec. Negative Emission Technologies, Front. Clim. 4:946735 (2022).

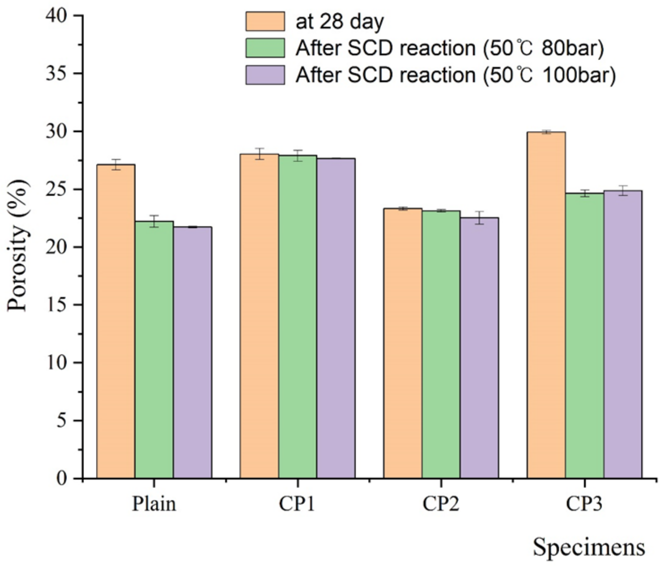
DURABILITY OF CALCIUM PHOSPHATE MODIFIED PORTLAND CEMENT EXPOSED TO SUPERCRITICAL CO_2 ENVIRONMENT

Kim, J. H.[1], Son, J. J. [1], Lee, Y. K.[2], Chung, C. W.[1]

[1]Division of Architectural and Fire Protection Engineering, Pukyong National University,
Busan, Republic of Korea
Email: kjh@pknu.ac.kr

Abstract

When CO_2 is captured, it is injected into deep geologic repository. Concrete is placed into annulus in order to prevent the leakage of CO_2. Since CO_2 is stored as a supercritical state, the carbonation reactivity is so aggressive. In such environment, Ca^{2+} ion concentration in portland cement concrete drops quickly due to the rapid consumption of calcium hydroxide to produce calcium carbonate polymorphs. Such an abrupt reaction can not only generate microcracks (by rapid formation of calcium carbonate polymorphs), but also decrease the stability of the calcium silicate hydrate. The durability of concrete used for sealing the annulus becomes a key issue for permanent safe sequestration of CO_2. In this work, to minimize the damage caused by the attack of supercritical CO_2, portland cement was modified by calcium phosphates to produce a more thermodynamically stable phase than calcite. Monocalcium, dicalcium and tricalcium phosphates were added to portland cement for this purpose. The results indicated that hydroxyapatite was successfully produced by the reaction between calcium phosphate and portland cement. Among three different calcium phosphates, monocalcium phosphate showed the most significant impact on hydration and rheology, and caused rapid stiffening with the delay in calcium silicate hydration. Although cement pastes modified by calcium phosphates showed less compressive strength than plain cement paste, they showed much higher resistance against supercritical CO_2. The potential applicability of calcium phosphate modification in portland cement was successfully proven.



Keywords
Calcium phosphate; hydroxyapatite; durability; supercritical CO_2

THE UTILIZATION OF RED MUD FOR URANIUM MINERALIZATION BY A LOW-TEMPERATURE SINTERING PROCESS

Minhua Su[1], Zheng Chen[1], and Jinfeng Tang[1]

[1]School of Environmental Science and Engineering, Guangzhou University, Guangzhou 510006, China
Email: jinfeng@gzhu.edu.cn

Abstract

Red mud, a highly alkaline aluminum industrial waste residue, is considered a promising sorbent for the removal of toxic metal ions from industrial effluents. Red mud has been used to enrich uranium(VI) ions and immobilize uranium(VI) into specific mineral phases. Here, a low thermal stabilization process was well-designed for stabilizing uranium(VI) in red mud into particular phase(s) such as geopolymer-like microstructures. Characterization and leaching experiments were further performed for uranium-containing RM samples thermally stabilized at various temperatures, and the stabilization behavior of uranium in the RM was determined. X-ray diffraction (XRD) revealed that a geopolymer-like phase whose main components were Al, Si and O was formed when the U(VI) adsorbed RM was treated at 600 °C; thus, uranium was successfully stabilized. When treated at higher temperatures (i.e., 900 °C), the RM formed feldspar minerals from the oxides of Si and Al, which originated from the broken geopolymers (a stable three-dimensional network), thus exposing uranium to the surface. The constant-pH leaching test, evidenced that the sintering temperatures of 600–800 °C facilitated the fixing of uranium by RM. The findings of this study offer us a promising strategy on the utilization of RM to eliminate and stabilize uranium(VI) from waste streams.

Keywords
Red mud; sintering; uranium; stabilization; waste treatment

NOVEL INSIGHT TO PREPARING HIGH CARBONATION REACTIVITY WOLLASTONITE FIBERS THROUGH PHASE TRANSFORMATION TREATMENT

Donglin Li[1], Pengjie Rong[1], Songhui Liu[1], Peiliang Shen[2], Jian-xin Lu[2], Chi-Sun Poon[2]

[1]Henan Key Laboratory of Materials on Deep-Earth Engineering, School of Materials Science and Engineering, Henan Polytechnic University, Jiaozuo, 454003, China
[2]Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong
Email: liusonghui@hpu.edu.cn

Abstract

The production of traditional cementitious material OPC is accompanied by massive CO_2 emissions, therefore the development of low CO_2 emission cementitious materials has received increasing research attention. This study prepared high carbonation reactivity wollastonite fibers through phase transformation treatment using natural wollastonite as raw material, and investigated the carbonation hardening properties, CO_2 sequestration capacity, phase composition, microstructure, and carbonation mechanism of the high carbonation reactivity wollastonite fibers. First-principles theoretical simulations were used to reveal the influence of CS mineral structure and electronic properties on carbonation reactivity. The results show that after phase transformation treatment, natural wollastonite transformed into Pseudo-wollastonite and Cyclo-wollastonite with high carbonation reactivity while retaining its original fibrous structure. After carbonation, it exhibited excellent mechanical properties (compressive strength and flexural strength of 80.72 MPa and 10.07 MPa, respectively), dense microstructure (calcium carbonate and silica gel), and significant CO_2 sequestration potential (~14.86%). The fibrous structure provided a larger specific surface area, finer particle size, higher carbonation reactivity and more CO_2 diffusion channel. Structural and electronic property calculations further revealed differences in mineral carbonation reactivity. The Ca-O bonds of high-temperature CS minerals have lower BO values and higher BL values, making chemical bonds easier to break. Additionally, the isolated ternary ring structure of [SiO_4] tetrahedra results in Ca^{2+} ions being more easily dissolved to participate in reactions. This study provides a novel approach for large-scale utilization of wollastonite to develop high carbonation reactivity cementitious materials.

Keywords: CO_2 sequestration; wollastonite fibers; high carbonation reactivity; first principles

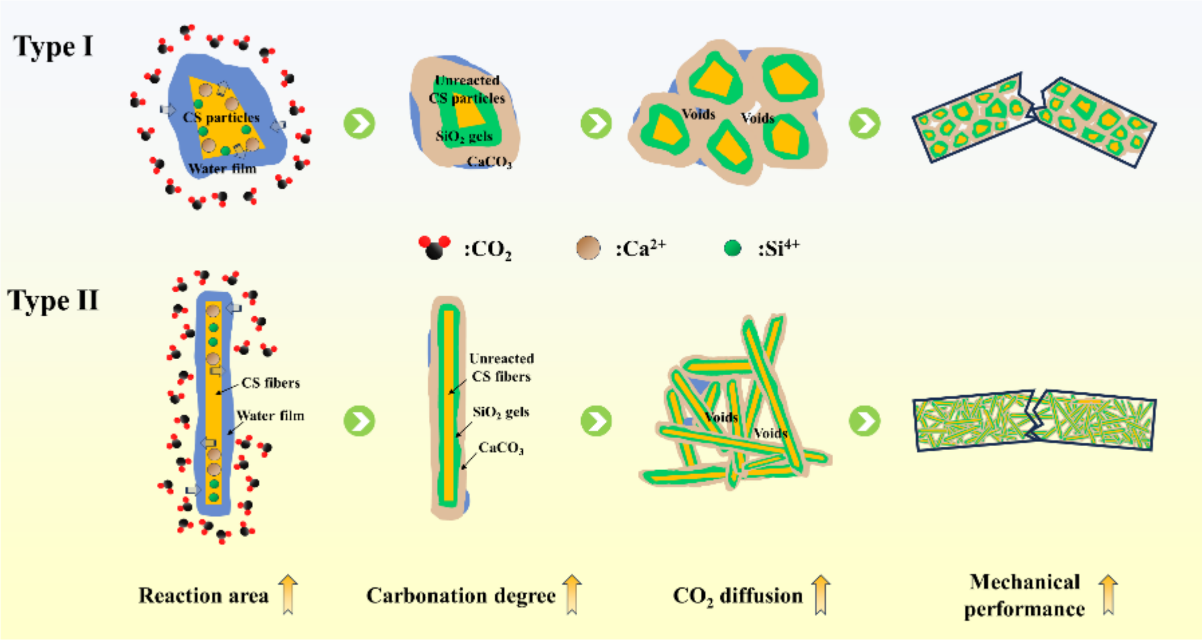


Fig. 1 Carbonation process and destruction process of different CS morphology

FEASIBILITY, PERFORMANCE AND CARBONATION MECHANISM OF TERNESITE AS AN ULTRA-LOW LIME CO_2 SEQUESTRATION BINDER

Xiaoyun Du[1], Jun Chang[1]

[1]School of Infrastructure Engineering, Dalian University of Technology, Dalian, 116024, China
Email: mlchang@163.com

Abstract

The carbonation hardening of low lime carbonatable binders has become an important way to reduce carbon emissions in the cement industry. In order to further reduce the carbon footprint of low lime binders, this work proposes using ternesite as a CO_2 sequestration binder. This study used a semi dry method to carbonation the compacts of ternesite at different water solid ratio, initial porosity, CO_2 pressure and reaction periods. The evolution of ternesite carbonation products and properties was studied by qualitative and quantitative analysis, and the kinetics equation of ternesite carbonation was established. According to the microstructure, crystal size and phase composition of carbonated products, a conceptual model of the carbonation hardening process mechanism of ternesite was constructed. The carbonation performance of ternesite was characterized under different process parameters and carbonation conditions. The results showed that ternesite CO_2 sequestration capacity and compressive strength of ternesite reached 15.1% and 91.95 MPa after carbonation for 120 min. Compared with other low lime minerals, ternesite as an ultra-low lime CO_2 sequestration binder has significant competitiveness in carbonation performance, limestone consumption, total carbon emissions, and other aspects, opening up the field of carbonatable binders.

Keywords

Ternesite; carbonation; carbonatable binders; CO_2 sequestration binder; low-carbon cement

Acknowledgements

This work was funded by the National Key Research and Development Program of China (2022YFE0208200).

DESIGN OF C_2S-CS LOW-CALCIUM PHASE SYSTEM FOR SYNERGISTIC IMPROVEMENT OF CO_2 SEQUESTRATION CAPACITY AND MECHANICAL PROPERTIES

Ning Tan[1], **Zengmei Wang**[1]

[1] School of Materials Science and Engineering, Jiangsu Key Lab. of Construction Materials,
Southeast University, Nanjing 211189, PR China.
E-mail address: 101011338@seu.edu.cn (Z. Wang)

Abstract

Mineral Carbonation Technology (MCT) enables low-calcium minerals to achieve excellent mechanical properties, serving as an energy-efficient binder with reduced emissions and CO_2 sequestration during curing. However, $CaSiO_3$ (CS), β-Ca_2SiO_4(β-C_2S), and γ-Ca_2SiO_4(γ-C_2S) individually lack optimal carbon sequestration capacity, strength, and durability. This paper presents a novel low-calcium C_2S-CS system, developed using Simplex-centroid designs to integrate γ-C_2S, β-C_2S, and CS, enhancing early carbonation and long-term strength. Contour maps assess the performance-mineral composition relationship, and interaction mechanisms are explored via carbonation products and reaction heat analysis. Synthesized from solid waste wollastonite tailings in a single step, the system streamlines preparation and reduces energy use. After 24 hours of carbonation curing, it achieves 129 MPa compressive strength, 217 kg/t CO_2 sequestration, and satisfactory durability, providing valuable insights for designing and optimizing low-calcium CO_2 sequestration cementitious materials (LCC) and suggesting approaches for industrial LCC manufacturing.

Keywords

α /β-CS; γ-C_2S; β-C_2S; low calcium CO_2 sequestration cementitious material; solid waste resource utilization

REVISITING THE CARBONATION BEHAVIOR OF C_3S_2 TO ELUCIDATE THE ROLE OF SILICA GEL IN THE MECHANICAL STRENGTH OF CARBONATED MATRIX

Zhang, Z.[1,2], Liu, Z.[1,2], Wang, F.[1], and Hu, S.[1]

[1]State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan, 430070, China
[2]School of Materials Science and Engineering, Wuhan University of Technology, Wuhan, 430070, China
Email: liuzc9@whut.edu.cn

Abstract

This work addresses the characteristics of C_3S_2 under enforced carbonation including the dissolution of ions, reaction degree, phase assemblage, microstructure and mechanical strength. The compression molded C_3S_2 specimens achieve a compressive strength of 108.4 MPa within 24 hours. The reaction degree of C_3S_2 calculated according to ^{29}Si MAS NMR spectra reaches 54.8% and 66.3% after 1 day and 21days of CO_2 exposure, respectively, which are much higher than that of reaction degree calculated according to CO_2 uptake. Different from the previous studies, silica gel is found to be distributed evenly between calcium carbonate in C_3S_2 resulting from the slower dissolution rate of Ca^{2+}. The interface between silica gel and calcium carbonate distracts crack advance and provides stress relief, which could be another contribution of silica gel on mechanical strength apart from filling of pores and the densification of the matrix.

Keywords

Rankinite; accelerated carbonation; silica gel; microstructure; mechanical property

PRODUCTION OF MSWI-BASED BELITE-TERNESITE CEMENT WITH ENHANCED CO_2 REACTIVITY

Xiaoli Wang[1], and Tung-Chai Ling[1]

[1]College of Civil Engineering, Hunan University, Changsha 410082, Hunan, China
Email: tcling@hnu.edu.cn; tcling611@yahoo.com

Abstract

The cement industry is responsible for approximately 7%~8% of global anthropogenic CO_2 emissions. Specifically, the colossal CO_2 footprint is predominantly linked to the calcination of limestone and the associated fuel combustion for the production of cement clinker mineral. Fundamentally, the primary source of the above carbon emissions is clinker mineral with high calcium content (e.g., 55-70% C_3S). This implies that, despite improvements in energy efficiency or the use of green energy in the production process, clinker production is a significant source of CO_2 emissions. Therefore, developing a low-carbon type of cement with a different chemistry than ordinary Portland cement clinker could be an important step in cement industry to achieve carbon neutrality target.

Unlike the C_3S phase, the belite (β-C_2S) phase with a relatively lower CaO/SiO_2 ratio allows the use of lower-grade limestone (or waste-based source) and alternative fuels with lower caloric values for sintering. Ternesite ($2C_2S{\cdot}CaSO_4$, $C_5S_2\bar{S}$), a typical intermediate phase, has been used as a constituent in sulphoaluminate cement (SAC) with a sintering temperature below 1250°C, and it releases around 14% less CO_2 than C_3S. Hence, β-C_2S and $C_5S_2\bar{S}$ are also typically considered low-carbon clinker phases, but they have significantly low hydraulic reactivity, especially in the early age. The hydraulic reactivity of β-C_2S is only about 1/20th of that attained by the C_3S phase, and the hydration degree of $C_5S_2\bar{S}$ is only 3% and 12% in 7 days and 2 years, respectively. Although many measures have been taken to improve their reactivity, the economically prohibitive nature limits their industrial applicability.

Advances in carbon mineralization have attracted extensive attention to provide a possible solution for carbonatable calcium silicate to sequestrate CO_2 back in cement-based materials. These carbonatable calcium silicate-based cements are estimated to reduce total CO_2 emissions from cement production by about 70%. Carbonation can activate the reactivity of β-C_2S due to its strong carbonation reactivity. So far, most reported studies on $C_5S_2\bar{S}$ have mainly focused on the preparation process and related enhancement of hydration reactivity. At present, only one study reported the effect of carbonation curing on the compressive strength of $C_5S_2\bar{S}$, and found that carbonation curing is more effective in gaining strength than normal curing.

Given the research gap in the combined utilization of β-C_2S and $C_5S_2\bar{S}$ minerals in carbonatable binders as low carbon cement, the key novelty of the present work is to break through the traditional hydration hardening of cement minerals and replace by carbonation hardening approach. Both CO_2 highly reactive minerals of β-C_2S and $C_5S_2\bar{S}$ were prepared using 100% MSWI residues-fly ash (FA) and bottom ash (BA). The CO_2 capture capacity, structural property, phase evolution, and carbonation hardening mechanism were studied to determine the optimal CaO/SiO_2 (C/S) ratio and sintering temperature from the perspective of carbonation reactivity.

The results showed thatβ-C_2S and $C_5S_2\overline{S}$ are the major mineral phases, accounting for 31.3-38.9% and 12.7-28.3%, respectively. Upon 1 day of CO_2 curing, about 21.0% β-C_2S and 15.1% $C_5S_2\overline{S}$ was consumed to produce 21.1% calcite and 6.8% gypsum. The average compressive strength of the carbonated cement was exceeding 100 MPa, and mineralized over 10% of CO_2. It should be noted that this specialty cement can gain very high strength within a short period of time. At 6 h of CO_2 curing, the cement paste achieved a strength of 66 MPa, which is comparable to the 7-day strength of grade 42.5 OPC paste. Interestingly, the morphology of $CaCO_3$ changed significantly over the carbonation time, the needle-like $CaCO_3$ with a length of 2-4 μm found at 2 h was gradually evolved into more compact clusters at 6 h, followed by a coarser growth pattern structure at 1 day. As expected, the porosity also decreased from 18.6% to 10.4%. This is mainly due to the fact that the mineral particles are uniformly bound together and filled with carbonation products, leading to a denser structural matrix.

Keywords

Municipal solid waste; belite-ternesite phase; CO_2 uptake; carbonation reactivity

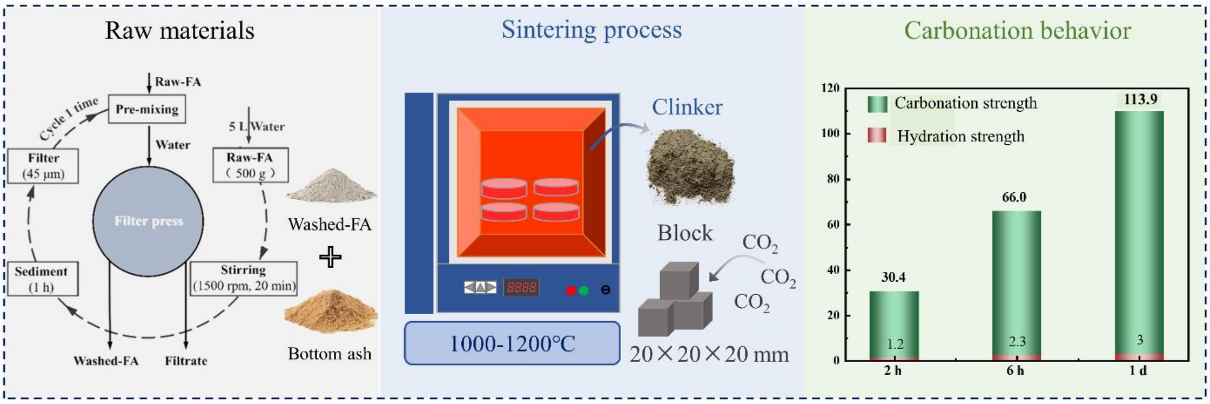


Fig. 1 The preparation process and carbonation behavior of belite-ternesite cement

CHLORELLESTADITE: AN APT BINDER OR AN SCM WITH CARBONATION REACTIVITY

Hanxiong Lyu[1], Shipeng Zhang[1], Chi Sun Poon[1]

[1]Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, 11 Yuk Choi Road, Hung Hom, Kowloon, Hong Kong, China
Email: shipeng.zhang@polyu.edu.hk

Abstract

Chlorellestadite (ideal formula of $Ca_{10}(SiO_4)_3(SO_4)_3Cl_2$, CE) is an intermediate mineral that forms during the fabrication of Portland cement or ecological clinkers from solid wastes containing both sulfate and chlorine. Due to its water-insolubility, CE presents challenges for application in typical construction practices. To indirectly promote the use of chlorine and sulfate-containing waste as feedstock in cement production, this work investigated the viability of employing CE as a binder or an SCM under a binary curing system of carbonation and hydration. Results showed that CE had carbonation reactivity, enabling the sequestration of 4.8% (by mass) of CO_2 and the development of a compressive strength of 25.8 MPa after undergoing 24 hours of carbonation curing with $CaCO_3$, $CaSO_4$, $CaCl_2$, and silica gel as carbonation products. Substituting 20 wt% CE in dry-cast pastes (CE20) resulted in a reduction in 1d compressive strength due to the decreased cement content. However, carbonation of CE introduced secondary gypsum into the binder system, promoting the formation of an additional AFt phase within pores during subsequent water curing. Consequently, the 28d strength of CE20 (93.4 MPa) surpassed the OPC reference by 21.1%. These findings underscore not only the potential of using CE as a binder or an SCM in dry-cast non-structural concrete but also the feasibility of utilizing waste materials containing sulfate and chlorine as feedstock for synthesis, thereby mitigating environmental risk associated with such wastes.

Keywords

Chlorellestadite; carbonation reactivity; binder; supplementary cementitious material; solid waste

DEVELOPMENT OF HIGHLY CARBONATION-EFFECTIVE CALCIUM SILICATES (β-C_2S): PHASE EVOLUTION, MICROSTRUCTURE, AND CARBONATION MECHANISMS

Miao Ren[1], **Peiliang Shen**[1], **Yong Tao**[1], **Chi-sun Poon**[1]

[1] [1]Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hong Kong, China
Email: peiliang.shen@polyu.edu.hk

Abstract

Carbonation of calcium silicate (CS) minerals to produce hardened construction materials has attracted increasing interest, but the efficiency of conventional CS minerals is low. This study prepared a highly reactive β-C_2S derived from a C-S-H precursor through hydrothermal synthesis. The potential CO_2 uptake ability, product compositions, microstructural characteristics, and carbonation mechanisms of the hydrothermal β-C_2S were investigated. The results showed that the hydrothermal β-C_2S achieved 3.7 times higher carbonation efficiency compared to the reference β-C_2S prepared by traditional calcination and reached a carbonation degree of 56.1% within 10min. Its high reactivity was attributed to large specific surface area, unique thin lath structure, and fine grain size. The novel type of C-S-H produced, special thin lath structure evolution and sufficient ACC led to an almost complete carbonation degree of hydrothermal β-C_2S. The developed carbonation process could be used to valorize recycled concrete waste which contains a large amount of C-S-H.

Session B1

CO_2 Activation of Recycled Concrete Aggregates

IMPACT OF CARBONATED RECYCLED AGGREGATES ON THE CARBONATION RESISTANCE OF CONCRETE

Etxeberria, M.[1]

[1]Department of Civil and Environmental Engineering, Universitat Politécnica de
Catalunya.BarcelonaTECH
E-mail (miren.etxeberria@upc.edu)

Abstract

Concrete construction has a high environmental impact due to the abundant energy consumption and the CO_2 emissions of cement production [1]. Several actions must be taken to reduce it. The use of recycled concrete aggregates (RCA) to replace natural aggregate partially lies in CO_2 emissions reduction in concrete manufacture and in the significant mitigation of the environmental impacts induced by stockpiling the respective waste [2]. In addition, the use of CO_2 for the carbonation process of RCA not only consumes a volume of emitted CO_2 but also improves the properties of RCA [3]. Moreover, the use of more sustainable types of cement produced using limestone, blast furnace slag (BFS) and fly ash (FA) as SCMs can also help reduce the environmental impact [4]. It is known that recycled aggregate concrete (RAC) has a more significant impact on durability than strength. The durability of RAC decreases as the amount of RCA used in the mix increases [5]. However, according to the carbonation resistance of RAC, these studies tend to be inconsistent [6] [7].

In this research, the carbonation resistance of structural concrete produced with 50% of carbonated and uncarbonated coarse RCA was analysed to be exposed to an XC4 environment. The two experimental phases were carried out. In Phase 1, the RCA was produced by crushing a parent concrete made with ordinary Portland cement (OPC), which was submitted to an accelerated carbonation process (C-RCA). All the concretes were made with 300 kg of cement of four types (CEM I, CEM IIAL, CEM IIAS and CEM IV of 42.5 MPa) and an effective water/cement (w/c) ratio of 0.48. In Phase 2, two types of recycled aggregates were used: RCA and RCA-FA, which were obtained by crushing the parent concrete made with OPC and fly ash (FA) blended cement. After the accelerated carbonation process, the aggregates C-RCA and C-RCA-FA were produced. The four types of RA (uncarbonated and carbonated) were used for concrete production. All the concrete mixtures were made using 350 kg of limestone cement (CEM IIAL) and an effective water-cement ratio of 0.52.

In both phases, the compressive strength and accelerated carbonation resistance were determined. Moreover, in phase 2, the physical properties and sorptivity were also determined and analysed. The results were also compared with those of conventional concrete (NC).

According to the obtained results, in phase 1, the recycled concretes achieved a 7% lower compressive strength than the NC concrete (58 MPa), except the concrete produced by C-RCA (carbonated), together with IIAS cement, which achieved a similar strength to NC. In addition, the concrete produced with RCA (uncarbonated) and the type IV cement achieved the lowest strength of 48 MPa. At 56 days, the concrete produced with RCA (uncarbonated) achieved the lowest strength (57.2 ±2.5MPa). In addition, C-RAC and NC achieved a compressive strength of 59.8 ± 3MPa and 64 MPa ± 3.5 MPa, respectively. According to the carbonation resistance, the C-RAC-IIAS (carbonated) achieved the lowest carbonation rate of 0.57 $mm/days^{0.5}$

followed by NC-I, RAC-IIAS, and NC-IIAS concretes. In addition, C-RAC-I (carbonated) achieved a value of 0.77 mm/days$^{0.5}$, a higher rate value than the obtained by RAC-I (uncarbonated) of 0.74 mm/days$^{0.5}$. There is little difference between the carbonation rate of concretes produced using carbonated or uncarbonated recycled aggregates when they were produced with the same type of cement. Nevertheless, the difference occurred when different types of cement were employed. The concretes produced with type IV cement achieved the highest carbonation rate, and the C-RAC-IV (carbonated) concrete achieved the highest carbonation rate of 1.1724 mm/days$^{0.5}$ among the concretes.

In Phase 2, at 28 and 56 days of curing, the CC concretes achieved the highest strength of 45.92MPa and 54 MPa, respectively. In addition, the RAC50 achieved 44.10MPa and 50.3MPa; the RAC50-C concrete obtained 45.71MPa and 50.65MPa; the RAC50-FA concrete reached 45.09 MPa and 49.90 MPa, and the RAC50-FA-C concrete obtained 43.82MPa and 49.07 MPa, respectively. According to carbonation, the NC concrete achieved the lowest carbonation rate, 0.926 mm/days$^{0.5}$, followed by the concretes produced with 50% RCA (0.958 mm/days$^{0.5}$, uncarbonated) and 50% RCA-FA (1.269 mm/days$^{0.5}$, uncarbonated). The CO_2-treated recycled aggregates worsened the carbonation resistance of the recycled concrete, and this worsening condition increased when the RCA-FA-C aggregates were used for concrete production. It achieved a value of 1.432 mm/days$^{0.5}$. The carbonation process of RCA-FA (originated from an FA blended cement) caused an increase in the mesopores and macropores of the pore size distribution. While the concrete produced with RCA-FA-C achieved the highest sorptivity value, the RCA-FA achieved the lowest sorptivity value of 0.032 mm/min$^{0.5}$, followed by the CC concrete with 0.035 mm/min$^{0.5}$ and the recycled concrete produced with 50% of the RCA-C (carbonated) with 0.036 mm/min$^{0.5}$.

It can be concluded that recycled concrete produced with carbonated aggregates achieved higher compressive strength than uncarbonated ones when the recycled aggregates were obtained from a parent concrete composed of OPC. However, the carbonation rate of concrete produced with carbonated or uncarbonated recycled aggregates was similar. This property depended on the type of cement used in recycled concrete production.

In contrast, the concrete produced with carbonated recycled aggregates achieved lower strength and higher carbonation rate when the recycled aggregates were obtained by crushing an FA parent concrete due to the increase in the mesopores and macropores of the pore size distribution on the paste of the recycled aggregate.

Keywords
Parent concrete with FA; recycled aggregate concrete; carbonation treatment; supplementary cementitious materials carbonation rate

[1] M. Schneider, “The cement industry on the way to a low-carbon future,” *Cem. Concr. Res.*, vol. 124, no. August, p. 105792, 2019.
[2] P. Plaza, I. F. Sáez del Bosque, M. Frías, M. I. Sánchez de Rojas, and C. Medina, “Use of recycled coarse and fine aggregates in structural eco-concretes. Physical and mechanical properties and CO2 emissions,” *Constr. Build. Mater.*, vol. 285, p. 122926, 2021.
[3] C. Liang, B. Pan, Z. Ma, Z. He, and Z. Duan, “Utilization of CO2 curing to enhance the properties of recycled aggregate and prepared concrete: A review,” *Cem. Concr. Compos.*, vol. 105, no. July 2019, 2020.

[4] J. M. Franco de Carvalho, W. C. Fontes, C. F. de Azevedo, G. J. Brigolini, W. Schmidt, and R. A. F. Peixoto, “Enhancing the eco-efficiency of concrete using engineered recycled mineral admixtures and recycled aggregates,” *J. Clean. Prod.*, vol. 257, 2020.

[5] C. Thomas, J. Setién, J. A. Polanco, P. Alaejos, and M. Sánchez De Juan, “Durability of recycled aggregate concrete,” *Constr. Build. Mater.*, vol. 40, pp. 1054–1065, 2013.

[6] H. Guo *et al.*, “Durability of recycled aggregate concrete – A review,” *Cem. Concr. Compos.*, vol. 89, pp. 251–259, 2018.

[7] A. Leemann and R. Loser, “Carbonation resistance of recycled aggregate concrete,” *Constr. Build. Mater.*, vol. 204, pp. 335–341, 2019.

WET-CARBONATION OF RCAs FOR IMPROVED CARBONATION EFFICIENCY AND MECHANICAL PROPERTIES OF CARBONATED RCAs AND RCA CONCRETE

Xiang Zhao, Qingli Dai and Zhanping You

Department of Civil, Environmental, and Geospatial Engineering,
Michigan Technological University

Abstract

With the increasing volume of buildings and constructions, the escalating amounts of construction and demolition waste (C&DW) pose a significant challenge to the environment. Addressing the management and treatment of C&DW is crucial. Recycled coarse aggregates (RCAs) typically exhibits weak interfacial transition zones (ITZ), high porosity, and micro-cracks due to residual mortar layers, resulting in suboptimal performance when reused in concrete for structural applications. Carbonated RCAs can be utilized in concrete, by enhancing their mechanical and durability properties. This study aims to improve carbonation efficiency of RCAs with the developed wet-carbonation method and to evaluate mechanical properties of carbonated RCAs and RCA concrete.

This study developed a wet carbonation process using glycine acid as an inducer. Glycine acid, containing carboxyl and amino functional groups, forms stable complexes with Ca^{2+} ions in RCA, increasing calcium ion solubility and thereby accelerating carbonation. RCA samples underwent carbonation in a glycine acid solution, followed by characterization using X-ray diffraction and scanning electron microscopy to evaluate mineralogical transformations and microstructural enhancements. The carbonation process promoted the formation of vaterite, a reactive polymorph of calcium carbonate, on RCA surfaces, which improved ITZ density and bonding strength with the cement matrix. As results, abrasion loss was achieved to 45.71% lower than 53.82% of untreated RCA, and California Bearing Ratio values increased by 28% at 5 mm penetration, indicating improved aggregate physical durability properties. Leachate analysis showed that carbonated RCA exhibited reduced to pH 8.47 and lower total dissolved solids at 0.263 mS, as the formations of calcium carbonate. Furthermore, recycled concrete mechanical tests indicated gains of 13% in compressive, 15% in tensile, and 12% flexural strength compared to untreated RCA concrete, reflecting increased strength gained and reduced porosity. These results demonstrate that glycine acid-induced carbonation enhances RCA properties and possibly offers a promising pathway for mitigating construction and demolition waste.

CO_2 FIXED BY RECYCLED AGGREGATE IN A MEDIUM-SCALE CARBONATION FACILITY

Takahiro Iwafuchi[1], Yoshiyuki Suzuki[1], and Kazuhiro Yamauchi[2]

[1]HAZAMA ANDO CORPORATION
[2]HAIKOU ONODA REMICON
Email: iwafuchi.takahiro@ad-hzm.co.jp

Abstract

Recently, the use of recycled aggregate with fixed CO_2 as a concrete material has been investigated. In previous studies, laboratory-scale experiments have confirmed that calcium carbonate is efficiently produced when recycled aggregate is changed from a wet to a dry state under high CO_2 concentration[1]. In this report, aiming for social implementation of this technology, a medium-scale CO_2 fixation facility was fabricated and tested, and the amount of CO_2 fixation and reaction were confirmed.

In this experiment, recycled aggregate with a grain size of 15 mm to 5 mm, which is used in pavement construction, was used. **Fig. 1** shows a photograph of the aggregate. This recycled aggregate is not concrete returned from the construction site, but aggregate that has been mixed and granulated for the experiment [2]. The recycled aggregate is coated with a cement paste, which reacts with CO_2 to produce $CaCO_3$ and fix CO_2.

Fig. 2 shows an overview of the fabricated general-purpose CO_2 fixation facility. The carbonation tank was filled with wet aggregate, and five dehumidifiers (specifications: dehumidification volume 30 L/day) were installed in the dehumidification tank. Highly concentrated carbon dioxide gas was supplied at a feed rate of 30 L/min, and a blower (4 m^3/min) circulated the air in the enclosed space. The thickness of the aggregate layer was 25 cm (about 0.25 m^3), and samples were taken for temperature measurement and CO_2 fixation analysis at the locations shown in **Fig. 3**. Carbonation time was 21 hours.

In this study, CO_2 content was determined by wet analysis[3] . Wet analysis is a method in which a sample is decomposed with hydrochloric acid, the generated CO_2 is introduced into a sodium hydroxide-barium chloride solution and absorbed, and the CO_2 content is determined by titration with a hydrochloric acid standard solution. For the concrete used in this experiment, the Ca content of each sample was measured by X-ray fluorescence to ensure that variations in the distribution of the originally contained lime-crushed sand ($CaCO_3$) did not affect the analytical results, and the CO_2 content was corrected so that the Ca value of the blank sample was the same as the Ca values of the other samples.

Fig. 1 Recycled aggregate used

Fig. 4 shows the temperature measurement results from the thermometer installed in the aggregate. At measurement points A and B, reaction heat of more than 10°C was generated, while no reaction heat was observed at measurement point C. The moisture content results shown in the figure also confirm that points A and B are dry, but point C is not sufficiently dry. It was

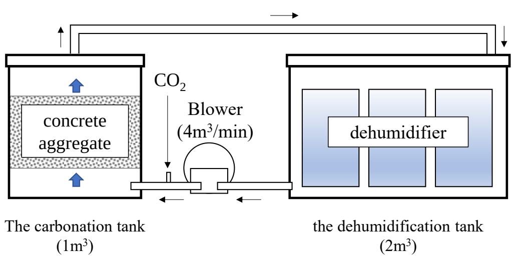


Fig. 2 Carbonation facility

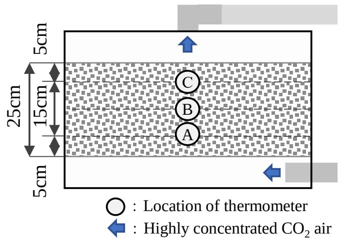


Fig. 3 Thermometer location

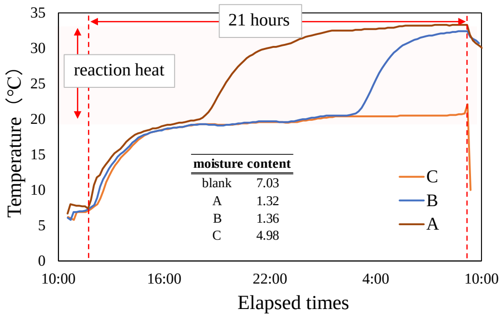


Fig. 4 Temperature measurement results

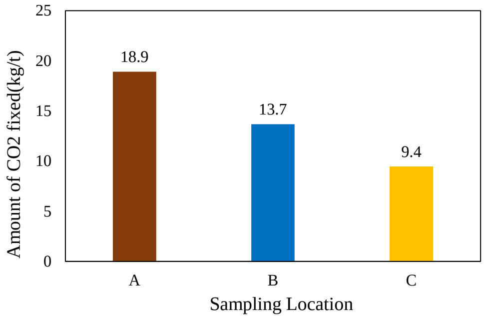


Fig. 5 CO_2 fixation results

confirmed that the generation of reaction heat started at the bottom of the charcoal oxidation tank where air is supplied.

Fig. 5 shows the results of the CO_2 fixation amount, defined as the difference in CO_2 content from the blank sample. At measurement point A, the CO_2 fixation result was 18.9 kg/t. At measurement point B, the result of CO_2 fixation amount was 13.7 kg/t, which was lower than that of measurement point A. The reason for this is that at measurement point A, the response was not as strong as at measurement point B. The reason for this is thought to be that the reaction had converged at measurement point A, but the reaction started later at measurement point B than at A, and the reaction had not yet converged. At measurement point C, the amount of CO_2 fixation was 9.4 kg/t, confirming that sufficient CO_2 fixation had not progressed.

In the future, with a view to social implementation of this facility, we plan to maximize the amount of CO_2 fixed by the entire facility and improve its production capacity by repeating wetting and drying and optimizing the CO_2 fixation time.

Keywords
Recycled bone; high concentration CO_2 gas; general purpose equipment; air cycle drying, CO_2 fixing capacity

This paper is based on results obtained from a project, JPNP21023, commissioned by the New Energy and Industrial Technology Development Organization (NEDO). We would like to thank the NEDO project personnel for their valuable comments and suggestions. We would like to express our gratitude to all parties involved.

[1] Rei Yoshino, et al: Study on Effective CO_2 Fixation Method to Recycled Aggregate, Proceedings of the Japan Concrete Institute, Vol. 45, pp. 34-39, 2023.7
[2] Akio Koyama: Resource Recovery of Residual Concrete by Granulation, Journal of the Japan Society of Material Cycles and Waste Management, Vol. 34, No. 6, 2023.
[3] Suzuki, Y. et al: Study on Evaluation of CO_2 Content by Wet Analysis and TG-DTA, Proceedings of the Japan Concrete Institute, Vol. 45, No. 1, pp. 1258-1263, 2023.7

STUDY OF CO_2 FIXATION IN RECYCLED AGGGREGATE USING POTASSIUM CARBONATE SOLUTION

Rei Yoshino[1], Yoshiyuki Suzuki[1], and Tsuyoshi Saito[2]

[1]HAZAMA ANDO CORPORATION
[2]Associate Professor, University of Tokyo
Email: iijma.rei@ad-hzm.co.jp

Abstract

Recently, CO_2 fixation technology through carbonation reaction of cement/concrete has been attracting attention. In addition, the utilization of CCUS (Carbon Capture, Utilization and Storage) is attracting interest, and effective utilization of the separated and recovered CO_2 is required. In this context, cement and concrete structures as well as waste materials are expected to serve as effective destinations for the utilization and storage of CO_2, given their vast global stock. Therefore, in this study, we aimed to improve the CO_2 fixation efficiency in cementitious recycled materials. A method of supplying CO_3^{2-} directly to the materials while maintaining a highly alkaline liquid phase was investigated using potassium carbonate (K_2CO_3) solution; changes in CO_2 fixation and hydration products were investigated.

In this study, we used recycled fine aggregate as a recycled cementitious material. **Table 1** shows the physical properties of the recycled fine aggregate used. The K_2CO_3 solution was prepared by dissolving K_2CO_3 with a purity of 99.0% or higher and a density of 2.29 g/cm^3 in distilled water at a specified amount. The CO_2 fixation amount was examined by air-dried recycled fine aggregate in the K_2CO_3 solution under the conditions shown in **Table 2**. For the analysis of CO_2 fixation amount, the recovered recycled fine aggregate after immersion in the K_2CO_3 solution was immersed in acetone to stop the hydration reaction, and then the samples were ground to less than 150 μm using a ball mill.

Table 1 Physical properties of recycled fine aggregate

Surface dry density (g/cm^3)	Water absorption (%)	Dry density (g/cm^3)	Fineness (%)	Fineness modulus (F.M.)	Amount of paste (%)
2.23	12.5	1.99	3.4	3.20	33.8

Table 2 Immersion conditions in K_2CO_3 solution

Condition	K_2CO_3 concentration (mass%)	Immersion time (h)	Solution Temp. (℃)
Effect of concentration	2.9, 23, 30, 40, 53	24, 48, 72	20
Effect of immersion time	23	3, 9, 15, 24	20

The amount of CO_2 fixation was calculated from the CO_2 content before and after immersion in K_2CO_3 solution by measuring the CO_2 content by simultaneous differential heat and thermogravimetric analysis (hereinafter referred to as TG-DTA). The CO_2 content, $Ca(OH)_2$ content, and $CaCO_3$ content are those for the sample in the dry state by vacuum drying.

Figure 1 shows the relationship between K_2CO_3 concentration and CO_2 fixation amount of recycled fine aggregate immersed in K_2CO_3 solution for 24 hours; the CO_2 fixation amount increases linearly with increasing K_2CO_3 concentration, reaching a maximum of 77 kg/t CO_2 fixation amount at 53% K_2CO_3 concentration. **Figure 2** shows the relationship between immersion time and the amount of CO_2 fixation amount at a K_2CO_3 concentration of 23%. CO_2

fixation increases with increasing immersion time, but the increase is slower from 24 to 72 hours.

Figure 3 shows the change in K_2CO_3 concentration and TG-DTA curve of the recycled fine aggregate. The recycled fine aggregate used in this study is considered to have been carbonated during the service period of the original concrete or during the production and storage of the recycled fine aggregate, because the CH around 450°C can hardly be confirmed even in the blank sample before immersion in the K_2CO_3 solution (K_2CO_3_0%). The DTA curve remains almost the same after immersion in K_2CO_3_23%, but the decrease in the weight from 550 to 900°C suggests that $CaCO_3$ is formed by carbonation reaction with other hydrates, such as Ca other than CH and perhaps C-S-H. At K_2CO_3 concentrations above 30%, an endothermic peak is observed around 540°C, but the weight decreases in the range of 550 to 900°C. It cannot be concluded that the mass loss is solely due to decarbonation of $CaCO_3$, as the exothermic peak is not clear. However, considering that the weight is almost converging within 900°C at a K_2CO_3_50%, it is highly likely that it is not decarbonation of K_2CO_3 itself, but rather a CO_2 content derived from some carbonate compound. These results show that the immersion of recycled fine aggregate in K_2CO_3 solution increased the amount of CO_2 fixation, although there was almost no CH present.

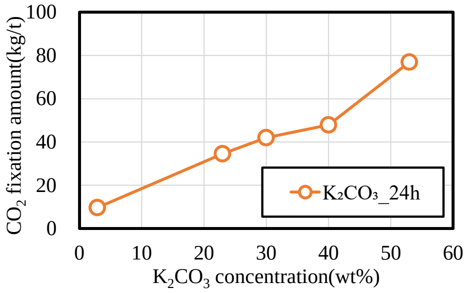


Figure 1 Relationship between K_2CO_3 concentration and CO_2 fixation amount by immersion in K_2CO_3 solution (24h)

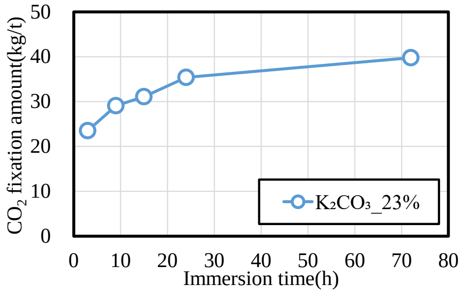


Figure2 Relationship between immersion time and CO_2 fixation amount in 23% K_2CO_3 concentration solution

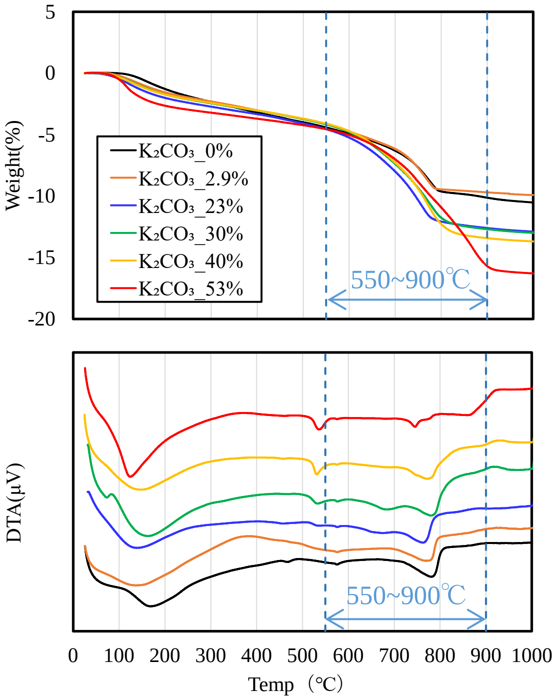


Figure3 K_2CO_3 concentration and change in TG-DTA curve

Keywords
Recycled aggregate; potassium carbonate solution; TG-DTA; carbonation

This paper is based on results obtained from a project, JPNP21023, commissioned by the New Energy and Industrial Technology Development Organization (NEDO). The authors would like to express their gratitude to all parties involved.

MECHANICAL PROPERTIES OF RECYCLED CONCRETE WITH CARBONATED RECYCLED SAND

Hong, K.C.[1], Chinzorigt, G.[2], Enkhbold, O.[2], Lee, H.S.[3], and Choi, D.U.[1]

[1]CECM Co Ltd, Korea, Anseong-si, South Korea
[2]Dept. Architectural Engineering, Hankyong National University, Anseong-si, South Korea
[3]Dongbu ENT, Anseong-si, South Korea
Email: ghdrudcks96@hknu.ac.kr

Abstract

The effect of using carbonated recycled sand was investigated. Natural sand was replaced by recycled sand by 15%, 30%, or 50% while natural coarse aggregate was 100% replaced by recycled coarse aggregate. Three different sands were used: Natural sand (NS), recycled sand (RS) and CO_2 treated recycled sand (CRS). CO_2 treatment of recycled sand improved physical properties such as absorption and density and the strength of recycled concrete. Compressive strength reduction was 11% or less at 100% recycled coarse aggregate and 15%–50% carbonated recycled sand replacement. CO_2 treatment of recycled sand resulted in up to 15% improved compressive strength of recycled concrete. Stress-strain relationship of recycled concrete was also investigated.

1. Introduction

Use of recycled sand (RS) in concrete is beneficial technology for its environmental benefits such as circularity of waste resources. However, the use of RS in recycled concrete (RC) often leads to reduced mechanical performance. CO_2 treatment for accelerated carbonation can be an efficient method to improve the properties of RC. Use of of CO_2-treated RS (CRS) on the mechanical behavior of RC was investigated, with focus on strength development and stress-strain relationship of RC.

2. Materials and test method

Type I ordinary Portland cement was used. 100% recycled coarse aggregates (RCA) replaced natural coarse aggregates (NCA) in all mixes except Control. Both natural sand (NS)/recycled sand (RS) were used. Table 1 shows the physical properties of all aggregates including carbonated RS (CRS). RS was carbonated in an environmental chamber with T = 20ºC, R.H. = 60%, CO_2 = 5% for 72 hours. Table 2 shows mix proportions. In Table 2, Control uses natural aggregates. In Series 1, RS replaces NS by 0, 15, 30, 50% while 100% RCA is used. In Series 2, CRS replaces NS by 15, 30, 50%. Water-to-cement ratio was 0.5 in all mixes. Aggregates were in SSD condition at the time of batching. ϕ100 × 200 mm cylindrical specimens were used for compressive strength test performed at 7d, 28d and split tensile strength test performed at 28d. 100 x 100 x 400 mm prisms were used to determine 28d flexural strength. Test specimens were demolded one day after casting and cured in water until test days. Three replicate specimens were used. Stress-strain relationship was determined using the compressive test specimens.

Table 1 Physical properties of aggregates.

Type	Absorption (%)	SSD Density (kg/m^3)	Crushing value (%)	Adhered mortar (%)	FM	% passing 0.08-mm sieve
NCA	0.48	2 690	17.4	-	7.3	-
RCA	3.84	2 430	21.2	24.2	7.4	-
NS	0.60	2 630	-	-	3.1	3.8
RS	5.52	2 450	-	-	3.2	7.4
CRS	3.92	2 510	-	-	3.2	7.4

Table 2 Mix proportion per m^3.

Series	Index	C kg	W kg	NCA kg	RCA kg	NS kg	RS kg	CRS kg	SP kg	Slump cm	Air %
1	Control	364	182	909	--	818	-	-	1.82	19.0	5.0
	RC-100-0				821	818	-	-		20.0	4.5
	RC-100-15					696	114	-		20.0	3.5
	RC-100-30					573	229	-		21.0	4.5
	RC-100-50					409	381	-		19.5	4.5
2	RC-100-15C					695	-	116		20.0	5.0
	RC-100-30C					573	-	232		21.0	4.5
	RC-100-50C					409	-	387		20.0	4.5

3. Test results

3.1 Compressive strength

- 7d: Average compressive strength of Control specimens was 29.9 MPa, while that of specimens with 100% RCA+15% RS was 27.3 MPa. It was 26.1 MPa and 28.5 MPa, respectively, for specimens with 100% RCA+30% CRS and 100% RCA+50% CRS. 7d compressive strength of specimens with 100% RCA+15, 30, 50% CRS was 110%, 96.3%, and 115% of corresponding specimens with RS, respectively.
- 28d: Average compressive strength of the Control specimens was 37.9 MPa, while that of specimens with 100% RCA+15, 30, 50% CRS was 35.8 MPa, 34.5 MPa, and 34.1 MPa, respectively. 28d compressive strength of specimens with 100% RCA+15, 30, 50% CRS was 101%, 113%, and 99.4% of corresponding specimens with RS, respectively.

3.2 Stress-strain relationship

Stress-strain behavior of Control and RC was determined using 1 200-kN capacity Instron 4495 UTM under displacement control mode at 0.1 mm/min ramp rate with results shown in Fig. 1. The stress-strain plots in Fig. 1 show that peak is lower for the RC than that of Control, stiffness is reduced for RC, and the strain at the peak tends to be delayed. The area under the stress-strain plot of RCs is typically smaller than that that of Control.

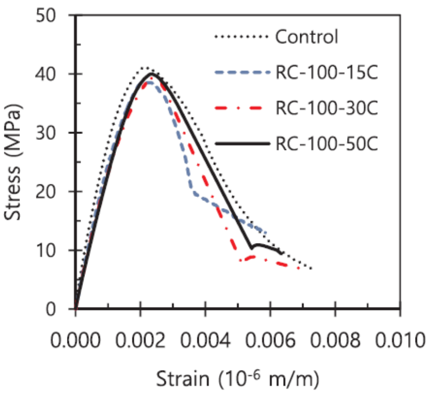


Fig. 1 Stress-vs-strain plot

4. Conclusion

The CO_2 treatment of recycled sand reduced absorption rate and increased density of RS. 28d compressive strength of RC with 100% RCA+15~50% CRS ranged between 90.0~94.5% of

Control and 99.4~113% of RC with 100% RCA+15, 30, 50% RS. Stress-strain behavior of RCs was similar to that of Control.

Keywords

Recycled sand; carbonation; recycled concrete; strength; stress-strain relationship

Acknowledgement

This work was supported by the Technology Innovation Program (20020839, Development of International Standard for low carbon recycled aggregate technology) funded by the Ministry of Trade Industry & Energy (MOTIE, Korea).

CHARACTERIZATION OF THE INTERFACIAL TRANSITION ZONE BETWEEN CARBONATED RECYCLED AGGREGATES AND NEW CEMENTITIOUS MATRIX: THE INTERACTIONS BETWEEN DIFFERENT CEMENTS AND CARBONATION PRODUCTS

Masunaga, K.M.[1], and Iyoda, T.[1]

[1]Shibaura Institute of Technology
Email: na22504@shibaura-it.ac.jp

Abstract

In order to reduce CO_2 emissions and enhance the circularity in the cement and concrete industry, the carbonation technology applied to concrete recycling products has been widely investigated. Also, reducing the clinker content by using, for example, Ground Granulated Blast Furnace Slag (GGBS) blended cement is an important factor to achieve concrete sustainability. When applied in powder cementitious materials waste, the carbonation products are Calcium Carbonate (CC) and decarbonated silica gel, a potential supplementary cementitious material that may influence the hydration of cement and the microstructure development, by physical filling and chemical effects. When carbonation is applied to hardened concrete recycling waste, such as recycled aggregates, the above described interaction between the carbonation products and the cement occurs in the interfacial transition zone (ITZ) and the possible changes in the exiting carbonated microstructure should be considered. In this context, the focus of this investigation was the characterization of the ITZ between carbonated recycled aggregates and new cementitious matrix, with different GGBS replacement ratios, evaluating the aggregate-cement interaction and its effect on concrete's mechanical performance.

Recycled aggregate concrete was produced replacing 100% of natural aggregates (NAgg) by saturated surface dry state coarse and fine non-carbonated recycled aggregates (RA) and carbonated recycled aggregates (CRA). The carbonation was conducted in a chamber 5% CO_2 concentration, 60% relative humidity and 20°C, and the carbonation was confirmed by the pH reduction, spraying phenolphthalein solution, by the increase in density and decrease in water absorption of the carbonated aggregates. The mix design is shown on the Table 1.

Table 1. Concrete's mix design.

Identification	W/B		s/a	Binder	Coarse and Fine Aggregates
OPC	50%		48%	Ordinary Portland Cement (OPC)	Natural Aggregates (NAgg) Recycled Aggregates (RA) Carbonated Recycled Aggregates (CRA)
B30				70% OPC + 30% GGBS	
B50				50% OPC + 50% GGBS	
B70				30% OPC + 70% GGBS	

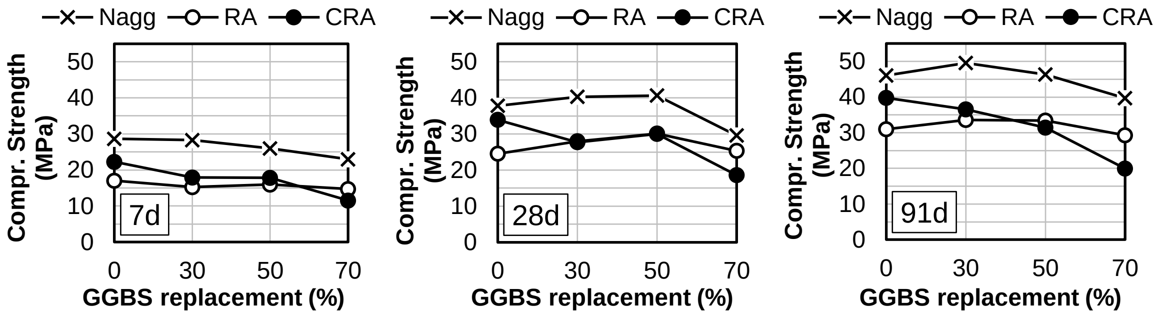


Figure 1. Influence of GGBS replacement ratio in the compressive strength (7 days, 28 days and 91 days curing) of concrete with Nagg, RA and CRA.

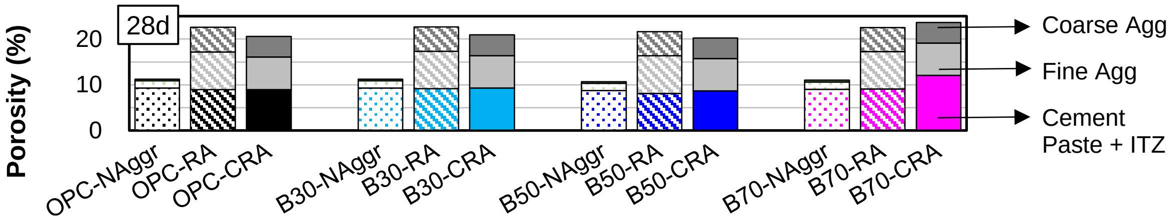


Figure 2. Porosity at 28 days of OPC and GGBS blended cement concretes, with Nagg, RA and CRA.

The compressive strength results of concrete are shown in the Figure 1. Compared to natural aggregates, the use of RA and CRA reduced the mechanical performance for all curing ages. This is caused by the attached mortar, resulting in RA and CRA having lower density and higher water absorption. As a consequence, recycled concrete's total porosity showed higher values (Figure 2), affecting the mechanical performance.

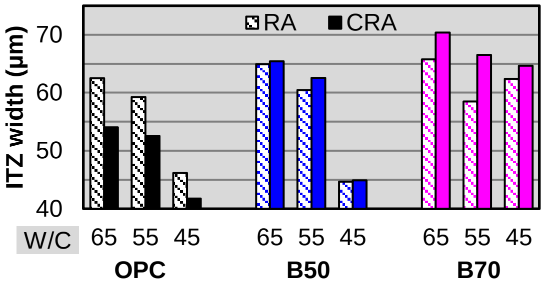


Figure 3. ITZ width between coarse RA/CRA and pastes different GGBS replacement ratios.

Comparing the effect of carbonation of aggregates on the strength, results were directly influenced by the GGBS replacement ratio. For no GGBS cement (OPC), using CRA resulted in higher strength compared to RA. But, for high GGBS replacement (B70), using CRA resulted in lower strength. So, even for the same aggregate, increasing GGBS replacement, decreased the strength of concrete with CRA, compared to RA. This suggests that carbonation treatment may have changed the surface of recycled aggregates in a way that affected negatively the aggregate-cement interaction and, consequently, the ITZ. This can be confirmed from the difference between B70-RA and B70-CRA concrete porosity's distribution (Figure 2). Even though the carbonation treatment reduced the porosity of both coarse and fine CRA, the total porosity increased for B70-CRA. This was caused by an increase in the (Cement Paste + ITZ) porosity, suggesting that aggregate-cement interaction occurred and that CC from carbonation may not be as stable as CC in natural limestone aggregate.

Then, the ITZ width between coarse RA or CRA (from demolished concrete with W/C of 60%, 55% and 45%) and cement pastes with 0%, 50% and 70% GGBS replacement (OPC, B50 and B70, respectively) was calculated, considering linear hardness measurements smaller than 80 HV, from Vickers Hardness test (Figure 3). High W/C of the demolished concrete increased the ITZ width because of the initial higher porosity of the recycled aggregates, but the changes in ITZ width with carbonation treatment was largely influenced by the GGBS content the new cementitious matrix. There was a decrease in the ITZ width between all 3 types of CRA and the OPC paste. However, an increase in the ITZ width between all 3 types of CRA and the B70 paste was observed. For CRA and B50 pastes, the ITZ width showed an intermediate behavior. These differences in ITZ depending on the cement type, even for the same aggregate, are an evidence of aggregate-cement chemical interaction, i.e. the interaction between CC and silica gel from carbonation treatment in the CRA's surface and high GGBS replacement cement pastes, which may have resulted in increasing the ITZ width. This explains the porosity increase (Figure 2) and the decrease in compressive strength (Figure 1), as the ITZ is considered the weakest part of concrete, where microcracks often initiate and propagate.

Keywords

Accelerated carbonation; recycled aggregate; ground granulated blast furnace slag; interfacial transition zone; aggregate-cement interaction

FULLY RECYCLED AGGREGATE CONCRETE (RAC) WITH RECYCLED CONCRETE POWDER (RCP): ENHANCEMENT USING CO_2 MINERALIZATION

Ligang Peng[1, 2], Yuxi Zhao[1], Peiliang Shen[2], Chi-Sun Poon[2]

[1]Institute of Structural Engineering, Zhejiang University, Hangzhou, 310058, Zhejiang, China
[2] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hong Kong, China
Email: yxzhao@zju.edu.cn

Abstract

Efficient utilization of concrete waste to produce recycled aggregate concrete (RAC) offers a significant technical approach towards green and sustainable development within the construction industry. However, the inferior workability, mechanical and durability properties of RAC along with the unsatisfactory reduction in CO_2 emissions have limited its application prospect in engineering practice. In this study, CO_2 mineralization technology was harnessed to enhance and refine recycled concrete aggregate (RCA) and recycled concrete powder (RCP). Carbonated RCA was utilized as a complete alternative for natural sand and gravel aggregates, while carbonated RCP was partially integrated in lieu of ordinary Portland cement to fabricate carbonated recycled aggregate concrete incorporating carbonated RCP (CRCP-CRAC). This innovative concrete was then subjected to a rigorous comparison with NAC and concrete solely utilizing RCA and RCP. A meticulous analysis was carried out to delve into the fundamental characteristics of RCA and RCP, both prior to and following the accelerated carbonation treatment. Furthermore, the mechanical properties, durability, and electrochemical behaviour of steel corrosion embedded within the CRCF-CRAC were thoroughly investigated. Through this comprehensive study, the evolution patterns of material properties and steel corrosion electrochemistry within the CRCP-CRAC were thoroughly grasped. The results show that the physical and mechanical properties of RCA were enhanced after CO_2 mineralization due to the reduction of porosity and the refinement of pore size, and the pore structure of RCP was refined as well along with a higher proportion of gel pores. Furthermore, CRCP-RAC exhibits satisfactory performance in compressive strength, water permeability resistance, chloride penetration resistance, and steel corrosion resistance due to the enhancement in matrix microstructure and aggregate-matrix interface. Using life cycle assessment with a system boundary from cradle to gate, the feasibility of utilizing this low-carbon and sustainable concrete was preliminarily validated from the perspectives of environmental and economic benefits.

Keywords
Recycled concrete aggregate; recycled concrete powder; CO_2 mineralization; steel corrosion; life cycle assessment

ACTIVATED BIOCHAR SATURATED WITH CO_2 AS INTERNAL CARBONATION CURING ACTIVATOR FOR ENHANCING MECHANICAL PROPERTY OF CEMENT PASTE

Renming Wu[1], Shipeng Zhang[1], Chi Sun Poon[1]

[1]Department of Civil and Environmental Engineering, The Hong Kong Polytechnic
University, Hung Hom, Kowloon, Hong Kong
Email: ren-ming.wu@connect.polyu.hk

Abstract

Incorporation of carbon-negative biochar into cementitious materials has been recognized as one of the most promising ways to achieve global carbon neutrality. In this study, we evaluated the carbon dioxide adsorption performance of biochar with different activation methods and investigated the effect of biochar saturated with CO_2 as internal carbonation curing supplier on the properties of cement paste. Two activation methods, chemical activation and physical activation, were employed to enhance the CO_2 adsorption capacity and sequence constraint on CO_2. Higher CO_2 adsorption was achieved in activated biochars compared to pristine biochar, and more CO_2 was retained in pores of activated biochars after being exposure to the air atmosphere. Improved compressive strength of specimens with adding CO_2-saturated biochars was observed, which can be attributed to the hydration acceleration resulted by filler and nucleation effect. This approach can effectively reduce the negative effect of adding high amount (>5%) of biochar on the mechanical property of cement paste, further helping achieve global carbon neutrality.

Keywords

Biochar; activation method; internal carbonation curing; cement paste

Session B2

CO_2 Activation of Fresh and Hardening Concrete

HIGH-TEMPERATURE PERFORMANCE OF SCMS BLENDED CEMENTITIOUS MATERIALS SUBJECT TO CO_2 CURING

Guo, M.Z.[1], **Song, Q.F.**[2], **and Ling, T.C.**[3]

[1]School of Civil Engineering, Shaoxing University, Shaoxing, Zhejiang Province 312000, China
[2]College of Civil Engineering, Hunan University, Changsha,
Hunan 410082, People's Republic of China
[3]School of Mechanics and Construction Engineering, Jinan University, Guangzhou,
Guangdong Province 510632, China
Email: guo.mz.guo@connect.polyu.hk

Abstract

Supplementary cementitious materials (SCMs) are increasingly used to partially replace cement in the construction industry due to the expected reduction in CO_2 emissions and improvement in the mechanical and durability performance. Meanwhile, the application of CO_2 curing in cement-based materials is garnering growing attention against a backdrop of current green and sustainable development trends However, the influence of CO_2 curing on the high-temperature properties of the SCMs blended cementitious materials remains largely unknown. This study seeks to understand the influence of CO_2 curing on the high-temperature performance of cement paste incorporated with different types of common SCMs (Fig. 1).

The results showed that for the cement paste containing SCMs, pure CO_2 curing reduced the 28-day compressive strength by approximately 10% compared with standard curing. Further standard curing of 2 days promoted the hydration of the unreacted cement and SCMs in the CO_2 cured specimens, compensating for the reduced compressive strength of the pure carbonated SCMs cement paste (Fig. 2).

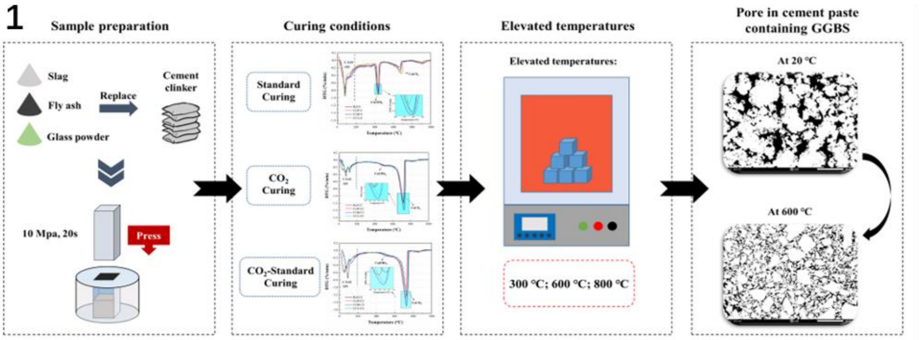
Due to the simultaneous generation of $CaCO_3$ and hydration products, the samples carbonated for 2 days and then standard cured for 26 days (CS curing) had the best high-temperature resistance, which was reflected by higher compressive strength, lower water absorption and decreased water sorptivity (Fig. 3). BSE imaged revealed that the porosity of the CS cured ground granulated blast furnace slag (GGBS)-incorporated cement paste decreased by 7.93% after exposure to 600 °C, corresponding to a 57% decrease in water sorptivity due to the transformation of large loose pores into more compact small pores (Fig. 4).

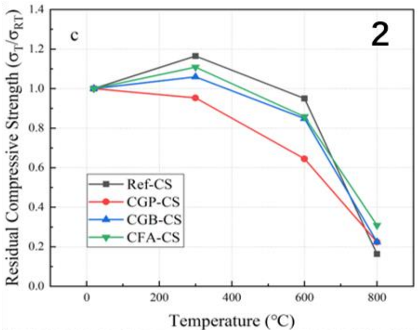
The DTG and XRD analysis demonstrated that Glass powder (GP) was inert in the cement matrix with no obvious pozzolanic reaction and negligible improvement of carbonation degree, leading to the lowest high-temperature performance; whereas, GGBS and fly as (FA) increased the generation of $CaCO_3$ by 19.05% and 15.08%, respectively (Fig. 5). Samples after two days of carbonation were completely carbonated, and subsequent standard curing further promoted the formation of $CaCO_3$, which was present as calcite and remained stable after exposure to 600 °C.

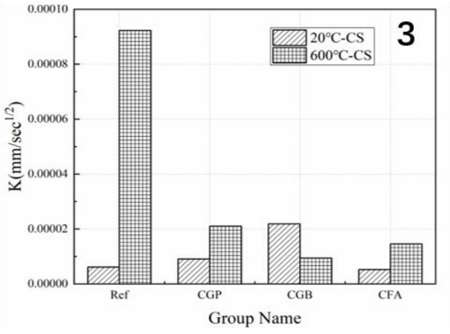
Findings from this study well demonstrate that the combined use of early CO_2 curing and subsequent standard curing to treat the SCMs blended binders has great potential to reduce carbon footprint and improve elevated temperature resistance.

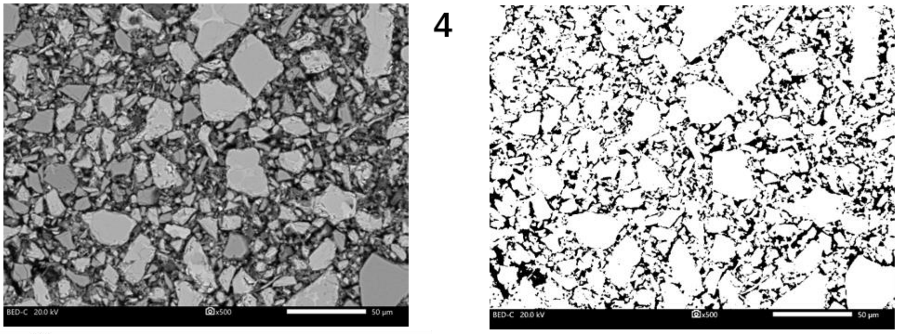
Keywords

Supplementary cementitious materials; CO_2 curing; steel slag; high-temperature performance; microstructure









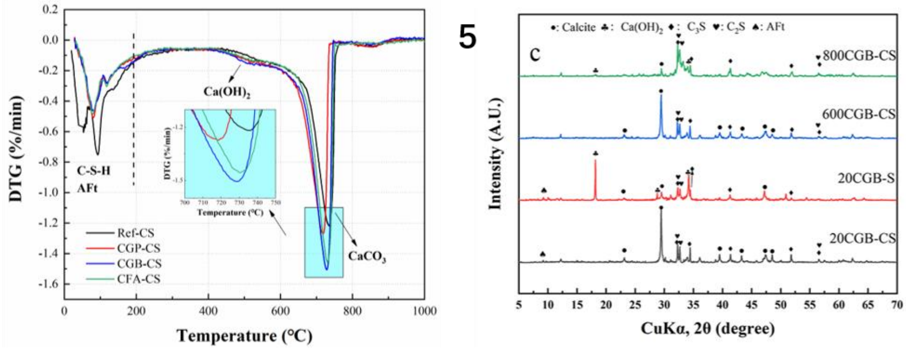


Fig. 1 Overview of the experimental design; **Fig. 2** Residual compressive ratio of the SCMs blended samples with CS curing; **Fig. 3** Comparison of water sorptivity of CS cured SCMs cement pastes at 20 and 600 ℃; **Fig. 4** BSE images of CS cured CGB samples; **Fig. 5** DTG curves and XRD patterns of CS cured GGB groups

Acknowledgment

The authors would like to acknowledge the funding support from the National Natural Science Foundation of China (52078202) and the Natural Science Foundation of Jiangsu Province (BK20221503).

DEVELOPMENT OF ADVANCED CARBONATION TECHNOLOGIES FOR CONCRETE AND SOLID WASTE

Peiliang Shen[1], Yi Jiang[1], Zihan Ma[1], Yingliang Zhao[1], Chi Sun Poon[1]

[1] Department of Civil and Environmental Engineering & Research Centre for Resources
Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hung Hom,
Hong Kong, China
Email: peiliang.shen@polyu.edu.hk; cecspoon@polyu.edu.hk

Abstract

The whole world is facing a critical environmental challenge due to the extensive release of CO_2 and the construction sector plays an important role. Meanwhile, with accelerated industrialization and urbanization, huge amounts of concrete waste are generated. Carbonation of waste concrete and its reuse can convert concrete into carbon sink due to the rapid chemical reaction between CO_2 and hydration products of cement, which could not only address the CO_2 release issue but also benefit to environmental protection and effective utilization of resources. Our recent research has shown that this approach is applicable to promote recycling of concrete waste by accelerated carbonation.

Our works focused on the application of different advanced carbonation technologies to achieve the total recycling of concrete and solid wastes. Advanced technologies including dry carbonation, pressurized semi-dry carbonation, flow-through semi-wet carbonation and wet carbonation methods were developed to treat concrete wastes with different size. Recent results indicated that the waste concrete can be turn into low-carbon products using different carbonation technologies, including carbonated coarse aggregate, carbonated fine aggregate and high-valued products (nano-silica gel, micro-fiber and superfine powders), aiming to achieving the total recycling of concrete waste by using accelerated carbonation. The accelerated carbonation processes can not only enhance the properties and values of the recycled concrete waste, but also sequestrate a large amount of CO_2. Furthermore, the performance of new concrete incorporating these carbonation products can be obviously improved. Therefore, the provided carbonation technologies of concrete waste can make great contribution to the waste recycling and reduction of CO2 emission of construction sector.

Keywords

Concrete waste; carbonation technologies; recycling; mechanisms

THE REACTION MECHANISMS OF CEMENT INCORPORATING FA AND GGBS DURING CO_2 CURING AND FURTHER HYDRATION

Pingping He[1, 2], **Caijun Shi**[2]

[1]School of Traffic and Transportation Engineering, Changsha University of Science and Technology, Changsha 410119, P. R. China
[2]Key Laboratory for Green & Advanced Civil Engineering Materials and Application Technology of Hunan Province, College of Civil Engineering, Hunan University, Changsha 410082, P. R. China

Abstract

In this study, the reaction mechanisms of cement incorporating FA and GGBS during CO_2 curing and further water curing were investigated. In CO_2-cured cement, the matrix and rim encompassing the cement particles contained both calcium carbonate and silica-rich gel. But the former was mainly in the matrix, while the latter was mainly in the rim. Besides, calcium carbonate crystals were surrounded by amorphous layer. In FA/GGBS-blended cement, FA or GGBS remained unreacted and calcium carbonate precipitated on FA and GGBS. The calcite precipitated on FA particles were hexagonal plate, while that on GGBS particles and cement particles were small rhombohedral crystals. After 28d of further hydration, cement was covered by cluster-shaped C-S-H gel. Calcium carbonate reacted with aluminum phase and formed Mc. Ettringite was also detected and mainly located in pores. The calcite content increased due to the further carbonation. After 90d of further hydration, the crystal size of calcite and ettringite increased. The content of calcite decreased due to the continuous reaction with aluminum phase, while more ettringite and portlandite was formed. Meanwhile, more C-S-H was formed and transformed to net shape. After further water curing, hexagonal calcite on FA particles was not observed. By contrary, some products surrounded FA particles due to the reaction between FA and calcite, forming Hc and Mc. The content of calcite decreased continuously during further hydration. For GGBS particles, they were also covered by calcite crystals and the surface of slag was not connected with matrix after CO_2 curing. After further hydration, slag was tightly connected with matrix as the hydration and reaction with calcite.

Keywords
CO_2 curing; reaction mechanisms; further hydration; microstructure

EFFECT OF PRE-TREATMENT METHOD ON ACCELERATED CARBONATION OF HARDENED CEMENT PASTE

Luge Cheng[1], **Ippei Maruyama**[1], **and Ryo Kurihara**[1]

[1]Department of Architecture, Graduate School of Engineering, The University of Tokyo,
Tokyo 113-8656, Japan
Email: chengluge@g.ecc.u-tokyo.ac.jp

Abstract

To predict the service life or evaluate the soundness of reinforcement structures, it is imperative to determine the resistance of concrete against carbonation. Accelerating the carbonation test with high CO_2 concentration is necessary to evaluate the carbonation resistance of cementitious materials to shorten the duration of experimental procedures in a laboratory setting.

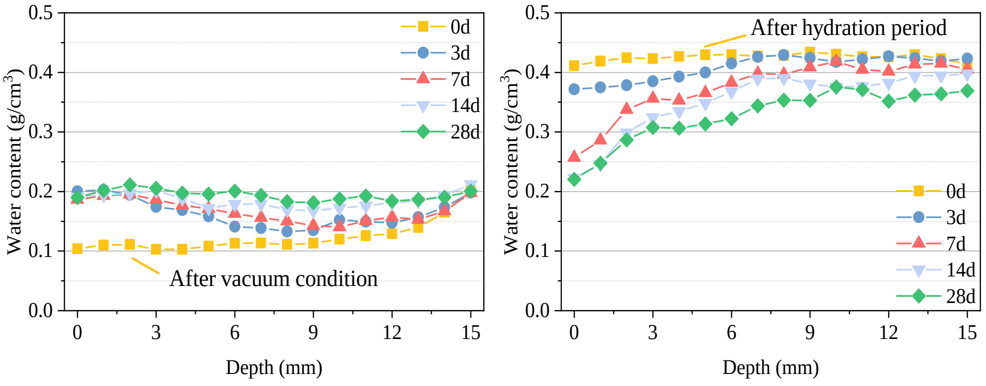
For the accelerated carbonation method proposed in the JIS standard [1] and ISO 1920-12 [2], samples are preconditioned to dry under the optimum RH for the carbonation reaction, reported to be approximately 60%, to remove excess water that obstructs CO_2 penetration through the pore path. However, based on our previous research [3], the plugging effect of water on the carbonation process occurs under accelerated carbonation conditions with such pre-drying procedures, which is a significant difference between natural carbonation and accelerated carbonation. Therefore, it is necessary to propose a proper accelerated carbonation method with a different pretreatment to avoid such plugging effect. This study proposes a new pretreatment method before the accelerated carbonation period: after sufficient hydration, drying the hardened cement paste (hcp) samples under vacuum conditions for three weeks before drying them under 60%RH for 28 day. With the preconditioning with (D) and without (V) vacuum pretreatment, the water content distribution before the carbonation period and the resultant carbonation rate coefficient under 5% and 1% CO_2 concentrations were compared and assessed.

The water content distribution was measured during the pre-drying period by the relaxometry measurements, which was conducted using single-sided 1H-NMR measurements with the Carr–Purcell–Meiboom–Gill (CPMG) technique, shown in Figure 1. The initial water content after vacuum pretreatment is approximately 0.1 g/cm_3 in D, which is four times lower than the initial value of approximately 0.42 g/cm^3 in V. For D with vacuum pretreatment, the water content increases from the surface over time, after 28 days, the water content along the penetration depth becomes constant, stabilizing at around 0.2 g/cm^3. For V without vacuum pretreatment, a continuous decline in water content is observed along the penetration depth, with a more pronounced reduction near the surface due to the greater evaporation. The final water content is much higher by V pretreatment than that by D pretreatment, ranging from 0.23 to 0.37 g/cm^3.

During the carbonation period, **Error! Reference source not found.** illustrates carbonation depth of the samples over the carbonation duration and the corresponding carbonation rate coefficient (Kc) under different CO_2 concentrations. The carbonation coefficient with D pretreatment is much higher than that with V pretreatment under 5% CO_2 concentration, while slightly higher than that with V under 1% CO_2 concentration.

These results reveal that the vacuum pretreatment before the drying process significantly affects the water content distribution in hcp. Consequently, the lower and stable water content achieved through vacuum pretreatment facilitates the penetration of CO_2 during the carbonation period,

thereby avoiding the plugging effect and advancing the carbonation process, especially under a higher CO_2 concentration of 5%. Therefore, implementing vacuum pretreatment as a preparatory step can enhance the efficiency and effectiveness of accelerated carbonation tests, providing a more accurate assessment of carbonation resistance that better represents natural carbonation conditions.



(a) D (b) V

Fig. 1 Volumetric water content distribution during the pre-drying period with (a) and without vacuum pretreatment (b)

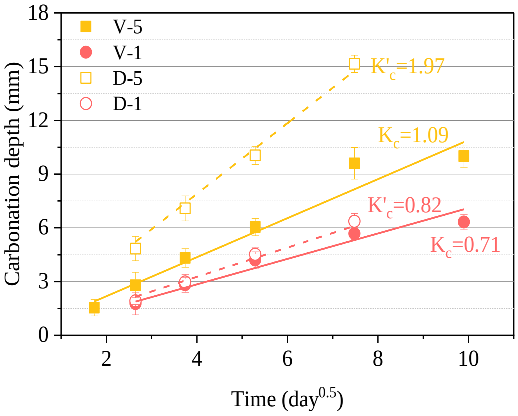


Fig. 2 Carbonation depth determined by image analysis with different pre-treatment conditions under 5% and 1% CO_2 concentration

Keywords

Accelerated carbonation; pretreatment; plugging effect; water content; hardened cement paste

Acknowledgment

This study was based on the results obtained from a project (JPNP21023) commissioned by the New Energy and Industrial Technology Development Organization (NEDO).

References

[1] J. Japanese Industrial Standard Committee: Tokyo, *JIS A 1153; 2012, Method of Accelerated Carbonation Test for Concrete*. 2012, p. 2012.

[2] I. O. for Standardization, *ISO 1920-12-2015Testing of concrete. Part 12: Determination of the carbonation resistance of concrete*. 2015.

[3] L. Cheng, R. Kurihara, T. Ohkubo, R. Kitagaki, A. Teramoto, and I. Maruyama, “Plugging effect of fine pore water in OPC and LC 3 paste during accelerated carbonation monitored via single-sided nuclear magnetic resonance spectroscopy,” pp. 1–50.

MICROWAVE-CO_2 COMBINED CURING TECHNOLOGY FOR LOW-CARBON DEVELOPMENT OF ULTRA-HIGH-PERFORMANCE CONCRETE (UHPC)

Yuan Feng[1,2], **Rui Yu**[1,2], **Jingjing Zhang**[1,3]

[1]State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan, China
[2]International School of Materials Science and Engineering, Wuhan University of Technology, Wuhan, China
[3]School of Materials Science and Engineering, Wuhan University of Technology, Wuhan, China
Email: r.yu@whut.edu.cn

Abstract

Ultra-high-performance concrete (UHPC) possesses excellent mechanical properties and durability. However, the extensive use of energy-intensive cementitious materials and prolonged curing times lead to increased carbon emissions, contradicting green and low-carbon development goals and impacting its sustainability. Numerous studies have achieved green development in UHPC production through mix optimization or the use of waste materials, but measures to reduce carbon emissions during the curing process remain limited. Therefore, in addition to changing raw material composition, improving the efficiency and reducing the carbon footprint of the curing process is also crucial. Based on this, this study proposes a microwave-CO_2 combined curing strategy. This strategy aims to rapidly enhance UHPC performance and achieve low-carbon development by stimulating UHPC's carbonation activity and incorporating negative carbon curing technology. The study also quantifies carbon emissions under different curing methods using the Life Cycle Assessment (LCA) method. Practically, the microwave treatment of UHPC promotes substantial CH generation and internal molecular thermal motion, activating carbonation activity and facilitating subsequent CO_2 absorption during carbonation curing. Additionally, combined curing enhances hydration reactions and carbonation, optimizing the macropore structure. Compared to standard curing (93.8%) and microwave curing (12.1%), the microwave-CO_2 combined curing account for only 9.56% of the total production process emissions. This ultimately demonstrates the feasibility of the combined curing system in reducing UHPC carbon emissions.

Keywords

Ultra-high performance concrete (UHPC); microwave-CO_2 combined curing; life cycle assessment (LCA); low carbon preparation and evaluation techniques

USE OF CARBONATED WATER FOR CARBON SEQUESTRATION IN OPC AND LOW-CARBON CEMENT SYSTEMS

Aswathy Rajendran[1], **Sripriya Rengaraju**[1], **and Abir Al-Tabbaa**[1]

[1]University of Cambridge, United Kingdom
Email: ar2221@cam.ac.uk

Abstract

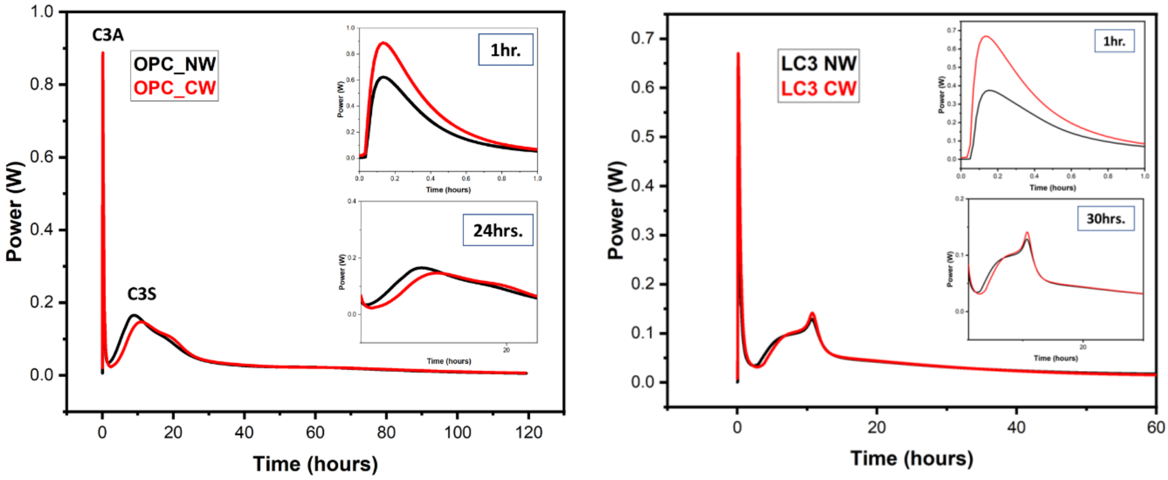
The concentration of carbon dioxide in the atmosphere has increased to 400 parts per million, reflecting a 35% rise from pre-industrial levels (Suescum-Morales et al., 2021). The industrial sectors of cement, steel, and chemicals are major contributors to this increase, accounting for 25% of emissions due to their energy consumption and industrial processes. Consequently, the construction industry, which heavily relies on these materials, has a significant responsibility in reducing these emissions. The Intergovernmental Panel on Climate Change (IPCC) has emphasized that to achieve a 43% reduction in greenhouse gas (GHG) emissions by 2030, global emissions must peak by 2025 (Ozkan & Custelcean, 2022). This means that emissions must stop growing and start declining. To meet this ambitious target, the construction industry must not only adopt low-carbon materials but also integrate carbon sequestration and utilization (CCU) into construction projects, thereby transforming structures into effective carbon sinks.

A widely researched pathway in CCU for construction is facilitating mineral carbonation which entails the reaction of CO_2 with naturally occurring materials or industrial wastes to form stable carbonates through carbonation. The chemical binding of CO_2 via mineral carbonation primarily occurs in the form of calcium or magnesium carbonate, as most construction materials, such as cement, aggregates, or waste ashes, contain reactive calcium/magnesium oxide. Among the materials mentioned above, cement exhibits the highest theoretical CO_2 uptake potential for mineral carbonation at approximately 48% (BS EN 16757), while SCMs have a CO_2 uptake range of 5-20% (Gunning et al., 2010). This study investigates the mineral carbonation potential of cement-based materials, which are highly effective at capturing CO_2 and are extensively used in the construction industry, using carbonated water.

The study investigated four cement mortar systems: (i) Ordinary Portland Cement (OPC), (ii) a blend with 50% slag, (iii) a blend with 80% slag, and (iv) Limestone Calcined Clay Cement (LC^3) with 50% calcined clay. A consistent water-to-binder ratio of 0.5 was maintained across all mixes, and a mid-range polycarboxylate ether (PCE) superplasticizer was used to ensure workable mixes with a flow range of 160-200mm, as measured by flow table tests. Each cement-type mortar was prepared using both normal and carbonated water and then cured for 7 days before evaluating their micro and macro properties.

The results revealed a significant interaction between carbonate ions and the hydration process, as evidenced by the heat of hydration curves (Figure 1). This interaction suggests potential binding with ettringite and carboaluminate phases. In the OPC and slag systems, the primary observation was a variation in the initial tricalcium aluminate (C_3A) peak and retardation of C_3S hydration. However, in the LC^3 system, there was no delay in tricalcium silicate (C_3S) peak and a more pronounced interaction due to more carboaluminate phases in them.

The use of carbonated water led to a slight reduction in workability across all mixes. Despite this, the compressive strength remained largely unaffected, indicating that the dosage level of carbonated water used in this study—0.3% by weight of cement—was insufficient to cause substantial CO_2 binding. Nevertheless, a trend of decreasing permeability was observed in all mortars when carbonated water was used. This reduction in permeability may be attributed to the formation of carbonated products from ettringite and carboaluminate phases, which typically have decreased volumes.



(a) *(b)*

Figure 1: Heat of hydration curves for (a) OPC and (b) LC^3 cement paste

The study recommends further exploration of carbon sequestration in cement systems, particularly those like LC^3 which are characterized by a higher amount of ettringite and carboaluminate formation. However, it also notes a key drawback: the pH of carbonated water, ranging from 4.5 to 5.3, lowers the initial pH of the cement paste. This impairs the hydration of calcium silicate bogue compounds, reducing the availability of carbonatable materials within the system. This limitation suggests that introducing CO_2 during the mixing process may not be the most effective strategy.

Instead, the study recommends employing mineral carbonation in cement or supplementary cementitious materials (SCMs) before their introduction into the concrete mix. This approach could better manage the complex interplay of pH and maximize the potential for CO_2 binding in the cement phases.

Keywords
Carbon sequestration; mineral carbonation; carbonated water

References

1. BS EN 16757-2022 - TC--[2024-07-14--12-00-26 PM]. (n.d.).
2. Gunning, P. J., Hills, C. D., & Carey, P. J. (2010). Accelerated carbonation treatment of industrial wastes. Waste Management, 30(6), 1081–1090. https://doi.org/10.1016/j.wasman.2010.01.005

3. Ozkan, M., & Custelcean, R. (2022). The status and prospects of materials for carbon capture technologies. MRS Bulletin, 47(4), 390–394. https://doi.org/10.1557/s43577-022-00364-9
4. Suescum-Morales, D., Cantador-Fernández, D., Ramón Jiménez, J., & María Fernández, J. (2021). Potential CO2 capture in one-coat limestone mortar modified with Mg3Al–CO3 calcined hydrotalcites using ultrafast testing technique. Chemical Engineering Journal, 415. https://doi.org/10.1016/j.cej.2021.129077

RAPID DEMOLDING OF PRECAST CEMENT MORTAR FOR CO_2 CURING: A CUSTOM MOLD DESIGN

Jin-Xin Wei[1], **Yin-Ze Hu**[2], **and Tung-Chai Ling**[1]

[1]College of Civil Engineering, Hunan University, Changsha 410082, Hunan, China
[2]Hunan Luvalley Architectural Technology Co., LTD, 410082 Changsha, China
Email: tcling@hnu.edu.cn; tcling611@yahoo.com

Abstract

Generally, the precast cement products are cured in the mold in the first 18-24 hours at a relative high temperature to ensure fast early strength gain before demolded for cure in the natural environment at plant setting. To explore the possibility of applying CO_2 curing in precast cement products, this study aims to introduce a custom mold design which consists of a hollow frame and a complementary mold that allow concrete surface exposure of 75% after partial demolding. The increase of concrete surface area can facilitate CO_2 gas penetration from external environment to concrete layer for carbonation. This custom mold is especially designed for precast cement products that could be partially demolded in relatively short time for CO_2 curing to enhance the early strength gain in plant setting.

According to the current adapted mix design of a local precast concrete factory in Changsha, the freshly prepared cement mortars (containing 20% calcium sulfualuminate cement) were prepared and poured in the 40 mm cubic custom mold, and cured for 2 h in room temperature before partially demolding for 2-h pre-drying to evaporate part of the surface moisture. Then, the samples were placed in a CO_2 curing chamber for curing for another 4 h (see Figure 1). For comparison, a same mix design cement mortar was cured in conventional mold for 24 h. After demolding, all samples were further cured in a standard curing chamber (20 ± 2 °C, RH >95%) up to 28 days for testing.

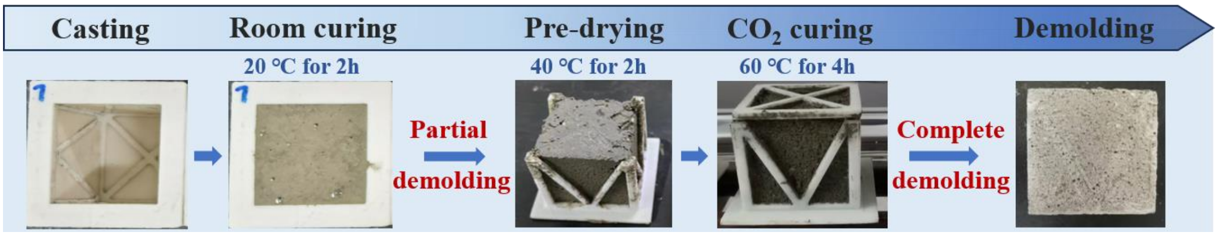


Figure 1. Cement Mortar Prepared by Custom Mold.

As can be seen from Figure 2, the compressive strength of the carbonated mortars at 8 hours can achieve 10.4 MPa, even higher than the reference mortar (9.6 MPa) tested at 24 hours. According to the TG and XRD tests results, clearly the hydration reaction within 5 mm of the sample surface was promoted by the carbonation reaction before demolding. In addition, the elevated temperature rose within the core area [1] can promote the hydration of calcium sulfoaluminate, resulting in excessive concentration of calcium ions inhibiting the reaction of C_3S [2].

After subsequent curing, the compressive strength of carbonated mortar further increased up to 28.2 MPa at 28 days. Obviously, the increment rate was relatively lower than the reference mortar due to the fact that the carbonated dense surface reduced the further hydration degree by hindering water complementary. In addition, the more blurred and looser ITZ in the carbonated mortars was the main reason for the lower compressive strength in the later stage.

In summary, high early strength obtained by CO_2 curing and custom mold has a significant advantage of shortening demolding time by 1/2-2/3 for precast cement products, which is greatly beneficial to the doubling of production efficiency and reduction of production costs. The slow increase in late strength is still sufficient to meet the engineering requirements of precast concrete factory.

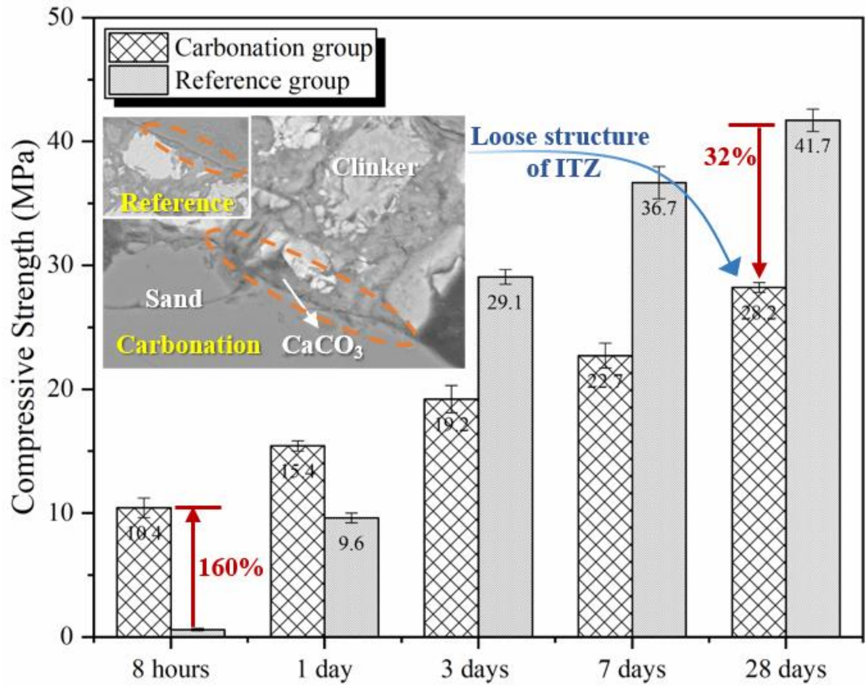


Figure 2. Compressive Strength Development and Microstructure of ITZ

Keywords
CO_2 curing; precast cement products; demolding time; compressive strength

[1] L. Xu, K. Wu, C. Rößler, P. Wang, H.M. Ludwig, Influence of curing temperatures on the hydration of calcium aluminate cement/Portland cement/calcium sulfate blends, Cem. Concr. Compos. 80 (2017) 298–306.
[2] S. Park, Y. Jeong, J. Moon, N. Lee, Hydration characteristics of calcium sulfoaluminate (CSA) cement/portland cement blended pastes, J. Build. Eng. 34 (2021) 101880.

INSIGHTS INTO THE SYNERGETIC ACTION OF HYDRATION AND CARBONATION OF PORTLAND CEMENT

Jionghuang He[1], Peiliang Shen[1], and Chi-Sun Poon[1]

[1] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University
Email: cecspoon@polyu.edu.hk

Abstract

Pretreatment plays a pivotal role in enhancing CO_2 transport for carbonation curing of cast concrete, yet the impact of initial hydration during pretreatment on subsequent carbonation remains underexplored. This study investigates how initial hydration affects carbonation and explores the synergistic action between hydration and carbonation. The phase assemblage, carbonation kinetics, microstructure and micro/macro mechanical properties of carbonated cement pastes (CCPs) with varying initial hydration durations were analyzed. The results reveled that unhydrated cement exhibited a peak carbonation rate of 0.65 W/g, which increased by approximately 83% when cement underwent an initial steam curing of 8 hours, highlighting a significant enhancement in the carbonation reactivity due to initial hydration. However, the carbonation efficiency of CCPs increased initially and then decreased as initial hydration extended. This trend was because while initial hydration enhanced the carbonation reactivity, it concurrently obstructed CO_2 transport. Furthermore, optimal initial hydration was crucial for the synergistic action of hydration and carbonation for CCPs, resulting in reduced porosity and a more homogeneous microstructure, thereby enhancing mechanical properties. These insights underscore the need to carefully consider the synergistic action of initial hydration and carbonation when designing pretreatment protocols.

Keywords

Carbonation; hydration; pretreatment; properties; curing

DAMAGE CHARACTERIZATION OF CARBONATED CEMENT PASTES WITH A GRADIENT STRUCTURE

Qinglong Qin[1], Boyang Su[1], Zihan Ma[1], Kai Cui[1], Weiwei Chen[1], Peiliang Shen[1], Qi Zhao[1], Chi Sun Poon [1]

[1]Department of Civil and Environmental Engineering & Research Centre for Resources Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hong Kong 999077, China
Email: peiliang.shen@polyu.edu.hk; cecspoon@polyu.edu.hk

Abstract

Carbonation curing concrete shows promise as a method to both reduce and sequestrate CO_2, nonetheless, it results in the formation of a gradient structure. In this paper, the damage evolution characteristics, composition, microstructure, and micromechanical properties of carbonated cement pastes with a gradient structure are investigated, aiming to delineate the relationship between the damage and inhomogeneous structure, as well as to explore its fracture mechanism. The results indicated that carbonated cement pastes display pronounced fluctuations in cracks initial stress, stress instability, and brittle damage at low strengths, which is closely related to its inhomogeneity. Moreover, carbonated cement paste is an inhomogeneous mass with a gradient structure. It displays a three-layer structure comprising an outermost layer, intermediate layer, and innermost layer. The outermost layer primarily comprises calcite, with minor amounts of aragonite and silica gel. Furthermore, its porosity, average micro-hardness, and elastic modulus are 17.03%, 58.62 HV, and 84.66 GPa, respectively. The intermediate layer consists mainly of calcite, aragonite, calcium hydroxide, C-S-H gel, and silica gel, with porosity, average micro-hardness, and elastic modulus of 21.88%, 37.21 HV, and 53.74 GPa, respectively. The innermost layer is composed of C-S-H gel, calcium hydroxide, calcite, aragonite, calcium hydroxide, and silica gel, with porosity, average micro-hardness, and elastic modulus values of 23.12%, 25.73 HV, and 58.87 GPa, respectively. The damage in cement pastes with a low degree of carbonation primarily arises from mixed tension-shear cracks, whereas in cement pastes with a high degree of carbonation, tensile cracks are the predominant cause of damage. Crucially, carbonation curing results in the formation of a “steel pipe concrete”-like structure, characterized by a strong exterior and a weak interior, which may affect the failure mechanism of the carbonated cement paste to a certain extent.

Keywords

CO_2 curing; cement pastes; gradient structure; damage characterization; failure mechanism

Acknowledgement

The authors extremely acknowledge the Global Cement and Concrete Association and China Resources Power Holdings (Shenshan) Co. Ltd for financial support.

UNVEILING THE CO_2 INTAKE IN C-S-H EFFECTED BY SURFACE PROPERTY

Li, G[1], Tao, Y.[1], and Poon, C.S.[1]

[1]Department of Civil and Environmental Engineering, and Research Centre for Resources Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong
Email: genli.li@polyu.edu.hk

Abstract

Carbon mineralization utilizing cement-based materials is widely used in building constructions, which not only enhances the physical-mechanical properties of building materials, but also promotes the carbon-negative scheme [1]. The feasibility of carbon mineralization is critically controlled by the carbonation efficiency, which relies on the CO_2 intake capacity of calcium silicate hydrate (C-S-H). Particularly, carbonation reaction occurs on the surface water film of C-S-H, which provides an incubator for the dissolution of interlayer Ca^{2+} and the formation of CO_3^{2-} species. The surface properties of C-S-H largely influences the water state in its nanometer-thick porewater and the solid-gaseous interactions so that determines its carbonation efficiency. However, current research studies fail to propose explicit mechanisms to elucidate the effect of surface properties on CO_2 adsorption and storage, especially the relation between surface properties, water saturation degree and CO_2 intake in C-S-H nanochannel. In this work, we employed the advanced grand canonic Monte Carlo (GCMC) methods to reproduce the CO_2 adsorption process in packing voids of C-S-H gel, which was mimicked by the slit pore model.

The packing voids in C-S-H grains store nanometer-thick water films under varying RH, which performs as nano-reactor for carbonation [2]. The water film thickness and state are highly depended on the surface properties of C-S-H, which is controlled by Ca/Si ratio. CO_2 adsorption and transportation in these nanometer-thick environment critically influences the carbonation efficiency. Figure 1 exhibits the total CO_2 adsorption amount in C-S-H nanochannel with respect to varying water content and Ca/Si ratio. Generally speaking, CO_2 adsorption amount increases with water content, indicating the higher water film thickness benefits the partial CO_2 solvation. The maximum CO_2 adsorption amount is reached when 4 – 5 water layers covers C-S-H surfaces. However, when the water coverage density exceeds a critical value, the CO_2 adsorption amount sharply decreases to a very low value. Under high water coverage conditions, the nanochannel is filled with liquid water, leaving little space for CO_2 diffusion, so that the adsorption efficiency is highly limited. The dissolved CO_2 and gaseous CO_2 amount is presented in Figure 1(b-c). With the increment of water coverage density, more CO_2 molecules diffuse into water film while the amount of free gaseous CO_2 gradually decreases. The obvious effect of Ca/Si ratio on CO_2 adsorption in C-S-H nanochannel is observed. Under low water coverage density (1 – 2 water layers), high Ca/Si ratio acquires high CO_2 adsorption capacity. However, the CO_2 adsorption capacity under high water coverage density becomes more complicated. Particularly, in 3 water layer systems, the Ca/Si ratio barely influences CO_2 adsorption amount. When water coverage density reaches 34.4 nm^{-2}, 43.0 nm^{-2} and 44.7 nm^{-2}, Ca/Si ratio of 1.2, 1.5 and 1.8 exhibits the strongest CO_2 adsorption capacity, respectively. These results indicate that the surface properties of C-S-H has an intricate effect not only on CO_2 adsorption, but also on H_2O state transformation.

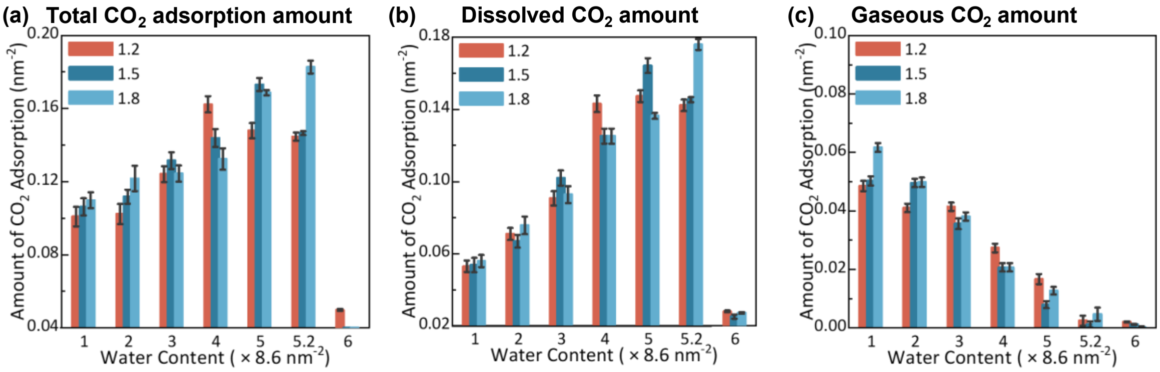


Figure 1. **CO_2 adsorption amount in 4 nm C-S-H nanopores.** (a) Total CO_2 amount in the entire channel, (b) Dissolved CO_2 amount in the interfacial water film, and (c) Gaseous CO_2 amount in the C-S-H channel.

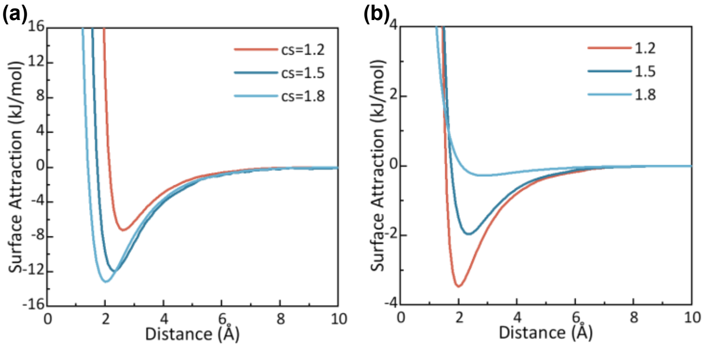


Figure 2. **Surface attraction of C-S-H with respect to (a) CO_2 and (b) H_2O.**

With the increase of Ca/Si ratio, C-S-H surface attraction obviously grows, inferring a stronger attraction to CO_2 molecules (shown in Figure 2(a)). A stronger attraction promotes the CO_2 capture on C-S-H surface. Therefore, high Ca/Si ratio enhances the balanced amount of CO_2 adsorption in dry and semi-dry conditions. With the increment of water film thickness, the solid-gas interaction sharply shrinks due to the adhesion contact is defined as a short-range interaction. At high water content, water condensation is found to benefit the CO_2 maximum intake. The higher surface attraction leads to a higher interaction range so that the hydrophilic surface with higher surface attraction to water moleucles requires a lower water film thick to active the surface condensation. In C-S-H system, high Ca/Si ratio leads to the decrease of surface attration to water molecules, thereby magnifying the water content to activate water condensation so that the maxium CO_2 intake in high Ca/Si ratio system requires higher water content (shown in Figure 2(b)).

Keywords

CO_2 adsorption, C-S-H, molecular simulation, water condensation, surface properties

References

[1] Y. Gao, Y. Jiang, Y. Tao, P. Shen, C.S. Poon, Accelerated carbonation of recycled concrete aggregate in semi-wet environments: A promising technique for CO_2 utilization, Cement and Concrete Research, 180 (2024) 107486.

[2] E. Placencia-Gómez, S.N. Kerisit, H.S. Mehta, O. Qafoku, C.J. Thompson, T.R. Graham, E.S. Ilton, J.S. Loring, Critical Water Coverage during Forsterite Carbonation in Thin Water Films: Activating Dissolution and Mass Transport, Environmental Science & Technology, 54 (2020) 6888-6899.

Session B3

CO_2 Activation of Ashes

MODIFICATION OF CARBONATION MINERALIZATION ON SUPPLIMENTARY CEMENTITIOUS MATERIALS

Gao, X.J.[1], **Chen, T.F.**[1], **and Li, L. S.**[1]

[1]School of Civil Engineering, Harbin Institute of Technology, Harbin 150090, China
Email: gaoxj@hit.edu.cn

Abstract

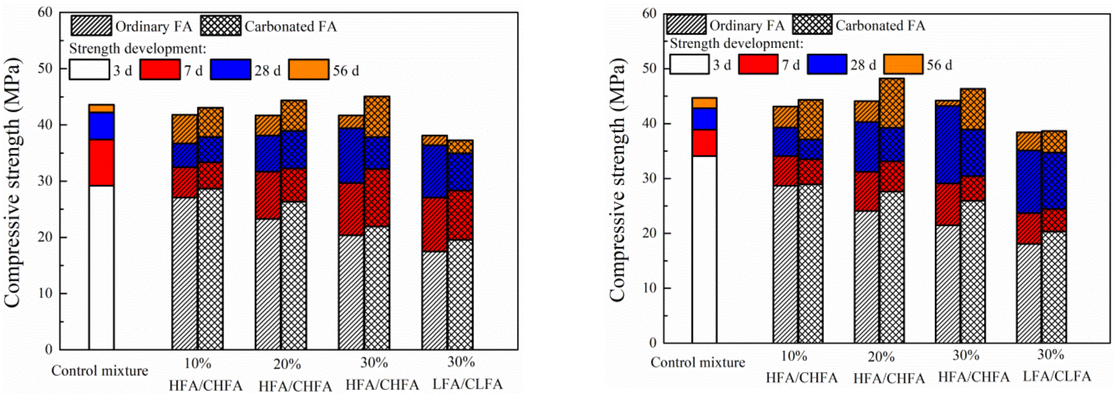
It is well known that carbonation curing can effectively sequester CO_2 and improve strength and durability of cement-based materials. Due to the low permeability, the carbonation reaction normally occurs in the outer layer of cement concrete specimens and the CO_2 capture potential could not be fully exploited. The carbonation mineralization of some industrial wastes has been also recognized as one of the most promising carbon capture and storage technologies and the carbonated material can be used as supplementary cementitious materials for cement concrete. In this study, fly ash and steel slag were pre-carbonated under different CO_2 concentrations and durations and then the carbonated fly ash and steel slag were used as mineral materials to replace Portland cement. The fly ash or steel slag added pastes and mortars were exposed to carbonation curing condition and the coupling effect of these two CO_2 capture processes were discussed based on both mechanical properties and microstructures.

The results showed that the content of free CaO and the hydration heat of fly ash were apparently reduced by the carbonation mineralization, but the pozzolanic reactivity and strength contribution of fly ash was improved. The synergy of carbonation mineralization and carbonation curing compensated for the strength loss caused by fly ash incorporation as shown in Fig.1. CO_3^{2-} on the surface of fly ash introduced by carbonation treatment competed with Cl^- for binding AFm, consequently led to the lower resistance to chloride ion penetration as shown in Fig.2. Carbonation curing mitigated this degradation by improving the pore structure of cement mortar significantly. In summary, carbonation curing can be an appropriate complement to carbonation mineralization of fly ash for performance improvement of cement-based materials and more efficient CO_2 sequestration.

The carbonation pretreatment induced the swelling deformation of steel slag particles, accompanied by the formation of calcium carbonate and amorphous silica gel as shown in Fig.3. It was found that the carbonation mineralization decreased the early-stage reactivity of steel slag due to the formation of a dense calcite barrier layer on mineralized steel slag particles. And the later-stage reactivity can be improved, being attributable to the accelerated hydration effect of silica-rich phase and nucleation effect of calcite. The synergistic effect of carbonation mineralization and carbonation curing can efficiently improve the CO_2 sequestration and compensate the early age strength reduction due to the addition of steel slag as shown in Fig.4. To facilitate the CO_2 capture and reduce the negative effect of carbonated products on hydration reaction of steel slag, porous biochar was added into aqueous steel slag slurry for carbonation mineralization. Results showed that the synergistic effect of carbonation mineralization and biochar is of great significance for enhancing the hydration reactivity of steel slag.

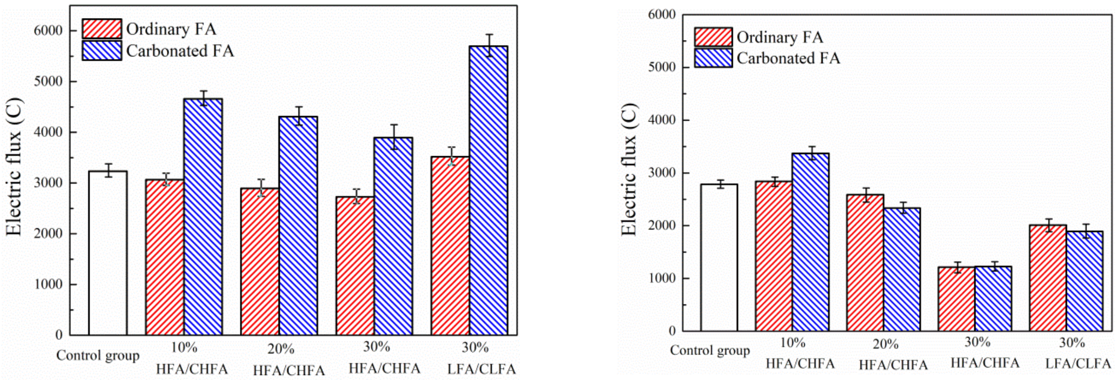
Keywords

Carbonation mineralization; carbonation curing; mineral admixture; strength; microstructure



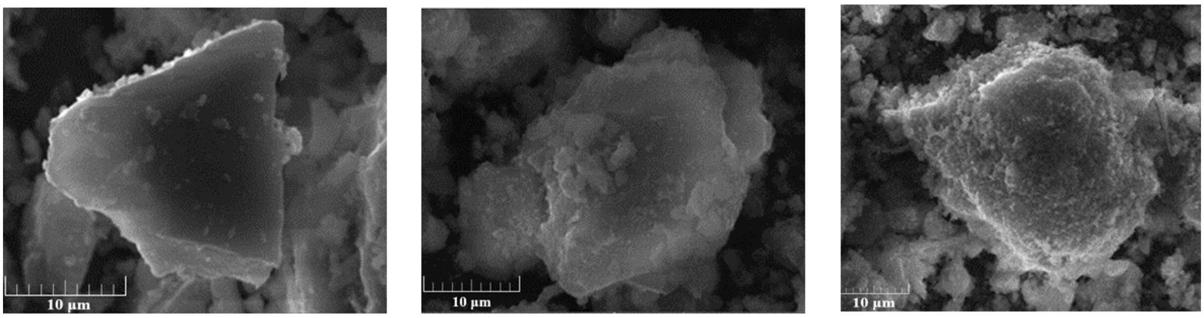
(a) Standard curing (b) Carbonation curing

Fig. 1 Compressive strength of standard and carbonation-cured cement mortar



(a) Standard curing (b) Carbonation curing

Fig. 2 Electric flux of cement mortar



(a) Uncarbonated (b) Partly carbonated (c) Fully carbonated

Fig.3 Apparent morphology of steel slag particles (SEM)

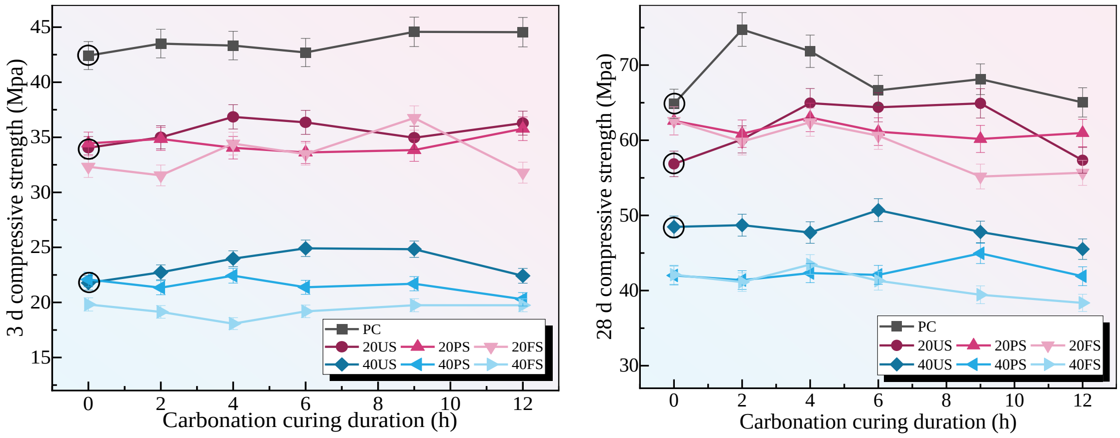


Fig.4 Compressive strength of steel slag-cement paste

EFFICIENCY OF PROCESSES FOR THE CARBONATION OF MUNICIPAL SOLID WASTES BOTTOM ASH

Courard, L.[1], **Kanjo, E.I.**[1], **Hubert, J.**[1]

[1]University of Liege, Urban and Environmental Engineering, Building Materials (Belgium)
Email: Luc.Courard@uliege.be

Abstract

Municipal solid waste incinerated bottom ash (MSWI-BA) is a byproduct from waste incineration process, which is used mainly as replacement of sand and natural aggregates in road foundations and concrete pavements. Usually, MSWI-BA undergoes a maturation treatment before being used by exposing it to the atmosphere for a period of minimum 18 weeks. This study investigates an accelerated carbonation process for MSWI-BA maturation, which aims to enhance both its engineering properties and its environmental sustainability. The results demonstrate significant improvements in shortening treatment duration without compromising physical properties such as density and water absorption while concurrently reducing greenhouse gas emissions through CO_2 sequestration.

The accelerated carbonation was carried out in CO_2 incubator maintaining a volume concentration of 12% CO_2 at atmospheric pressure for 0/20 and 0/2 sieving samples. The temperature and relative humidity inside the incubator were kept at 30±1 °C and 60±3%, respectively.

The experiment was carried using two approaches:

- Approach A: the MSWI-BA was conditioned in a climate chamber at a temperature of 30±1 °C and relative humidity of 60±3% until its weight stabilized (4 days). The moisture content of the samples decreased from 20% to 2% for both BA0-20_A and BA0-2_A before starting the accelerated carbonation.
- Approach B: the carbonation was carried out directly without any type of pre-conditioning (BA0-20_B and BA0-2_B).

Samples were collected after being carbonated for 1, 2, 4, 8, 24, 48 and 168 h where CaCO3 content, CO2 uptake, pH value and moisture content were measured. The Figure shows that the CO_2 uptake of BA0-20 rises proportionally with time, reaching its maximum value at 4 hours for approach A and 8 hours for approach B. Beyond these points, CO_2 uptake stabilizes. Notably, approach B exhibits a more pronounced increase in equivalent CO_2 uptake where it reached almost 14g/kg of MSWI-BA compared to approach A (only 10 g/kg). A similar behavior is observed for BA0-2, where the CO_2 uptake increases with time up to 8 hours for approach B, after which the results remain constant. However, with approach A, the gain in CO_2 uptake was negligible compared to approach B even after 7 days of carbonation.

The results are similar to those obtained by Lin et al (2015a), Lin et al (2015b) and Yoa et al (2022).

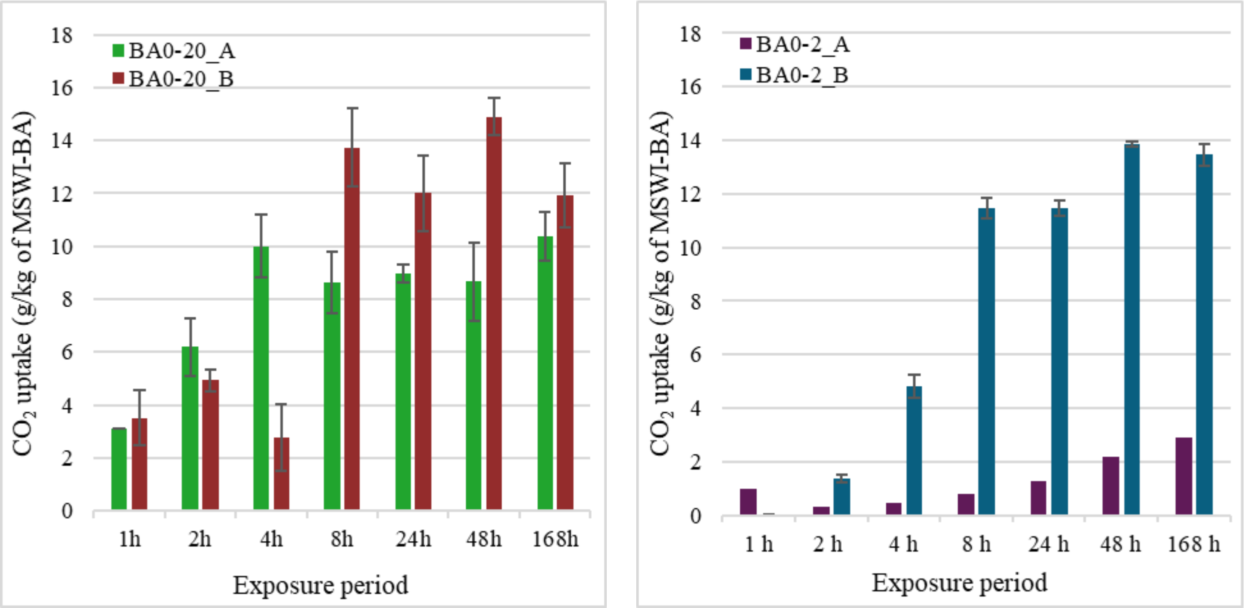


Figure 1 : CO_2 uptake of BA0-20 (left) and BA0-2 (right) after different exposure periods to accelerated carbonation

For both fractions studied, moist carbonation (approach B) was showed to be more effective regarding carbon capture and pH stabilization and more practical since no pre-conditioning was required. Regarding the type of treatment used (accelerated carbonation or traditional maturation) there was no significant improvement in the physical properties of MSWI-BA.
These findings have established that accelerated carbonation could be an effective maturation technique for MSWI-BA, thus offering new insights for its practical implementation in waste management and resource recovery strategies.

Keywords
Carbonation; incinerated municipal solid waste; moisture; duration; CO_2 uptake

Acknowledgements
The authors extend their gratitude to the Walloon region and the European Union (NextGeneration EU) for providing funding for this research within the framework of the PNRR REMIND CARBOC project.

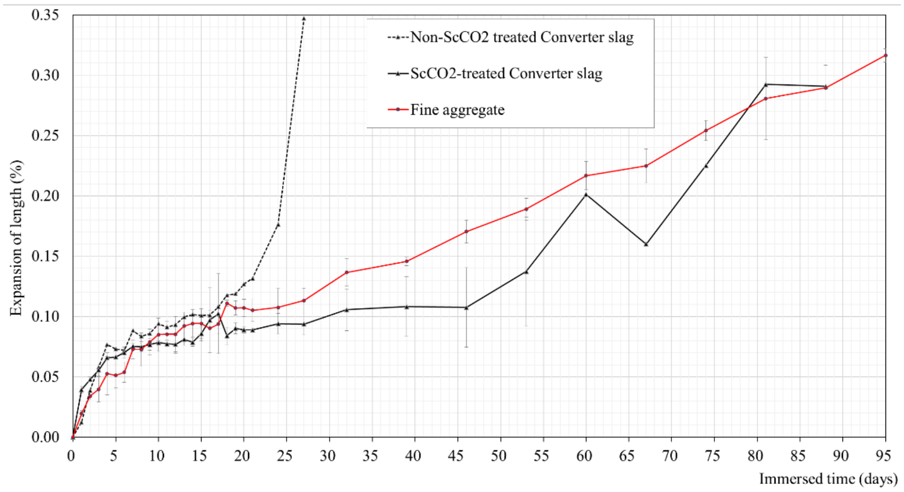
MINERAL CARBONATION AND STABILIZATION OF CONVERTER SLAG USING SUPERCRITICAL CO_2

Chung, C. W.[1], Tran, V. C. [1], Noh, G. M. [1], Moon, H.[2], Kim, J. H.[1]

[1]Division of Architectural and Fire Protection Engineering, Pukyong National University, Busan, Republic of Korea
[2]Korea Institute of Civil Engineering and Building Technology, Goyang-si, Gyeonggi-do, Republic of Korea
Email: cwchung@pknu.ac.kr

Abstract

The converter slag that are produced during steelmaking process has been abandoned or landfilled because the utilization of converter slag as sources of aggregates has been strongly limited. The main reason was associated with its highly expansive reaction in concrete, caused by the hydration of free lime (CaO) and periclase (MgO) to produce portlandite ($Ca(OH)_2$) or brucite ($Mg(OH)_2$). To avoid such problem, approximately 6 months or more of aging period, accompanied by water spraying and mechanical agitation, is suggested. In this work, stabilization of converter slag was attempted by mineral carbonation using supercritical CO_2. Since supercritical CO_2 reacts more aggressively with minerals than gaseous CO_2, it can rapidly induce the formation of calcite ($Ca(CO)_3$) and magnesite ($Mg(CO)_3$) from converter slag by reaction of free lime and periclase. This approach has two significant benefits: 1) to mitigate CO_2 emission from steelmaking industry and 2) stabilization of converter slag in short period of time to promote recycling of this material as an aggregate. After the reaction of converter slag with supercritical CO_2, it was successfully carbonated, showing approximately 5 to 10% of CO_2 uptake by its weight. Results from mortar bar expansion test by ASTM C 1260 procedure showed that mortar bar specimens made of unreacted converter slag showed much higher expansion than those made of reacted converter slag. The 28-day compressive strength of mortar with converter slag was slightly reduced after reaction with supercritical CO_2, but it was still higher than the plain mortar made of conventional siliceous sand.



Keywords
Converter slag; supercritical CO_2; mineral carbonation; expansion

EFFECT OF CARBONATED SLUDGE POWDER ON COMPRESSIVE STRENGTH OF MORTAR

Lett Wai Nwe[1], Atsushi Ueno[1] and Tomohisa Kamada[1]

[1] Department of Civil and Environmental Engineering, Tokyo Metropolitan University, Japan
Email: n-lett@tmu.ac.jp

Abstract

Concrete plant emits not only CO_2 but also large amounts of sludge water (washing wastewater) during concrete production process. These lead to an increasing environmental impact. However, sludge water is rich in calcium ions, hydrated and unhydrated cement minerals. Hence, it has a high potential of CO_2 capturing, and after carbonation solids in the sludge water may be reused as a concrete material. Also, the powdery material obtained after carbonation of the sludge water has a potential to reduce the usage and production of Portland cement, CO_2 emission, and environmental loads. Therefore, we investigated the effects of carbonation of sludge powder (SP) on the compressive strength of mortar and activity index of SP.

Two types of sludge water were used to make carbonated sludge powder (CSP): one was simulated sludge water (5% of cement concentration) prepared in laboratory, and the other was actual sludge water (11.4% of solid concentration) collected from a ready-mixed concrete plant. CO_2 gas was supplied to the simulated sludge water with 1 L/min flow rate to form CSP. Besides, the actual sludge water was used to prepare default sludge water (non-carbonated) and carbonated actual sludge water by supplying CO_2 gas with 2 L/min flow rate. After that, using a spray dryer, the three types of sludge water were dried and powdered into carbonated simulated sludge powder (CSP-S), carbonated actual sludge powder (CSP-A), and non-carbonated actual sludge powder (NSP-A). Particle size distribution, density, and contents of calcium hydroxide (CH) and calcium carbonate (CC) in CSP-S, CSP-A, and NSP-A were analyzed by using laser diffraction technique, JIS A 6201 and TG-DTA, respectively. Mortar specimens were prepared with water to cementitious materials ratio (W/B) of 0.5 and fine aggregate to cementitious materials ratio (S/B) of 3.0. Ordinary Portland cement (density=3.16 g/cm^3, volume average particle size (mv)=28.6 µm) and standard sand as fine aggregate (density=2.64 g/cm^3) were used. Replacement ratio by SPs was 0%, 12.5%, and 25% mass of the cement. Compressive strength test of mortars for 7, 28 and 91 days were conducted. The details of carbonation process, test methods and mortar preparation were explained in our previous research [1].

Table 1 shows the basic properties of three SPs. The mv of NSP-A (16.34 µm) was larger than that of CSP-S and CSP-A, indicating that carbonation process can reduce the particle size of SP and it is more significant with the increase in CO_2 flow rate and carbonation time [2]. The density of CSP-S, NSP-A and CSP-A were not clearly different, and carbonation process did not produce any change on the density of SP. The CC content of NSP-A was 10.98% and its initial content increased to 57.88% in CSP-A after 50 min of carbonation time, and CC of CSP-S increased to 52.86% from 5.44% (0min carbonation). It can be considered that not only CH, but also other unhydrated cement minerals contained in the SP were carbonated and formed into CC during the carbonation process.

Changes in compressive strength of mortar and activity index of SP at 20 C° water curing age of 7, 28, and 91 days are shown in **Figure 1**. The experimental results of all CSP-S, NSP-A, and CSP-A mortars showed that the compressive strength increased from 7 to 91 days of the age although the reduction in the compressive strength increased as the replacement ratio by SP increased. Further, the SP's activity index of CSP-A had the lowest changes from 7 to 28 days compared to that of CSP-S and NSP-A. Comparing the NSP-A mortars with the CSP-A mortars, the compressive strength showed no significant difference except for 25% replacement at 28 days. It may be that CSP operates as a filler in the mixture during the early ages rather than a cementitious mineral, and its reaction increases gradually with the later ages due to the hydration of unhydrated clinker in CSP as well as the slow pozzolanic reaction of CSP. Moreover, in both CSP mortars, the compressive strength of CSP-A mortar was higher than that of CSP-S mortar at 7 and 28 days of age, assuming that the smaller particle size in CSP-A resulted in a more filler effect and pozzolanic reactivity in mortar, which led to the higher strength. Therefore, it can be concluded that CO_2 capture by SP does not have a negative influence on the strength of concrete, however, CSP may have the probability of being used as a cementitious material for approaching to reduce CO_2 emissions and environmental loads.

Table 1. Carbonation of sludge water and basic properties of different sludge powders

Sludge powder types	CO_2 flow rate	Carbonat-ion time	mv (μm)	Density (g/cm^3)	CH (%)	CC (%)
CSP-S (carbonated simulated sludge powder)	1 L/min	30 min	13.14	2.55	3.07	52.86
NSP-A (non-carbonated actual sludge powder)	-	0 min	16.34	2.53	6.88	10.98
CSP-A (carbonated actual sludge powder)	2 L/min	50 min	8.12	2.50	4.03	57.88

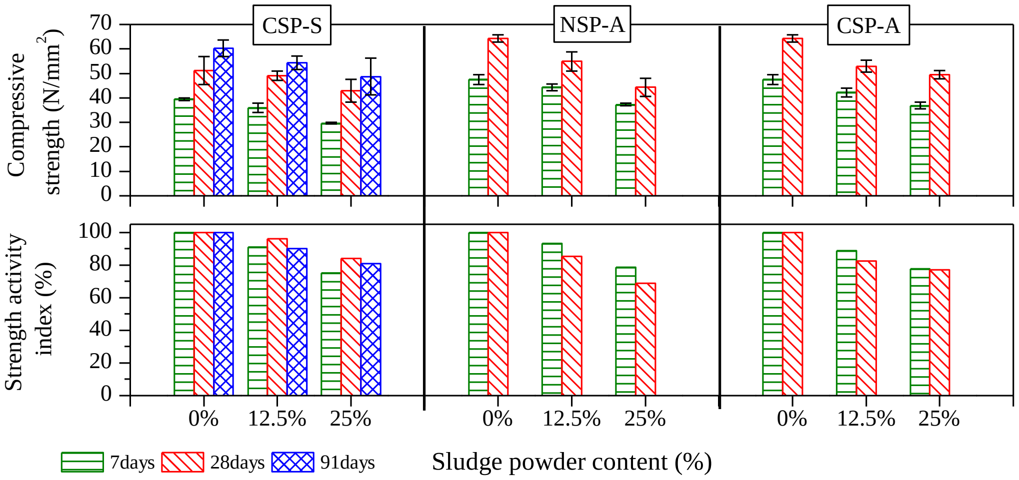


Figure 1. Compressive strength and activity index

Keywords
Carbonation; carbonated sludge powder; compressive strength; activity index

This research was conducted as a part of the project “JPNP21023” commissioned by the New Energy and Industrial Technology Development Organization (NEDO). The authors are grateful to all concerned from the NEDO’s project for their valuable discussion. The authors appreciate Mr. Iwao Mimoto from Uchiyama advance Corp. to supply of actual sludge water sample.

[1] N. Shimizu, L.W. Nwe, A. Ueno, T. Kamada, Fundamental properties of simulated sludge water-derived powder with CO_2. Proceedings of the Japan Concrete Institute. 46, 2024.
[2] Y. Mao, S. Drissi, P. He, X. Hu, J. Zhang, C. Shi, Quantifying the effects of wet carbonated recycled cement paste powder on the properties of cement paste, Cem. Concr. Res. 175, 2024.

PREPARATION OF IN-SITU GROWN NANO-SILICA BASED ON FLY ASH BY CARBONIZATION METHOD: IMPROVING NANO-SILICA DISPERSITY AND SEQUESTERING CO_2

Hailong Sun[1], **Ganghua Pan**[2]

[1,2]School of Materials Science and Engineering, Southeast University, Nanjing, PR China
[1,2]Jiangsu Key Lab of Construction Materials, Southeast University, Nanjing, PR China
Email: ghpan@seu.edu.cn

Abstract

Nano-silica (NS) is an enticing possibility for use as a nano reinforcement in geopolymers. However, one of the most significant barriers to using NS is the difficulty of establishing an ample dispersion in the matrix, which has an enormous effect on its working performance. An innovative method for preparing NS was described in this study, in which carbon dioxide gas was infused into solution to in-situ grow NS (INS) on the surface of fly ash. Temperature, silicon source concentration, carbon dioxide flow rate and other parameters were investigated, and the growth mechanism of INS was explored. The results showed that when the concentrations of Na_2SiO_3 and polyethylene glycol are 0.25 M and 2%, respectively, at 40°C and the CO_2 flow rate is 500-800 mL/min, silica with an average particle size below 100 nm can be produced on the surface of fly ash. In comparison to commercial NS, INS can preserve greater fresh properties and rheology. In addition, the byproduct Na_2CO_3 can be utilized as an activator for geopolymers. This work opens a new strategy for the development of NS as a nano-reinforcing material for geopolymer and provides feasible methods for fixing carbon dioxide and reducing carbon emissions.

Keywords

Carbonation; in-situ grown nano-silica; high dispersion; CO_2 sequestration

UTILIZATION OF CARBONATED STEEL SLAG AS SUPPLEMENTARY CEMENTITIOUS MATERIAL: A REVIEW OF MECHANISM, METHODS AND APPLICATIONS

Xingtong Yue[1,2,3], Jiajie Li[1], and Anlin Shao[3]

[1]School of Civil and Resource Engineering, University of Science and Technology Beijing, Beijing 100083, China
[2]Ansteel Beijing Research Institute Co. Ltd., 102209, China
[3]Institute of Minerals Research, University of Science and Technology Beijing, Beijing, 100083, China
Email: xingtongyue1215@sina.com

Abstract

The burgeoning production of steel slag as an industrial by-product has led to significant environmental and sustainability challenges. The conventional disposal method of landfilling steel slag leads to the wastage of valuable materials and poses environmental risks. Direct application of untreated steel slag in construction is also problematic due to its potential to degrade the structural integrity and longevity of construction materials. To mitigate these issues, researchers have been exploring the accelerated carbonation process as a means to transform steel slag into a more sustainable and environmentally friendly material. Recently, preparing the supplementary cementitious material (SCM) from carbonated steel slags (CSS) has aroused the interest of a wide range of researchers. This paper presents a comprehensive review of the mechanisms underlying steel slag carbonation and its subsequent application as the SCM.

The carbonation process is explored in detail, elucidating the chemical reactions that occur when steel slag reacts with carbon dioxide (CO_2). This reaction not only facilitates the sequestration of CO_2 but also endows the steel slag with enhanced cementitious properties. The paper delves into the influence of key parameters such as temperature, CO_2 concentration, pressure and moisture content on the carbonation efficiency, highlighting the optimal conditions necessary for effective SCM production.

The review further investigates the pozzolanic and hydraulic activities of CSS, which are crucial for its performance as an SCM. The improved reactivity and the formation of stable calcium carbonate ($CaCO_3$) and amorphous silica during carbonation are identified as key factors that augment the strength and durability of cementitious composites.

Practical applications of carbonated steel slag as an SCM in the construction industry are also examined. The paper demonstrates how the incorporation of this material can lead to improved mechanical properties in concrete, such as increased compressive and flexural strengths, as well as enhanced resistance to environmental degradation.

In summary, this review underscores the environmental and economic benefits of utilizing steel slag carbonation for the production of SCMs. It advocates for further research and development in this area, with the aim of optimizing carbonation techniques and expanding the use of carbonated steel slag in sustainable construction practices.

Keywords

Steel slag; carbonation; supplementary cementitious materials; mechanism; environmental sustainability

MECHANICAL PROPERTIES AND MICROSCOPIC CHARACTERISTICS OF TERNARY COMPOSITE CARBONATED CEMENTITIOUS MATERIALS OF S95 GRADE BLAST FURNACE SLAG-STEEL SLAG-CALCIUM CARBIDE SLAG

Ma, Wene[1], Tong, Liyuan[1], Zhang, Tiantian[1], Zheng, Shuwen[1], and Sun, Dafang[2]

[1]Transportation College, Southeast University, Nanjing 211189, China
[2]Nanjing Metro Town Development and Construction Group Co., Ltd., Nanjing 211135, China
Email: 1224696716@qq.com

Abstract

With the gradual promotion of green, circular and low-carbon production methods in building material industry, the utilisation of industrial solid waste and carbon dioxide emission reduction is an important way to solve the environmental resource problems and achieve the dual-carbon target.

In this paper, S95 grade blast furnace slag(GGBS), steel slag and calcium carbide slag(CS) were used as cementitious materials. The mechanical properties and microscopic characteristics of ternary composite cementitious materials after carbonation were researched by compressive strength test, flexural strength test, X-ray diffraction test and scanning electron microscope test. The carbonation mechanism of the material was analysed, the recommended mixing ratio and carbonation time for practical application were proposed, and the feasibility of the application of the material in practical engineering was discussed.

Firstly, the main chemical composition and content of the three curing agents were measured by X-ray fluorescence spectroscopy analysis. Then, the optimum mixing ratio of GGBS&steel slag: CS(GGBS: steel slag = 1:1) was researched, and the test factors and level parameters are shown in Table 1. Finally, based on the above optimum mixing ratio, the optimum mixing ratio of GGBS and steel slag was studied. The water-cement ratio was 0.5. The slurry after mixing the curing agent and water was poured into a 40mm*40mm*160mm triple flexural mould in three layers and vibrated in layers. The completed specimens were demoulded after 1 day of standard curing. After demoulding, the specimens of carbonation groups were carbonised in a carbonation chamber at 25°C, 75% humidity and 20% CO_2 concentration, and the comparison specimens were tested after ordinary curing at the same temperature and humidity conditions. The average value of three parallel specimens was taken as the representative value.

Table 1. Orthogonal test factors and horizontal parameters

level parameter	test factor	
	Curing age/day	GGBS&steel slag:CS （GGBS:steel slag = 1:1）
1	1（carbonation）	4:1
2	2（carbonation）	3:2
3	4（carbonation）	2:3
4	6（carbonation）	1:4
comparison group 2	2（ordinary）	-
comparison group 4	6（ordianry）	-

As shown in Figure 1, the compressive strength increases with carbonation curing age. For 1~4 days, it rises with the growth of GGBS&steel slag: CS dosage ratio(GGBS: steel slag = 1:1); for 4~6 days, the compressive strength growth rate of 4:1 ratio is only 4.57%. Figure 2 reveals that only the flexural strength of specimen with 2:3 ratio increases continuously for 1~6 days. Thus, the optimum ratio of GGBS&steel slag: CS is selected as 2:3. Then, as shown in Figures 3 and 4, the compressive and flexural strengths of the specimens with a ratio of 2:1 between GGBS and steel slag are the highest for 4~6 days, so the ratio of GGBS and steel slag is selected as 2:1. Moreover, the compressive and flexural strengths of the carbonation-conditioned specimens are greater than those of the ordinary-conditioned specimens. In summary, the optimal ratio of ternary composite carbonation cementitious material is GGBS&steel slag: CS=2:3 (GGBS:steel slag=2:1), and the carbonation curing age is 4~6 days. Figure 5 shows that the main product of carbonation curing is $CaCO_3$, with 1:4 and 2:3 groups having higher $CaCO_3$ peaks than 4:1 group. But there's $Ca(OH)_2$ in 1:4 group. Figure 6 reveals that under the ratio of 2:3, the structural densification of the material is greatly enhanced, with the microscopic pore spaces filling with $CaCO_3$, C-S-H, etc. The reasonableness of this dosage ratio was further verified.

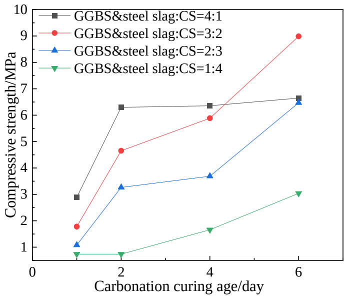


Figure 1. Compressive strength

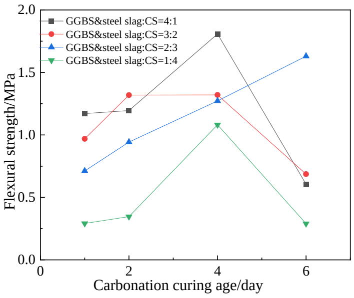


Figure 2. Flexural strength

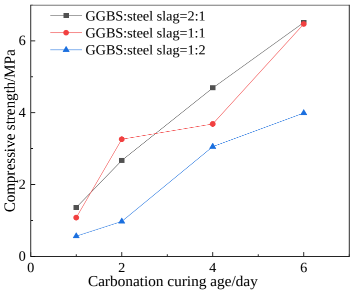


Figure 3. Compressive strength

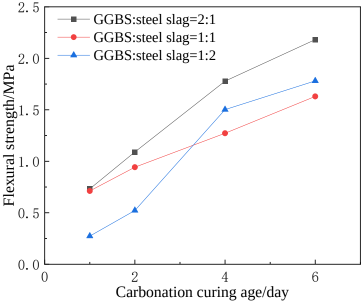


Figure 4. Flexural strength

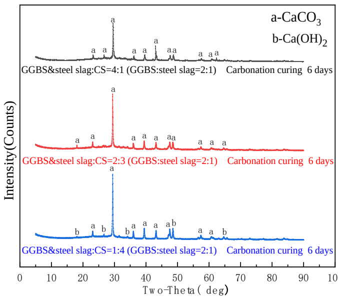


Figure 5. XRD

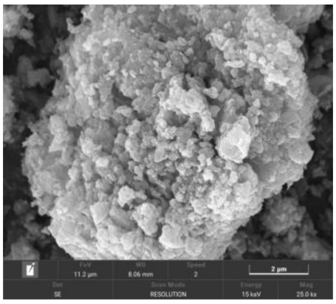


Figure 6. SEM

In conclusion: (1) When the content of CS is high, there are more OH^- ions, and it is easy to sustain the carbonation reaction. The resulting $CaCO_3$ fills the pores in the material and enhance material strength. However, the content of CS should not out of range, too much $Ca(OH)_2$ will hinder the hydration reaction and become the weakness, resulting in a strength decrease. (2) The carbonation effect of GGBS is better than that of steel slag, because the content of CaO, Al_2O_3, SiO_2 and MgO in GGBS is higher than that in steel slag, which is able to generate more carbonate salt and gelling substances such as C-S-H. (3) Through XRD and SEM tests, it can be determined that the main product is C-S-H when it is not carbonised, and $CaCO_3$ after carbonation. The prolongation of carbonation time can effectively promote the generation of $CaCO_3$. (4) It is recommended that the optimal mixing ratio of ternary composite carbonation cementitious materials is GGBS&steel slag:CS=2:3 (GGBS:steel slag=2:1), and the carbonation curing time is 4~6 days.(5) The GGBS-steel slag-CS ternary composite cementitious material has good performance after carbonation, and it can be used as an alternative material for the carbonation and curing project of soft soil.

Keywords
Carbonation; industrial solid waste; cementitious materials; mechanical properties; microscopic properties

IMPACT AND MECHANISM OF ULTRASONIC CARBONATED MSWI FLY ASH ON CO_2 MINERALIZATION CURING OF CEMENT

Jie Chen[1], Xiaoqing Lin[1], Xiaodong Li[1], and Jianhua Yan[1]

[1]State Key Laboratory of Clean Energy Utilization, Institute of Thermal Power Engineering of Zhejiang University, Hangzhou 310027, Zhejiang, China
Email: linxiaoqing@zju.edu.cn

Abstract

Our team developed an innovative approach by integrating ultrasonic chemical methods with accelerated carbonation treatment technology for MSWI FA, which enhanced its carbon sequestration capacity, heavy metal stabilization, and dechlorination efficiency [1]. The resulting Ultrasonic Carbonated FA (UFA) was characterized by its high calcium carbonate content, low chlorine levels, small particle size, and high degree of homogenization. Subsequent research confirmed UFA's feasibility as a partial cement replacement material. After 28 days of room-temperature steam curing, the unconfined compressive strength of cement paste test blocks containing 50% UFA surpassed 30 MPa [2]. Furthermore, UFA demonstrated extremely low risks of heavy metal leaching in both acidic and alkaline environments. To further mitigate the cement industry's carbon emissions, traditional steam curing was proposed to be replaced with CO_2 mineralization curing. During this process, calcium ions in cement reacted with CO_2 to form $CaCO_3$, enhancing the mechanical properties of cement by filling its pores [3].

Therefore, leveraging the high calcium carbonate content in UFA, we proposed a novel CO_2 mineralization and curing method for UFA-doped cement. By evaluating the compressive strength and carbon fixation depth of cement paste test blocks across varying curing periods, we aimed to determine the optimal UFA blend ratio and its impact on cement's compressive strength and carbon fixation rate. Additionally, the physical and chemical properties (crystalline structure, functional groups, surface micromorphology, porosity) of cement slurries with varying UFA contents were characterized to explore the interaction mechanisms during the CO_2 mineralization curing process. Heavy metal leaching tests on different FA and cement paste samples further validated the stabilization effect of this treatment on heavy metals in FA. As evidenced in Figure 1(a), increasing UFA content from 0 to 50% initially enhanced and then diminished the compressive strength of cement paste. At 10% UFA addition, the compressive strength peaked at 50.23 MPa, attributed to the nanoscale $CaCO_3$ in UFA that not only filled cement pores effectively but also provided nucleation sites for the CO_2 mineralization reaction, further enhancing pore filling. The porosity results in Figure 1(b) showed decreased porosity at 5% and 10% UFA levels. However, at 50% UFA, both compressive strength and porosity declined significantly due to the inert nature of $CaCO_3$, which impeded hydration reactions and continuous C-S-H growth. Figure 2 indicated that adding 5% to 20% UFA did not enhance the carbon fixation efficiency of cement, primarily because it reduced surface porosity, thus hindering CO_2 diffusion into the cement interior. Conversely, at 50% UFA, increased porosity facilitated both CO_2 diffusion and the carbonation reaction. Even at high UFA levels, the concentration of leached heavy metals remained below the "Groundwater Quality Standards" for the limit of class IV water, highlighting the efficacy of ultrasonic carbonation treatment in reducing heavy metal leaching toxicity from original FA and its further improvement by cement solidification.

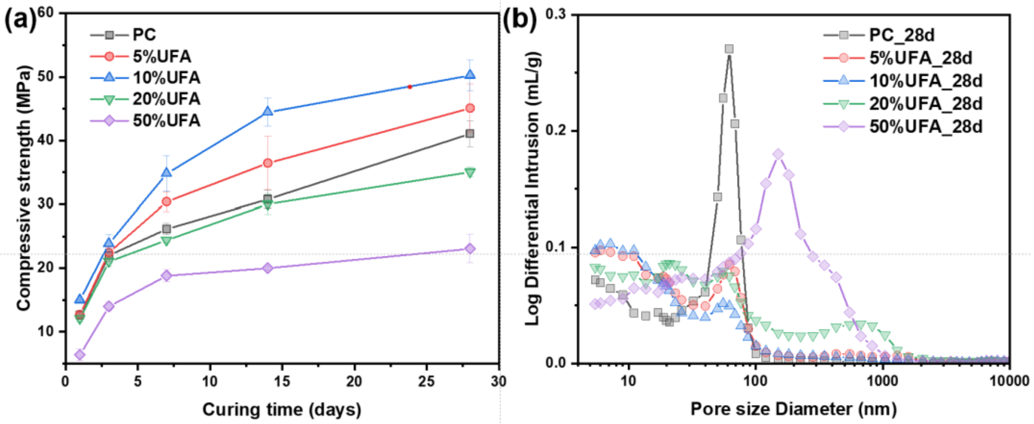


Figure 1. Compressive strength (a) and pore distribution characteristics (b) of different cement pastes

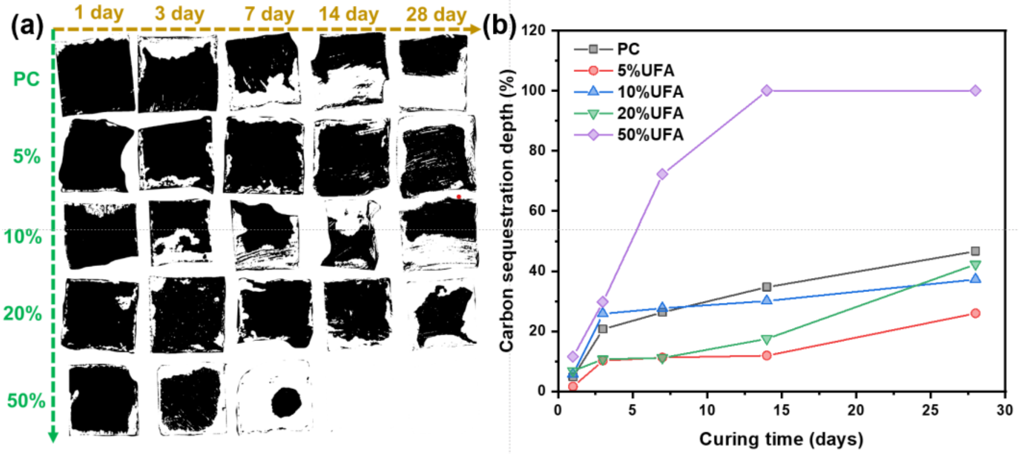


Figure 2. Cross-section binarization (a) and carbon fixation depth (b) of different cement pastes

Keywords

MSWI fly ash; ultrasonic carbonation; CO_2 sequestration; mineralization; cement

Acknowledgement

This study was financially supported by the National Natural Science Foundation of China (52076192, 52376192).

References

[1] J. Chen, C. Fu, T. Mao, Y. Shen, M. Li, X. Lin, X. Li, J. Yan, Study on the accelerated carbonation of MSWI fly ash under ultrasonic excitation: CO2 capture, heavy metals solidification, mechanism and geochemical modelling, Chemical Engineering Journal. 450 (2022) 138418. https://doi.org/10.1016/j.cej.2022.138418.

[2] J. Chen, W. Zhu, Y. Shen, C. Fu, M. Li, X. Lin, X. Li, J. Yan, Resource utilization of ultrasonic carbonated MSWI fly ash as cement aggregates: Compressive strength, heavy metal immobilization, and environmental-economic analysis, Chemical Engineering Journal. 472 (2023) 144860. https://doi.org/10.1016/j.cej.2023.144860.

[3] Z. Tu, M.Z. Guo, C.S. Poon, C. Shi, Effects of limestone powder on CaCO3 precipitation in CO2 cured cement pastes, Cement and Concrete Composites. 72 (2016) 9–16. https://doi.org/10.1016/j.cemconcomp.2016.05.019.

HIGH-TEMPERATURE CARBONATION BEHAVIOR, REACTION KINETICS AND MICROSTRUCTURAL CHANGE OF HYDRAULIC AND NON-HYDRAULIC CALCIUM SILICATES

Hao Yu[1], **Xing-Yuan Lei**[1], **Tung-Chai Ling**[1]

[1]College of Civil Engineering, Hunan University, Changsha-410082, China
Email: tcling@hnu.edu.cn; tcling611@yahoo.com

Abstract

Diverse hydraulic characteristics of calcium silicates can result in variations in hydration and carbonation reactions, particularly when exposed to elevated temperatures curing. In this study, the influence of hydration degree on high-temperature (60-140°C) carbonation was studied, and tricalcium silicate (C_3S) and γ-dicalcium silicate (γ-C_2S) were chosen as precursor minerals to examine their carbonation products and the evolution of microstructure at elevated CO_2 temperature curing. Specifically, considering carbonation degree with time, the reaction kinetics for two minerals were also simulated at 120°C, for a better understanding of the carbonation rate with curing temperature.

The results showed that high temperature can generally increase the carbonation efficiency, while simultaneously altering the calcium carbonate (Cc) crystalline, C-S-H morphology and products distribution. Calcite, vaterite and sheet-like C-S-H were detected in C_3S carbonation, wherein the C-S-H product was nucleated around the unreacted C_3S particles, densifying the matrix and inhibiting further CO_2 diffusion. As for non-hydraulic γ-C_2S, a loose microstructure with three layers of inner γ-C_2S, interlayered C-S-H gel and outer calcite was presented, which was favorable to CO_2 diffusion but not benefiting to the matrix densification.

Due to the water loss under high temperature condition, an improved two-stage kinetic model is developed for the simultaneous incorporation of temperature effect on the intrinsic reactivity and the moisture retainment of C_3S and γ-C_2S. The first stage is within 15~30 min, where the revised nucleation model and phase-field model can be applied in this stage. It implies that elevating curing temperature increases the apparent reaction rate constant and thus improves the carbonation efficiency, which is more significant in γ-C_2S reaction. After that, the reaction kinetics depends on the water content inside of the sample, where a diffusion-controlled model can be employed with available water in sample. Nevertheless, the reaction would be terminated with the absence of water. This also highlights the fact that the initial carbonation rate is fast at high temperatures, while the carbonation degree is attenuated for long-term curing.

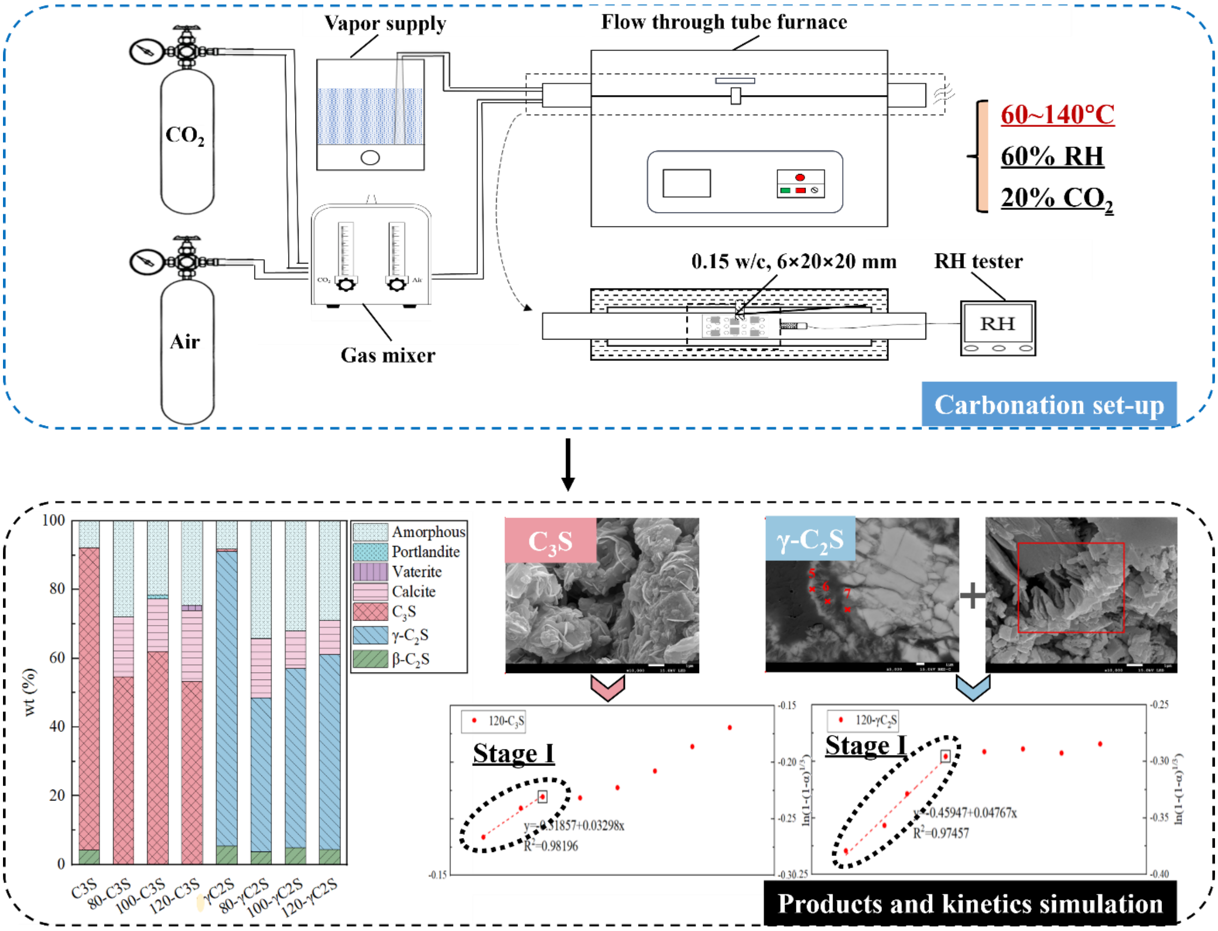


Figure 1. High-temperature CO_2 set-up and characterization of carbonated C_3S and γ-C_2S compacts

Keywords

High-temperature carbonation curing; hydraulic characteristics; reaction kinetics; C_3S; γ-C_2S

Session B4

Carbonation Modelling

COMPREHENSIVE MOLECULAR-SCALE INSIGHTS ON THE INTERFACIAL CO_2 MINERALIZATION OF PORTLANDITE

Roland J.-M. Pellenq[1], **Xinping Zhu**[2], **Romain Dupuis**[2], **Katerina Ioannidou**[2], **Yong Tao**[3] **and Chi-Sun Poon**[3]

[1] IEM, CNRS and University of Montpellier, France
[2] LMGC, CNRS and University of Montpellier, France
[3] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hong Kong
Email: roland.pellenq@cnrs.fr

Abstract

Carbon dioxide (CO_2) mineralization of the widely available alkali earth-metal sources has great potential in migrating the global anthropogenic CO_2 emissions for dozens of Gt per year. Yet, the large-scale CO_2 sequestration through this mineralization method remains a challenge without comprehensively elucidating the CO_2 mineralization reactions.

Unveiling the early-stage molecular-scale physicochemical progress along the complete CO_2 mineralization path is paramount to advancing this method. Herein, we implement a series of advanced molecular simulations (Grand Canonical Monte-Carlo (GCMC), reactive force field (ReaxFF), and well-tempered metadynamics) to track the complete interfacial CO_2 mineralization path of a CO_2-thin water film-portlandite system.

Five steps, including water adsorption, ion dissolution, CO_2 adsorption, CO_2 speciation, and the formation of $CaCO_3$ precursor, are progressively observed. In particular, we analyze the effects of molecular surface roughness of the mineral and the acid-base environment of the nanometric water film on these steps. Finally, we propose the complete interfacial CO_2 mineralization path for the portlandite. Our results suggest that the kink is the reactive site. The Ca dissolution and CO_2 speciation processes compete with each other for the acid-base environment of the film. It turns out that the neutral water film yields the optimal carbonation degree, which has been further validated by our carbonation experiment.

Keywords
Carbonation; GCMC; reactive force field; well-tempered metadynamics; free-energy

STUDY OF CARBONATION KINETICS OF A RECYCLED SAND

Khaoula Mebrek[1,2], **Rachid Cherif**[1], **Abdelkarim Aït-Mokhtar**[1]

[1]LaSIE UMR CNRS 7356, University of La Rochelle, Avenue Michel Crépeau, 17042 La Rochelle Cedex 1, France
[2]Agence de La Transition Ecologique (ADEME), 20 Avenue du Grésillé B.P.90406, 49004 Angers Cedex 01
Email: rachid.cherif@univ-lr.fr

Abstract

The Waste Framework Directive 2008/98/EC mandates that each European Union member state must achieve a minimum recycling rate of 70% of construction and demolition wastes (CDW) since 2020 (Song and Ryou, 2014). To meet this target, recycling CDW into construction materials like aggregates, cement, concrete, and sands is imperative (Kou et al., 2012; Sormunen and Kärki, 2019). Otherwise, sand holds a primary position as the most utilized material in the construction industry with river sand being the most natural aggregate employed in the production of concrete materials throughout the entire construction sector (Kou et al., 2012).

The aim of this paper is to monitor the carbonation kinetic of a recycled sand with a particle size fraction of 0/4. The study was performed on a sand crushed from a mortar mixed in laboratory and based on standardized non-carbonated sand. 24 hours After mixing, the mortar was cured in water during 28 days in order to avoid carbonation. After that, the sand crushed was exposed to accelerated carbonation in laboratory during 7 days (until a maximum carbonation). A monitoring of the mass gain, pH evolution and carbonation products by TGA was carried out on the sand during carbonation.

Tests and Procedure

The recycled sand (RS) employed in this research were produced in laboratory conditions from a standardized mortar. A purely siliceous standardized sand (0/4 mm) was used to ensure that the sand does not contain carbonates. Ordinary Portland cement CEM I 52.5 N according to the European Standard EN 197-1 was used. The mass fractions of the principal clinker phases provided by the manufacturer (Calcia, Bussac-France) and based on the Bogue calculation method are 65% C_3S, 13% C_2S, 7% C_3A, 13% C_4AF and 4.9% gypsum. The chemical composition and density of the cement used is given in Table 1.

Table 1. Chemical composition and density of the cement used.

Composition	Cao	SiO_2	Al_2O_3	Fe_2O_3	SO_3	K_2O	Na_2O	Density
CEM I (wt%)	64.20	20.50	5.00	3.90	2.50	0.29	0..05	3.80

The water-to-cement ratio (w/c) of the mortars used was 0.6, while the sand-to-cement ratio was 3. They were cast in cylindrical molds of Ø 12x22 cm². After 24 hours, the mortar specimens were demolded and stored in water at an ambient laboratory temperature until the age of 28 days. Next, these mortars were crushed into various particle sizes. The desired RS particle distribution size (0/4mm) was obtained by sieving and then exposed to accelerated carbonation. Initial state of the recycled sand (RS) used was assessed through phenolphthalein spraying before carbonation.

Accelerated carbonation

Plastic bins containing bed of recycled sand with a particle size fraction of 0/4 mm were placed in a carbonation chamber with a CO_2 concentration of 3% to 4% and relative humidity of 65%. A fan is positioned at the top of the chamber to ensure the homogenization of the ambient air. Relative humidity control is achieved using saturated saline solutions composed of magnesium nitrate. In order to monitor qualitatively and quantitatively the carbonation process of the recycled sand, colorimetric phenolphthalein test, mass gain, pH evolution and thermogravimetric analysis (TGA) were carried out on sand samples during the carbonation at the following exposure times: 0; 8; 24; 32; 48; 56; 72; 80; 96; 104; 120; 128; 144; and 152 hours.

Some results and discussion

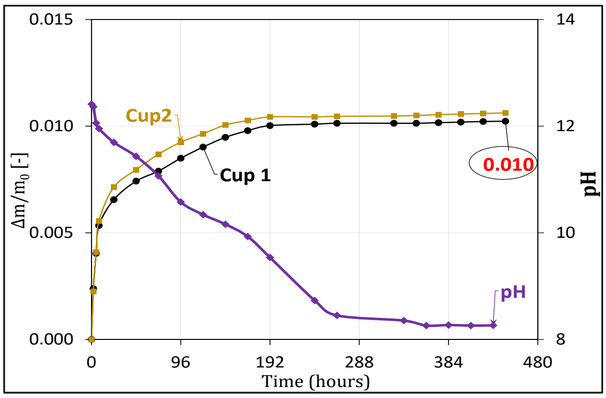


Fig. 1. Mass gain and pH evolution of recycled sand during carbonation.

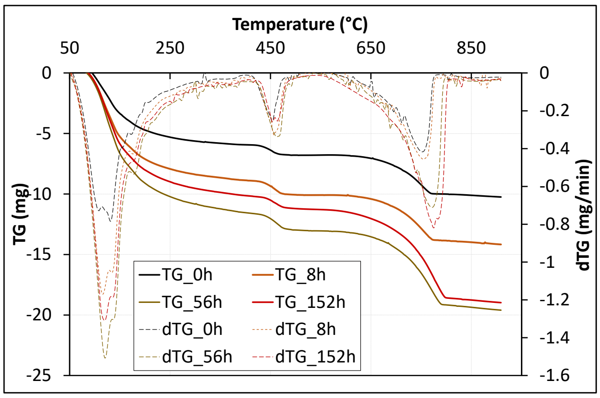


Fig. 2. TG/dTG curves of recycled sand carbonation**.**

1. A significant rise in mass within the first hours of carbonation (eight for our study), which points out a quick response from the cementitious matrix. After 4-day carbonation, the mass evolution of the carbonated recycled sand was relatively stable.
2. The gradual pH decrease of carbonated recycled sand over time is consistent with carbonation reactions, indicating a continuous precipitation of calcium carbonate. After 6-day carbonation, the pH of sand was not yet stable, contrary to mass gain, which could mean that the recycled sand carbonation was not yet maximum. pH monitoring is, a priori, more sensitive to carbonation than mass monitoring.
3. A substantial initial increase in $CaCO_3$ production during the first eight hours of carbonation, followed by a consistent rise in $CaCO_3$ content until 152 hours. This indicates a prolonged equilibrium phase in the carbonation process, suggesting difficulty in CO_2 diffusion into the inner part of RS beyond the initial intensive carbonation phase.

References

Song, I.H., Ryou, J.S., 2014. Hybrid techniques for quality improvement of recycled fine aggregate. Construction and Building Materials 72, 56–64. https://doi.org/10.1016/j.conbuildmat.2014.08.041

Sormunen, P., Kärki, T., 2019. Recycled construction and demolition waste as a possible source of materials for composite manufacturing. Journal of Building Engineering 24, 100742. https://doi.org/10.1016/j.jobe.2019.100742

Kou, S., Zhan, B., Poon, C., 2012. Feasibility study of using recycled fresh concrete waste as coarse aggregates in concrete. Construction and Building Materials 28, 549–556. https://doi.org/10.1016/j.conbuildmat.2011.08.027

Sormunen, P., Kärki, T., 2019. Recycled construction and demolition waste as a possible source of materials for composite manufacturing. Journal of Building Engineering 24, 100742. https://doi.org/10.1016/j.jobe.2019.100742

NANOSCALE MECHANISMS OF CO_2 DOCKING IN MINERAL MESOPORES AT DIFFERENT RELATIVE HUMIDITIES

Gen Li[1], Yong Tao[1], Xinping Zhu[2], Yining Gao[1], Peiliang Shen[1], Binbin Yin[1], Romain Dupuis[2], Katerina Ioannidou[2], Roland J.-M. Pellenq[3], Chi Sun Poon[1]

[1]Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong
[2]Laboratoire de Mécanique et Génie Civil, CNRS and Université of Montpellier, 490 Rue St Priest, 34090 Montpellier, France
[3]Institut Européen des Membranes, CNRS and Université of Montpellier, 300 avenue du Professeur Jeanbrau, 34090 Montpellier, France
Email: yong.tao@polyu.edu.hk; roland.pellenq@cnrs.fr; chi-sun.poon@polyu.edu.hk

Abstract

Carbon mineralization utilizing calcium/magnesium-rich minerals or solid wastes is one of the most promising carbon-negative schemes targeting global carbon neutrality. Its feasibility critically relies on the carbonation efficiency of CO_2-reactive minerals, which is to a great extent influenced by the water content and state within the mesopores of the materials. Yet, the pivotal role of confined water in regulating mineral carbonation at the nano-/meso-scale is not well understood. In this atomistic simulation work, we reveal the optimal thermodynamic condition (relative humidity) for CO_2 docking in hydrophilic mesoporous materials, which represents a critical step towards efficient mineral carbonation.

Using Portlandite [$Ca(OH)_2$] slit mesopore models of various pore sizes within a Grand Canonical frame, we find that the maximum CO_2 intake occurs precisely when the mesopore reaches a critical water amount that corresponds to the onset of the water capillary condensation (phase transition). At this transition state, the mesopores are filled with metastable density-fluctuating water layer adsorbed on the mesopore surface, creating an ideal environment for CO_2 docking. This critical water amount, interpreted as optimal relative humidity, was found to be dependent on the pore size, which was theorized with a Kelvin-like equation that eventually allowed us to propose an engineering model that can predict the optimal relative humidity for CO_2 intake in practical hydrophilic materials.

Building upon classical theories of phase transition of confined water in mesopores, this study unveils the surprising CO_2 docking ability of the metastable water film approaching capillary condensation transition in hydrophilic mesopores. It also enhances the engineering of optimizing carbonation conditions in pursuit of high-efficiency carbon mineralization in real-world applications under global carbon neutrality strategies.

Keywords
Carbon mineralization; grand canonical monte carlo; CO_2 adsorption; hydrophilic mesopores

HYGRO-THERMO-CHEMICAL MODELING OF ACCELERATED CARBONATION CURING OF CEMENTITIOUS MATERIALS AT EARLY AGE

Yang, L.[1], **Di Luzio, G.**[2], **Cusatis, G.**[3], **Su, H.**[4], **and Dai, J.G.**[1]

[1]Department of Architecture and Civil Engineering, City University of Hong Kong, Hong Kong, China
[2]Department of Civil and Environmental Engineering, Politecnico di Milano, Milan, Italy
[3]Department of Civil and Environmental Engineering, McCormick School of Engineering and Applied Science, Northwestern University, Evanston, IL, USA
[4]College of Water Conservancy and Hydropower Engineering, Hohai University, Nanjing 210098, China
Email: jiangdai@cityu.edu.hk (Dai, J.G.)

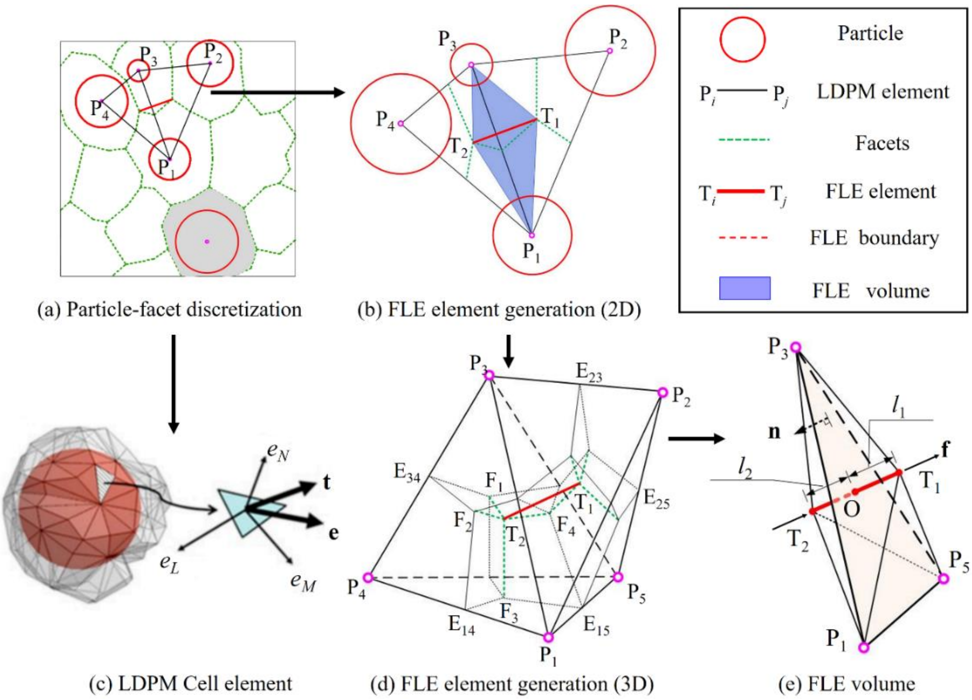
Abstract

Accelerated carbonation curing has been regarded as a promising curing method and CO_2 capture technique that can enhance the early strength and increase the CO_2 sequestration capacity of cement-based materials. Carbonation in cement-based materials involves outside gaseous CO_2 diffusing into solid mass, and a series of chemical reactions, in which not only hydration products, but also unhydrated clinker minerals can participate in [1-3]. Many scholars made much efforts on the improvement of treatment efficiency of carbonation curing, while the understanding on coupling effect of carbonation and hydration reactions at early age is still needed to optimize the carbonation curing scheme.

This study proposes a theoretical formulation for the kinetics of hydration and carbonation reactions at early age. Starting from the micromechanisms of surface reactions and the governing reaction equations, the effects of curing history are analyzed to explore the coupling effects between hydration and carbonation reactions. Based on the kinetics model, this work further proposes a hygro-thermo-chemical model for cementitious materials under carbonation curing suitable for the analysis of moisture transport, heat transfer, and chemical process at early age and beyond. Classical mass and energy conservation laws were formulated by using relative humidity and temperature as primary variables and by taking into account explicitly various chemical reactions, i.e. cement hydration and carbonation.

The hygro-thermo-chemical model was formulated within the Multiphysics Lattice Discrete Particle Model (M-LDPM) framework [4] which combines the Lattice Discrete Particle Model (LDPM) [5] with the dual Flow Lattice Element (FLE) framework. LDPM is a mesoscale model for simulating the mechanical response of concrete under a wide range of loading and environmental conditions. The FLE framework simulates the flow behavior in concrete and is inherently coupled with the LDPM system. Within the LDPM geometry (as shown in Fig. 1c for a 3D visualization), a dual lattice system for coupled simulation of chemical/transport processes is constructed from a FLE system. The FLE system is anchored to the LDPM system and is constructed in such a way that each FLE connects two tet-points belonging to two adjacent tetrahedra (as shown in Fig. 1a for a 2D visualization). The FLE, visualized by the solid thick line segment in Figs. 1b, d and e, connects the two points T_1 and T_2, in which the field variables are defined, i.e. temperature, T, and relative humidity, h. The hygro-thermo-

chemical model for heat transfer and moisture transport was based on Di Luzio and Cusatis' work [6].



(a) Particle-facet discretization (b) FLE element generation (2D)
(c) LDPM Cell element (d) FLE element generation (3D) (e) FLE volume

Figure 1 Visualization of basic element in dual lattice system

The proposed model was calibrated and verified by comparison with experimental test data. Based on the sensitivity analysis, the scheme of carbonation curing can be further optimized and analyzed.

Keywords
Carbonation; hydration; kinetics; multiphysics lattice discrete particle model; early age

Acknowledgement
The authors gratefully acknowledge the financial support from the Belt and Road Special Foundation of National Key Laboratory of Water Disaster Prevention (2023nkms07).

References

[1] Poon, C. S., Shen, P., Jiang, Y., Ma, Z., & Xuan, D. (2023). Total recycling of concrete waste using accelerated carbonation: A review. *Cement and Concrete Research*, 173, 107284.
[2] Wang, X., Guo, M. Z., & Ling, T. C. (2022). Review on CO2 curing of non-hydraulic calcium silicates cements: Mechanism, carbonation and performance. *Cement and Concrete Composites*, 133, 104641.
[3] Chen, T., Gao, X., & Qin, L. (2019). Mathematical modeling of accelerated carbonation curing of Portland cement paste at early age. *Cement and Concrete Research*, 120, 187-197.
[4] Yang, L., Pathirage, M., Su, H., Alnaggar, M., Di Luzio, G., Cusatis, G. (2021) Computational modeling of temperature and relative humidity effects on concrete expansion due to alkali-silica reaction. *Cement and Concrete Composites*, 124: 104237.
[5] Cusatis, G., Pelessone, D., & Mencarelli, A. (2011). Lattice discrete particle model (LDPM) for failure behavior of concrete. I: Theory. *Cement and Concrete Composites*, 33(9), 881-890.
[6] Di Luzio, G., & Cusatis, G. (2009). Hygro-thermo-chemical modeling of high performance concrete. I: Theory. *Cement and Concrete composites*, 31(5), 301-308.

PROBING Al UPTAKE IN C-S-H GELS VIA DFT AND MOLECULAR DYNAMICS SIMULATIONS: TOWARDS MAXIMIZING Al/Si RATIOS AND CHLORIDE ION ADSORPTION

Li Zhang[1], **Songhui Liu**[1], **Xuemao Guan**[1]

[1]Henan Key Laboratory of Materials on Deep-Earth Engineering, School of Materials Science
and Engineering, Henan Polytechnic University, Jiaozuo 454003, China
Email: zhangli_hpu@126.com

Abstract

Based on the concept of sustainable design of cementitious materials, various types of aluminum-rich supplementary cementitious materials (SCMs), such as fly ash and blast furnace slag, are often introduced in the design of cementitious materials. The Al^{3+} released from the hydration of these SCMs is easily doped into the calcium silicate hydrate (C-S-H) gel structure, forming calcium aluminosilicate hydrate (C-A-S-H) gel, which in turn affects the C-(A)-S-H gel structure and chloride ion adsorption mechanism. In this paper, the C-(A)-S-H structures with Ca/Si ratio = 1.0 were designed to investigate the present state, uptake site, uptake stability, and uptake amount of Al in C-S-H gels from the defect formation energy, lattice distortions, bond order-bond length (BO-BL) distributions, the density of states (DOS) and electron density differences (EDD) based on first-principles simulation, to construct reasonable C-(A)-S-H substrate structure models. On this basis, the molecular structure models of NaCl solution transport in the C-(A)-S-H pore channel were constructed, and the Al/Si ratios were set to 0.05, 0.1, 0.15, and 0.2, respectively. The effect and microscopic mechanism of Al doping on the adsorption of chloride ions in C-S-H gels were investigated based on molecular dynamics simulations.

The results show that Al could not replace Ca sites and had different tendencies to replace different types of Si sites in C-S-H gels. Aluminum occupies preferentially the bridge silica-oxygen tetrahedral sites rather than the pairing ones and the chain end positions. In addition, Al is present in the tetracoordinate form at the bridging sites. This is due to the bridge positions being less geometrically constrained and Al[IV] can be deformed by twisting to reach the energy minimum state. As the initial Al/Si ratio increases, the actual Al/Si ratio and the proportion of Al occupying bridging Si sites both increase, but their growth rates gradually decrease and level off. The upper limits of the two are only 0.12 and 50%, respectively. Keeping the Ca/(Si+Al) ratio constant, Al uptake at the Si site enhances the Cl^- adsorption capacity of C-(A)-S-H gels, and the optimal Al/Si ratio is 0.1, i.e., Al/Si ratio ≤ 0.1, the higher the Al/Si ratio, the stronger the Cl^- adsorption capacity of C-(A)-S-H gels. For Al/Si > 0.1, the higher the Al/Si ratio, the weaker the Cl^- adsorption capacity of C-(A)-S-H gels, but still higher than the pure C-S-H gels. Therefore, to enhance the ability of C-(A)-S-H gel to bind Cl^-, its Al/Si ratio should be increased at a fixed Ca/(Al+Si) ratio (Al/Si≤0.1). The above results can provide the theoretical reference and guidance for the optimal admixture of SCMs dopants in cement for service in the offshore environment.

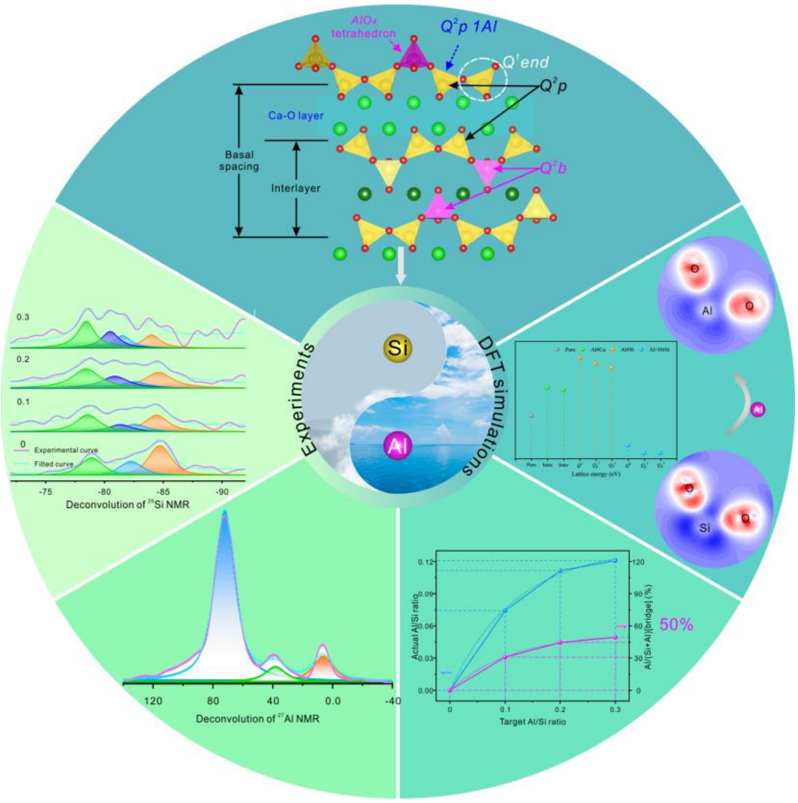


Fig. 1 Uptake behaviors of Al in C-S-H gels

Keywords
Aluminium uptake; C(-A)-S-H gel; Al/Si ratio; micro- and electronic structures evolution; theoretical simulations

IMPACT OF Fe DOPING ON THE REACTIVITY OF γ-DICALCIUM SILICATE: INSIGHTS FROM DFT CALCULATIONS

Zhao, M. [1, 2], **Wang, F.** [1], **Liu Z.** [1, 2], **and Hu, S.** [1]

[1]State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan 430070, China
[2]School of Materials Science and Engineering, Wuhan University of Technology, Wuhan 430070, China
Email: fzhwang@whut.edu.cn

Abstract

Accelerated carbonation curing (ACC) is a promising approach for reducing the carbon footprint of the cement industry. γ-Dicalcium silicate (γ-C_2S) has emerged as a key material for ACC due to its high carbonation reactivity and low manufacturing energy. However, the influence of minor elements, such as Fe, on the reactivity of γ-C_2S remains poorly understood. This study employs density functional theory (DFT) calculations to systematically investigate the impact of Fe doping on the reactivity of γ-C_2S.

We explore the substitution sites and equilibrium mechanism of the impurity Fe in γ-C_2S by analyzing the formation energies of isolated point defects and defect complexes in different charge states. The results reveal that Fe substitution for Si is thermodynamically more favorable than substitution for Ca. This preference is attributed to the smaller influence range and bond difference when Fe replaces Si compared to Ca. The defect formation energy is found to be independent of the mass percentage of impurities, emphasizing the importance of using sufficiently large supercells when studying defects in calcium silicates.

Reactivity assessments are conducted through quantitative analyses of atomic charges and Ca-O bonds. The introduction of Fe impurities strengthens the nearest-neighbor Ca-O bonds and reduces the effective charge of O atoms, thereby inhibiting the reactivity of γ-C_2S. However, ab initio molecular dynamics (AIMD) simulations of γ-C_2S|H_2O interfaces reveal that hydroxylation and dehydroxylation of Fe-O groups can decrease the O coordination of surface Ca ions, promoting their dissolution (see Figure 1). The overall impact of Fe doping on γ-C_2S reactivity is found to be a balance between these opposing factors, depending on the specific reacting molecules. In pure H_2O systems, the inhibitory effect on reactivity dominates, while in systems containing H_2CO_3, the enhancement of reactivity prevails.

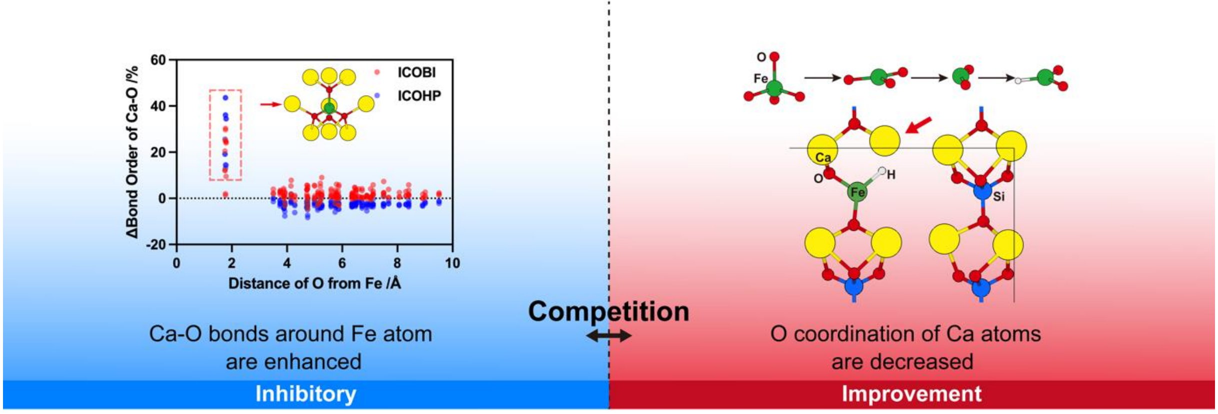


Figure 1 Impact of Fe doping on the reactivity of γ-C_2S

Experimental investigations support the AIMD findings, showing that Fe-doped γ-C_2S exhibits faster initial dissolution when in contact with water but releases fewer Ca ions overall compared to undoped γ-C_2S. The introduction of CO_2 significantly accelerates the reaction rate of Fe-doped γ-C_2S, due to the strong interaction between H_2CO_3 and Fe, which promotes the dissolution of Ca ions around the Fe impurity.

This study provides novel insights into the effects of ion doping on the reactivity of γ-C_2S. The findings contribute to the development of γ-C_2S-based carbonatable binders and the optimization of ACC processes for low-carbon building materials.

Keywords
γ-dicalcium silicate; Fe doping; reactivity; carbonation; density functional theory (DFT)

THE CARBONATION AND HARDENING PROPERTIES OF LARNITE, ÅKERMANITE AND MERWINITE IN STEEL SLAG: A STUDY FROM EXPERIMENTS AND DFT CALCULATIONS

Xinyu Zhang[1], **Donglin Li**[1], **Li Zhang**[1], **Xuemao Guan**[1], **Jianping Zhu**[1], **Songhui Liu**[1]

[1] Henan Key Laboratory of Materials on Deep-Earth Engineering, School of Materials
Science and Engineering, Henan Polytechnic University, Jiaozuo, 454003, China
Email: liusonghui@hpu.edu.cn

Abstract

Reducing anthropogenic CO_2 emissions remains a global challenge, with silicate minerals in steel slag (SS) significantly influencing its carbonation reactivity. This study synthesized three key mineral phases in SS - larnite, merwinite, and åkermanite (β-Ca_2SiO_4, $Ca_3MgSi_2O_8$, $Ca_2MgSi_2O_7$) - using solid-phase sintering. Their carbonation reactivity and hardening properties were systematically characterized, with intrinsic differences investigated using density functional theory (DFT). Carbonation reactions for all minerals concentrated in early stages, with 24-hour compressive strength of carbonated β-Ca_2SiO_4 reaching 84.70 MPa, significantly higher than $Ca_3MgSi_2O_8$ (37.89 MPa) and $Ca_2MgSi_2O_7$ (35.18 MPa). Mg substitution negatively impacted carbonation reactivity and altered products from calcite to Mg-calcite, aragonite, and magnesite. Theoretical simulations showed calcium silicates exhibit higher carbonation reactivity than calcium-magnesium silicates due to Ca/Mg-O bond energy differences. The carbonation reaction involves electrophilic and nucleophilic attacks on ions, with electron cloud localization varying in structures. $[Si_2O_7]^{6-}$ in $Ca_2MgSi_2O_7$ may affect ion dissolution during carbonation, potentially explaining its lower reactivity. This study provides valuable guidance for utilizing SS in high-performance CO_2 sequestration materials.

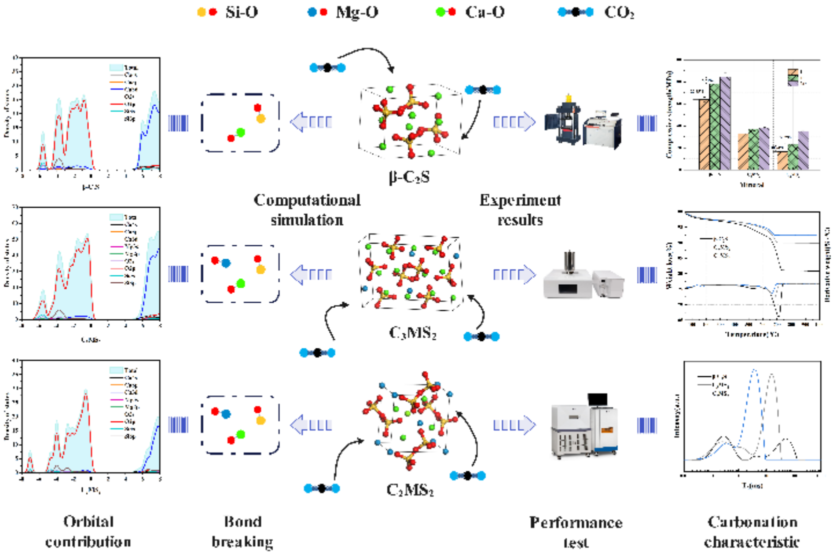


Fig. 1 Carbonation mechanism of three key mineral phases in SS

Keywords

Carbonation; steel slag; density functional theory (DFT); Ca/Mg-bearing silicate; CO_2 sequestration

NUMERICAL STUDY OF LIMESTONE PARTICLES CALCINATION IN A DROP TUBE FURNACE

Song, S.[1,2], **Park, D.G.**[1], **Hwang, J.**[2], **and Jeong, H.J.**[1]

[1]Korea Institute of Industrial Technology
[2]Yonsei University
Email: hyojae.jeong@kitech.re.kr

Abstract

In the context of dual fluidized bed gasifiers applying Sorption Enhanced Reforming (SER), optimal design of the combustion stage is crucial for regenerating CaO to regain its CO_2 capture capability through calcination of $CaCO_3$ transferred from the gasifier. This study develops a numerical analysis model based on ANSYS Fluent to design the combustor (riser) in a dual fluidized bed biomass gasifier using the limestone as a sorbent. The CFD model incorporated a calcination reaction for limestone particles using the Uniform Conversion Model (UCM) and employed the Langmuir isotherm equation to account for CO_2 partial pressure effects. The external and Pore diffusions of particles were also integrated. The numerical model describing calcination were implemented in ANSYS Fluent via user-defined functions

In this study, the developed CFD model was validated against experimental data from the drop tube furnace (DTF) at INCAR-CSIC, which is 6.5 m long with an inner diameter of 80 mm. The maximum error rate was about 0.2% when comparing the CFD results with experimental data based on the mole fraction of CO_2 at positions 1.0 m and 2.5 m from the $CaCO_3$ inlet. The conversion rates of 2,840 limestone particles over residence time in an air atmosphere, demonstrating similarity between numerical analysis and experimental results. The residence time of limestone particles in the DTF was calculated to be approximately 4 seconds, and it was observed that the slope of the conversion rate with respect to residence time increases with rising reaction temperature.

Keywords

Calcination; $CaCO_3$; sorption enhanced reforming; CFD; diffusion

NUMERICAL ANALYSIS OF CaO PARTICLES CARBONATION

Choi, H.S.[1,2], **Park, D.**[1], **Hwang, J.**[2], **and Jeong, H.J.**[1]

[1]Korea Institute of Industrial Technology
[2]Yonsei University
Email: hyojae.jeong@kitech.re.kr

Abstract

Many countries face the imperative to minimize greenhouse gas emissions while mitigating economic repercussions, aiming to achieve carbon neutrality by 2050. In recent years, hydrogen has emerged as an eco-friendly fuel, with significant interest focused on its production methods. Sorption-enhanced reforming (SER) is a technology applicable to dual fluidized bed (DFB) gasifiers to enhance hydrogen yield, utilizing limestone as a sorbent material. This study provides a numerical analysis approach for the carbonation reaction between calcium oxide (CaO) particles and CO_2 in the design of a DFB gasifier. The carbonation reaction rate was calculated using the Random Pore Model (RPM) in CFD modeling, considering the pore structure and external diffusion effects of CaO particles. The driving force due to CO_2 partial pressure in the gasifier and the diffusion effects from the $CaCO_3$ product layer formed during the carbonation reaction were also considered. The equations describing carbonation were implemented in ANSYS Fluent via user-defined functions.

In this study, the developed CFD model was validated by experimental data from the drop tube furnace (DTF) at INCAR-CSIC, which is 6.2 m long with an inner diameter of 100 mm. The maximum error rate was 3.40% when comparing the CFD results with experimental data based on the mole fraction of CO_2 at positions 2.4 m and 5.4 m from the CaO inlet. The conversion of 1,600 CaO particles over residence time at CO_2 mole fractions of 0.05, 0.15, and 0.2 demonstrated that the numerical analysis results were similar to the experimental data.

Keywords
Carbonation; CaO; sorption enhanced reforming; CFD; random pore model

Session C1

CO_2 Activation of Metallurgical Slag

CO-CARBONATION BEHAVIORS OF METALLURGICAL SLAG-CEMENT IN AQUEOUS CONDITIONS

Wang, Xue[1,2], **Wang, Qiang**[2]

[1] School of Civil and Resource Engineering, University of Science and Technology Beijing, Beijing 100083, China
[2] Department of Civil Engineering, Tsinghua University, Beijing, 100084, China
Email: w-qiang@tsinghua.edu.cn

Abstract

Blast furnace ferronickel slag (BFNS) and argon oxygen decarburization stainless steel slag (AODS) have the potential to sequester CO_2 for building materials. Carbonation kinetics of BFNS and AODS, as well as the effects of cement on the carbonation behaviors of them in slurry condition are studied in this research.

The results show that co-carbonation of 70% BFNS and 30% cement (B70C30) shows higher CO_2 uptake capacity (20.57%) than the theoretical value (16.93%). While co-carbonation of cement and AODS (A70C30, 70% AODS + 30% cement) has little effect on their carbonation capacity. Carbonation products of BFNS and AODS are calcite and metastable monohydrocalcite. Addition of cement transfers the carbonation products from monohydrocalcite to relatively more stable low-Mg calcite. Therefore, addition of cement is good for stability of the formed carbonation products. Besides, it was found that carbonation reactivity of Mg can be promoted by co-precipitation of Ca and Mg when two conditions are met: (1) containing enough Ca; (2) leaching more Mg. In B70C30, cement provides more Ca and promotes leaching of Mg for BFNS, so carbonation capacity is enhanced. However, in AODS, the above two conditions are already met, which makes cement less effective in promotion of carbonation capacity in A70C30 system.

Carbonation kinetics showes that pure BFNS and cement reaches almost complete carbonation after 7h, while B70C30 takes 20h to reach a stable state, resulting from the fact that Mg calcite forms in B70C30 at a low rate, due to incorporation of Mg and calcite crystal structure during growth. Besides, co-carbonation of AODS and cement accelerates the early carbonation and slows down the later reaction rate, as Mg calcite forms in A70C30 at a low rate when pH drops to below 10 at 11h.

This study provides theoretical guidance for AODS-cement and BFNS-cement CO_2 capture to make building materials.

Keywords

Blast furnace ferronickel slag; argon oxygen decarburization stainless steel slag; cement; monohydrocalcite; Mg calcite; co-precipitation

HYDRATION AND CARBONATION REACTIONS OF NATURAL HYDRAULIC LIME UNDER DIFFERENT CO_2 CONCENTRATIONS

Conghao Shao[1], Ze Liu[1], Zhiyuan Xu[1], Beibei Wang[1], Dongmin Wang[1], Junxia Li[1], Liguizu Liang[1]

[1]School of Chemical and Environmental Engineering, China University of Mining and
Technology, Beijing 100083, China
Email: liuze@cumtb.edu.cn

Abstract

Natural hydraulic lime (NHL) is a type of cementitious material that has both water-hardening and air-hardening properties. Its main constituents are C_2S and $Ca(OH)_2$ and it is typically produced within a temperature range of 800 to 1200°C. Unlike cement-based materials, carbonation is a major contributor to the mechanical performance of NHL. The $Ca(OH)_2$, C_2S and the hydration product C-S-H gel in NHL all exhibit good carbonation reactivity. In addition, as a lime-based binder, NHL paste is characterized by its porous and breathable nature, which facilitates the diffusion of CO_2 during the carbonation process. As a result of these properties, NHL exhibits higher carbonation reactivity, greater CO_2 diffusion efficiency and lower production energy consumption compared to cement-based materials. These advantages make NHL a superior carbon sequestration cementitious material compared to conventional cement-based materials.

In this study, using NHL5 as the primary material, samples with different degrees of hydration were prepared by varying the steam curing time of the NHL5 paste at 60°C (1 day, 3 days and 7 days). These samples are then subjected to carbonation curing under conditions of 3±1% CO_2, 25±3°C and 70±5% relative humidity. The objectives of the research are to investigate the mechanisms of influence of changes in different component contents on the carbonation reaction, to identify the preferential carbonation reaction of different carbonatable components within the system, to elucidate the carbonation reaction mechanisms of NHL5 paste at different hydration levels, and to provide a detailed explanation of the microstructural and phase composition changes that occur during the carbonation process. The experimental results indicate an inverse relationship between the degree of hydration and the diffusion range of CO_2 within the paste; higher hydration levels result in greater difficulty for CO_2 to diffuse into the paste. There is also a direct correlation between the degree of hydration and the compressive strength of the carbonated samples. For identical carbonation reaction times, samples with higher hydration levels exhibit greater compressive strength. After 28 days of carbonation, the compressive strengths of the samples subjected to 1 day, 3 days and 7 days of steam curing (S1C28, S3C28 and S7C28) were 30.5 MPa, 42.9 MPa and 46.2 MPa respectively. The quantitative XRD, TG-DTG and ^{29}Si NMR results at different carbonation times show that the content of the hydration product C-S-H increases with the degree of hydration. Samples with higher C-S-H gel content show higher degrees of carbonation for the same carbonation time. Combined with the conclusion that higher hydration levels make it more difficult for CO_2 to diffuse into the paste, it is evident that the carbonation reactivity of the components in the system follows the order: $Ca(OH)_2$ > C-S-H gel > C_2S. As the carbonation reaction progresses, the carbonation product $CaCO_3$ and silica gel continuously refine and fill the pores, thereby increasing the strength of the system. Based on the above, it is observed that as the degree of hydration increases, the C-S-H gel content in the system increases while the C_2S content

decreases. This results in a denser sample which affects the diffusion of CO_2. However, C-S-H gel has a higher carbonation activity compared to C_2S, resulting in better CO_2 sequestration efficiency and improved mechanical properties in samples with higher hydration levels.

In this study, a combined method of steam curing and carbonation curing was used to fully activate the hydration potential of NHL and to exploit its excellent carbonation reactivity. The result was the development of carbonated lime products with favorable mechanical properties. In addition, the study evaluated the specific effects of different hydration levels on the accelerated carbonation performance and mechanical properties of NHL and compared the reactivity of different carbonation components. The evolution of phase composition and microstructure during the carbonation process was elucidated, providing a theoretical basis for the application of NHL in the production of carbonated precast components.

Keywords
Natural hydraulic lime; carbonation; calcium hydroxide; dicalcium silicate; calcium silicate hydrate; calcite; CO_2 storage

UTILIZATION OF LOCAL RAW MATERIALS AND MINE WASTE TO MANUFACTURE CEMENT IN THE NORTHWEST TERRITORIES, CANADA

Huang, G.[1,2,4], Lambiv Dzemua, G.[3], Cairns, S.[3], and Liu, W.V.[2]

[1] State Key Laboratory of Geomechanics and Geotechnical Engineering, Institute of Rock and Soil Mechanics, Chinese Academy of Sciences, Wuhan, 430071, China
[2] Department of Civil and Environmental Engineering, University of Alberta, 9211 116th Street, Edmonton, Alberta, Canada T6G 1H9
[3]Northwest Territories Geological Survey, 4601 52nd Ave, Yellowknife, Northwest Territories, Canada X1A 1K3.
[4]IRSM-CAS/HK PolyU Joint Laboratory on Solid Waste Science, Wuhan, 430071, China
Email: victor.liu@ualberta.ca

Abstract

Currently, all the cement consumed in the Northwest Territories (NWT), Canada, is purchased from other provinces (e.g., Alberta) and shipped with long-distance (~ 1,800 km) truck freight. Transporting cement over long distances not only raises its cost, but also results in a higher carbon footprint. Locally producing cement is a potential low-carbon and economic solution for the local industry. However, it is unknown if the local raw materials are suitable for cement manufacturing; and there is a lack of a critical raw material—Iron ore—for cement manufacturing. Instead of iron ore, there are iron-rich tailings from a local rare earth element mine; the disposal of these tailings has been a significant challenge for the mining company. Towards low-carbon and circular economy, this study explored the use of local raw materials (i.e., limestone, clay, and gypsum) and mine waste (iron-rich tailings) to manufacture cement in the NWT and successfully produced the first bag of cement in the history of the NWT. Followed by cement manufacturing, the performance of NWT cement was evaluated and compared with commercial ordinary Portland cement (OPC) and standard specifications. The results showed that the chemical compositions of NWT cement met the Canadian standard specification for Portland cement. NWT cement-based samples had better workability and similar volume stability when compared with commercial OPC-based samples. Concrete samples made with NWT cement achieved comparable strength of commercial OPC-based concrete. For example, the 7 days' unconfined compressive strength (UCS) of NWT cement-based concrete reached 39.8 MPa, being 8.4% higher than that of commercial OPC-based concrete. In addition, an emission estimation suggested that locally producing cement in the NWT has the potential to reduce 3.0% - 61.7% of CO_2 emissions when compared with importing cement from other provinces. In conclusion, the raw materials and mine waste from NWT are suitable for cement manufacturing. Utilization of local raw materials and mine waste to locally produced cement can help reduce the carbon footprint related to cement consumption in the NWT and provide a sustainable solution for mine waste management. In addition, it also can help increase the value of local geological materials and diversify the local economy.

Keywords

Mine waste; cement manufacturing; concrete; circular economy; cementitious materials

IN-SITU WET CARBONATION OF STEEL SLAG POWDER PASTE MADE WITH CARBONATED WATER: INTERACTION MECHANISM BETWEEN CARBONATION AND HYDRATION

Wei Wang[1], **Zedi Zhang**[1], **and Yamei Zhang**[1,2]

[1]School of Materials Science and Engineering, Jiangsu Key Laboratory of Construction
Materials, Southeast University, Nanjing 211189, China
[2]Nanjing Institute for Intelligent Additive Manufacturing Co., Ltd, Nanjing 210000, China
Email: wang-wei@seu.edu.cn; 230228631@seu.edu.cn; ymzhang@seu.edu.cn

Abstract

The utilization of steel slag powder is severely limited by its low activity. This study reveals that in-situ wet carbonation with carbonated water can significantly enhance its hydration activity. The influencing mechanism of carbonation on the hydration process and its kinetics during hydration are explored, and on this basis, the interaction between carbonation and hydration is discussed. It is found that both hydration and carbonation exhibit distinct characteristics before and after the acceleration stage. Before the acceleration stage, the hydration rate is affected by mineral dissolution rate, which is dependent on carbonation reaction, with carbonation rate determined by the CO2 concentration. After entering the acceleration stage, the hydration rate is promoted by the increased nucleation effect (e.g., nano-CaCO3 precipitation). The hydration in this period dominates carbonation by controlling mineral dissolution rate, with carbonation rate dependent on the mineral dissolution rate. As a result, mineral dissolution is shown to act as a bridge between hydration and carbonation interactions. Based on these results, the lack of further increase in hydration activity of steel slag powder with higher concentrations of carbonated water is also explained. This study provides a deep understanding for wet carbonation in steel slag powder paste.

Keywords

Steel slag powder; in-situ wet carbonation; carbonated water; hydration mechanism; carbonation kinetics

TRANSFORM AOD SLAG TOWARD A HIGH-REACTIVE MINERAL ADMIXTURE WITH APPRECIABLE CO_2 SEQUESTRATION

Mo, L.W.[1, 2] **and** **Liu, P.**[1]

[1]College of Materials Science and Engineering, Nanjing Tech University, Nanjing, 211800, China
[2]State Key Laboratory of Material-Oriented Chemical Engineering, Nanjing Tech University, Nanjing, 211800, China
Email: andymoliwu@njtech.edu.cn; pengliu94@njtech.edu.cn

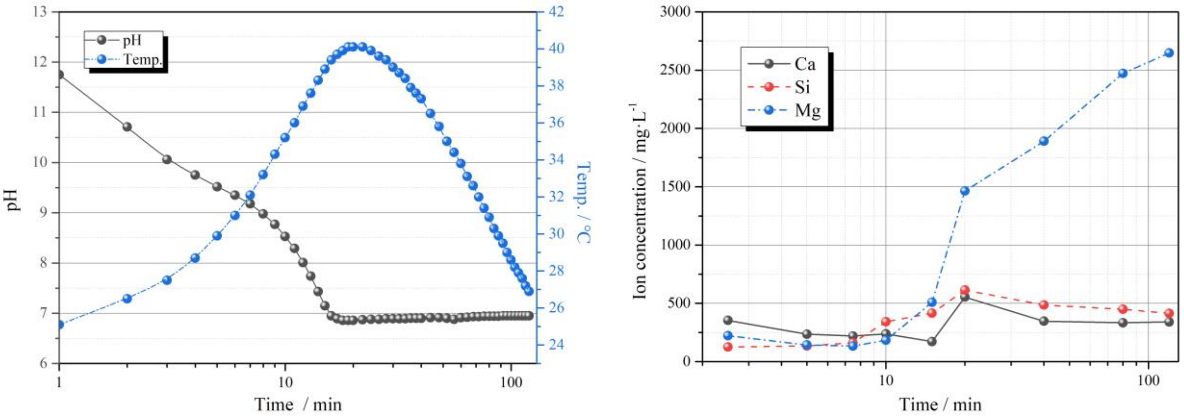
Abstract

Argon oxygen decarbonization slag (AODS) is a by-product of the secondary refining process of stainless steel [1]. The use of AODS as mineral admixture is an important way for its large-scale utilization. However, the unsoundness caused by excessive contents of f-CaO or f-MgO and the low hydration reactivity caused by the high contents of γ-C_2S and calcium/magnesium silicate minerals restrict its use as mineral admixture [2-4]. In this paper, the feasibility of transforming AODS toward a high-reactive mineral admixture with appreciable CO_2 sequestration were investigated. The evolution of the compositions and microstructures of AODS with carbonation time was systematically studied. The hydration behavior of carbonated AODS was revealed.

Results indicated that γ-C_2S and $Ca(OH)_2$ in AODS could quickly react with CO_2 to form $CaCO_3$. When the slurry pH dropped below 7.0, MgO and bredigite would also dissolve, leading to a significant increase in the concentration of Mg^{2+} in the slurry (Fig. 1). NMR results showed that the carbonation product of AODS also contains amorphous SiO_2 gel. As the carbonation time increased, the particle size of $CaCO_3$ gradually increased, from 100-200nm at 5min to 0.5-1μm at 20min (Fig. 2), and the content of SiO_2 gel also increased gradually, which constructed a porous microstructure, leading to an obvious increase in specific surface area of AODS. The CO_2 sequestration capacity of AODS can reach approximately 20g/100g.

The $CaCO_3$ and SiO_2 gel were involved in cement hydration to form monocarboaluminate and C-S-H respectively, which increased the hydration degree and compressive strength of composite cement. For instance, compressive strength of cement mortar mixed with 20wt.% carbonated AODS was increased by 25.8% compared with that of the mortar incorporating noncarbonated AODS (Fig. 3). This study provides a novel way to utilize the AODS to prepare high-reactive mineral admixture for being used as a replacement of cement.

Keywords
AOD slag; mineral admixture; CO_2 sequestration; microstructure evolution; hydration behavior



(a) pH value and temperature (b) ion concentration

Fig. 1 The evolution of pH value, temperature and ion concentration of the slurry during carbonation

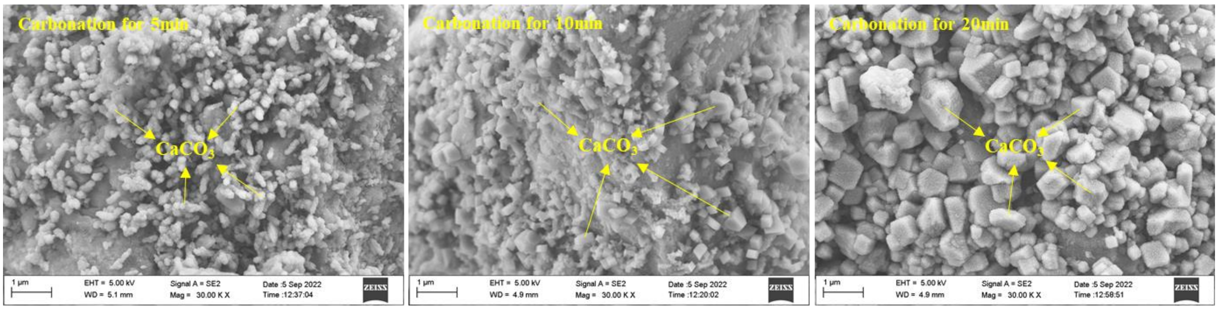


Fig. 2 SEM images of AODS with different carbonation time

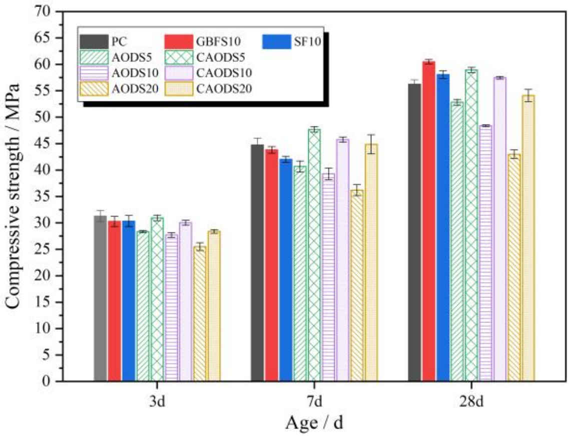


Fig. 3 Effect of carbonated AODS on the compressive strength of cement mortars

References

[1] H. Wang, G. Zhou, Y. Mu, M. Zhang, M. Guo, Enhanced carbon dioxide sequestration and Cr detoxification: Direct carbonation of AOD slag with additives under ambient conditions, Journal of Cleaner Production, 443 (2024) 141181.

[2] J. Zhao, Q. Liu, K. Fang, Optimization of f-MgO/f-CaO phase in ladle furnace slag by air rapidly cooling, Materials Letters, 280 (2020) 128528.

[3] O. Najm, H. El-Hassan, A. El-Dieb, Ladle slag characteristics and use in mortar and concrete: A comprehensive review, Journal of Cleaner Production, 288 (2021) 125584.

[4] T. Jiang, K. Cui, J. Chang, Development of low-carbon cement: Carbonation of compounded C_2S by β-C_2S and γ-C_2S, Cement and Concrete Composites, 139 (2023) 105071.

APPLICATION OF CARBONATION-BASED TREATMENTS TO VALORISE RESIDUES FROM EAF STEEL PRODUCTION

Masi, A.[1], **Alva Araujo, J.P.** [1], **Gubinelli, A.** [1], **Costa, G.**[1]

[1]Department of Civil Engineering and Computer Science Engineering, University of Rome
Tor Vergata, Rome 00133, Italy
Email: masi@ing.uniroma2.it

Abstract

Accelerated mineral carbonation has been proposed as an alternative and more sustainable way to manage the considerable amount of alkaline byproducts that are generated globally (around 7 billion tons per year (Renforth, 2019)) from different industrial sectors such as steel and concrete production, waste incineration, construction and demolition and mining. While these residues are currently mainly landfilled or under-utilised (Gomes et al., 2016), carbonation processes would allow to valorise them obtaining marketable products to be used, according to the selected treatment route, for several industrial applications, while at the same time permanently storing CO_2 in a solid form within the carbonates that originate from the reactions between Mg and especially Ca oxides-bearing reactive species, which are commonly found in alkaline waste, and carbon dioxide (Woodall et al., 2019).

The study we propose to present at the Conference is being carried out within the activities of the ongoing Rome Technopole project, funded by the Italian National Recovery and Resilience Plan, aimed at promoting the creation of a virtuous system within the Lazio Region for universities, national research centres, institutions and business associations to cooperate to tackle some critical and heavily-debated issues such as that of decarbonisation and circular economy (Fondazione Rome Technopole, 2024).

In this framework, two waste materials deriving from an EAF steel mill were considered: a coarse fraction, with dimensions ranging approximately from 2 to 13 mm, and a fine fraction, characterised by particles with a dimension below 0.06 mm in diameter. The residues were first characterised from an elemental, mineralogical and environmental point of view, and then subjected to preliminary carbonation tests considering enhanced wet and slurry carbonation conditions, in order to understand their reactivity with CO_2. Additional carbonation tests proved that the coarse sample only reacted when previously milled, which was not considered a feasible option, given that it was proposed to use this material as a substitute for natural aggregates. The fine sample, instead, presented interesting characteristics to be employed in further carbonation-based treatments considering both direct and especially indirect routes. For the direct route, carbo-granulation processes were tested to obtain aggregates, which were later characterised according to their CO_2 uptake, environmental behaviour and mechanical strength.

Further experiments are currently being carried out considering the indirect route, in order to obtain pure precipitated calcium carbonate. In particular, the objective of this specific phase of the study is that of testing, alongside with more traditional pH-swing processes, that are usually studied in the case of indirect carbonation, also more innovative treatments for both the Ca-extraction and the carbonation steps to reduce the use of reagents and improve the sustainability of the treatment.

Keywords
Carbonation; CO_2; industrial alkaline residues

References

Fondazione Rome Technopole. (n.d.). Retrieved 28 May 2024, from https://www.rometechnopole.it/en/

Gomes, H. I., Mayes, W. M., Rogerson, M., Stewart, D. I., & Burke, I. T. (2016). Alkaline residues and the environment: A review of impacts, management practices and opportunities. Journal of Cleaner Production, 112, 3571–3582. https://doi.org/10.1016/j.jclepro.2015.09.111

Renforth, P. (2019). The negative emission potential of alkaline materials. Nature Communications, 10(1), 1401. https://doi.org/10.1038/s41467-019-09475-5

Woodall, C. M., McQueen, N., Pilorgé, H., & Wilcox, J. (2019). Utilization of mineral carbonation products: Current state and potential. Greenhouse Gases: Science and Technology, 9(6), 1096–1113. https://doi.org/10.1002/ghg.1940

DECODING CARBONATED STEEL SLAG: A VISUALIZATION STUDY OF THE BARRIER LAYER

Linshan Li[1, 2], **Xiaojian Gao**[1], **and Guoqing Geng**[2]

[1]School of Civil Engineering, Harbin Institute of Technology, Harbin 150090, China
[2]Department of Civil and Environmental Engineering, National University of Singapore, 117576, Singapore
Email: Xiaojian Gao (gaoxj@hit.edu.cn) or Guoqing Geng (ceegg@nus.edu.sg)

Abstract

Carbonation treatment is widely acknowledged as an effective method for enhancing the volume stability of steel slag. However, the mechanisms behind the deactivation of steel slag through continuous carbonation treatment remain underexplored. This study employs innovative visualization techniques, including BSE-SEM, AFM, 3D X-Ray, and FIB-TEM, to elucidate the structural changes in carbonated steel slag. The results reveal that carbonated steel slag comprises four distinct layers: calcite, amorphous SiO_2, Ca_2SiO_4-twinning, and an uncarbonated Ca_2SiO_4 core. The "barrier layer" of calcite and amorphous SiO_2 constitutes up to 84.80% of a slag particle's volume. Compared to the uncarbonated core, the barrier layer exhibits a 6.4-fold increase in surface roughness and a porosity of 1.62%. The volume expansion during carbonation induces lattice defects, leading to the accumulation of Ca_2SiO_4-twinning. This comprehensive analysis provides deeper insights into the carbonation mechanism of steel slag, facilitating its improved application.

Keywords

Steel slag; carbonation treatment; visualization; nanostructure

PREPARATION OF CARBON-NEGATIVE ARTIFICIAL LIGHTWEIGHT AGGREGATES BY CARBONATING SINTERED RED MUD (SRM): CO_2 SEQUESTRATION, MICROSTRUCTURE AND PERFORMANCE

Maochun Xu[1] **and Liwu Mo**[1,2]

[1]College of Materials Science and Engineering, Nanjing Tech University, Nanjing, Jiangsu, 211800, PR China
[2]State Key Laboratory of Materials-Oriented Chemical Engineering, Nanjing Tech University, Nanjing, Jiangsu, 211800, PR China
Email: andymoliwu@njtech.edu.cn

Abstract

To efficiently utilize the low-value sintering red mud waste (SRM) on a large scale, a CO_2 curing method was proposed for the activation of cement-free SRM artificial aggregates. The effects of different curing methods (CO_2 and air curing) and varying fly ash (FA) dosages on the properties of SRM artificial aggregates were explored. In the case of SRM artificial aggregates subjected to CO_2 curing, the calcium silicates depleted accompanied by the precipitation of calcite and aragonite. The carbonated SRM artificial aggregates exhibited a denser microstructure, with the pores of the SRM itself filled with carbonation products. For carbonated artificial aggregates composed entirely of SRM, up to 10.07 wt.% CO_2 was sequestered and in turn produced calcium carbonate, which resulted in a 149.0% enhancement in crushing strength and a 29.5 % reduction in water absorption compared to the similar aggregate under air curing. The introduction of FA led to an accelerated strength development in the carbonated artificial aggregates during subsequent air curing, attributed to the hydration between the carbonation products $CaCO_3$ and aluminates derived from FA to form carboaluminates. From an environmental perspective, carbonated SRM artificial aggregates was carbon-negative, presenting a cleaner and sustainable alternative to natural aggregates. The results of the study provided a feasible solution for the effective treatment of SRM on a large scale and can be used as a reference for subsequent studies.

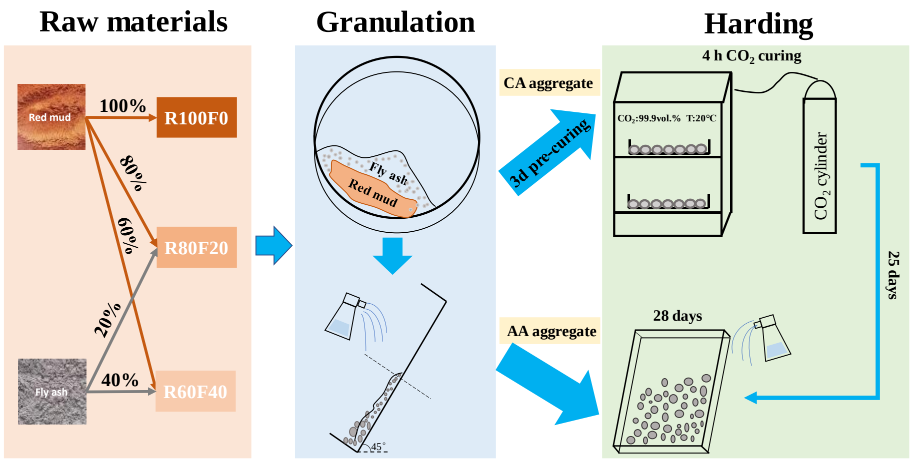


Fig. 1 Schematic process of the production of carbonated SRM artificial aggregates

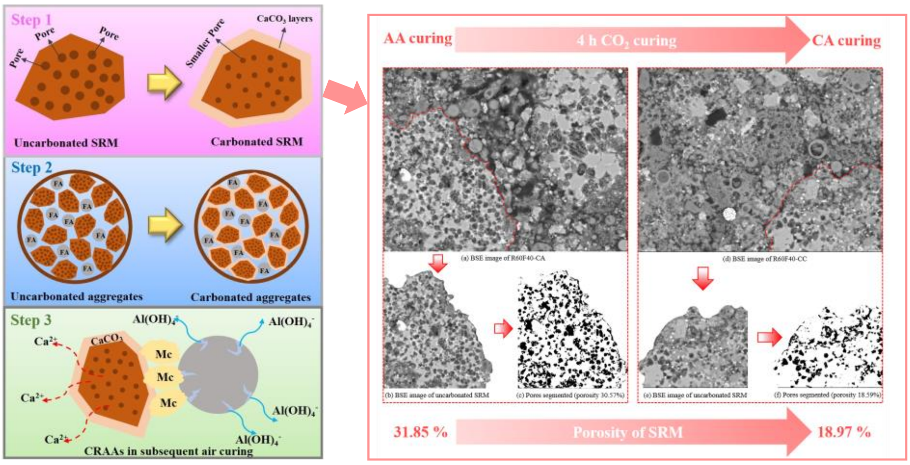


Fig. 2 Schematic of the mechanical property enhancement mechanisms of carbonated SRM artificial aggregates

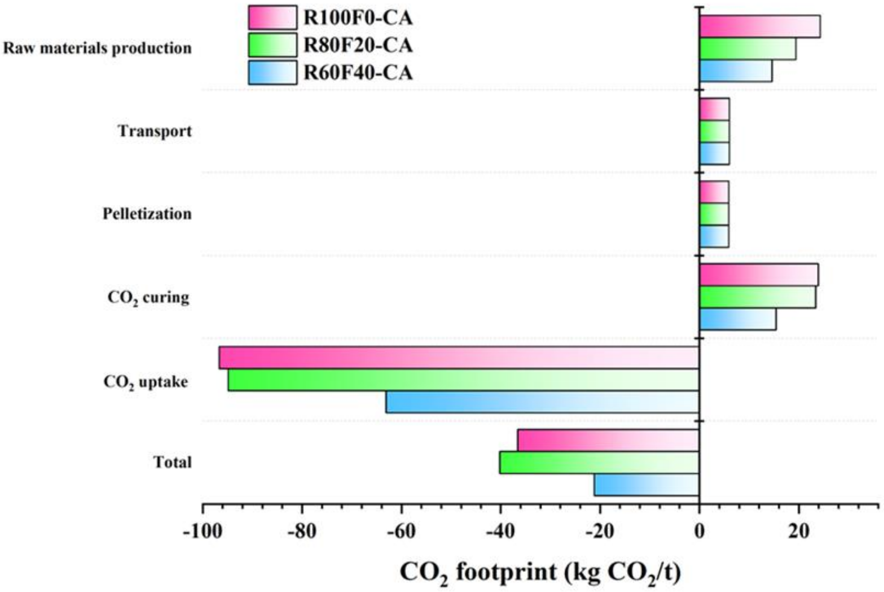


Fig. 3 CO_2 footprint of carbonated SRM artificial aggregates

Keywords
Red mud; artificial lightweight aggregates; CO_2 sequestration; microstructure; CO_2 footprint

MICROSTRUCTURAL STUDY ON THE CARBONATION OF GLYCINE-ACTIVATED STEEL SLAG

Seohyun Kim[1], and Juhyuk Moon[1,2]

[1] Department of Civil and Environmental Engineering, Seoul National University, Seoul 08826, Republic of Korea
[2] Institute of Construction and Environmental Engineering, Seoul 08826, Republic of Korea
Email: juhyukmoon@snu.ac.kr

Abstract

In this study, a novel method for using steel slag as a cementless binder was proposed. The steel slag pastes were hydrated under 40℃ for one day, and then carbonated for seven days. The addition of glycine accelerated the hydration of C_2S and C_2F, forming siliceous-hydrogarnet and AFm. During the carbonation, the hydration products and C_2S were carbonated to form $Al(OH)_3$ gel and various polymorphs of $CaCO_3$. Calcite was formed without the use of glycine while vaterite along with calcite were formed in the sample with 1% of glycine. Almost no crystalline $CaCO_3$ was precipitated when 3% and 5% of glycine was added. Glycine also induced the reaction of Mg ion in RO phase and hydrotalcite-like phase was formed. The compressive strength of steel slag pastes with 3% of glycine reached 77.8 MPa in seven days of carbonation.

Keywords

Steel slag; carbonation, glycine; raman microscopy

Introduction

Ordinary Portland Cement (OPC) is widely used as a construction material globally, but its production emits a substantial amount of CO_2 during the calcination process. Consequently, the need to identify alternative materials has become increasingly critical. Steel slag has emerged as a potential candidate to replace OPC; however, its low reactivity has limited its effective utilization. Nonetheless, steel slag contains reactive minerals, such as C_2S and C_2F, which possess latent hydraulic properties. Therefore, enhancing the reactivity of these minerals is crucial to maximizing the potential of steel slag in construction applications. The reactivity of C_2F can be improved through the use of various chemical activators, while C_2S exhibits significant carbonation reactivity. Additionally, metal ion chelating agents can be employed to further enhance the reactivity of C_2F. Carbonation curing, a method associated with carbon capture, utilization, and storage (CCUS), has gained attention as a promising technique for sequestering CO_2 through mineral carbonation. In this study, glycine, a chelating agent, was utilized as an activator for a steel slag-based binder, and carbonation curing was applied to investigate the resulting mechanical and microstructural properties.

Experimental

Table 1 shows the mix proportions of the steel slag pastes and curing conditions, and Figure 1 shows the schematic illustration of experimental procedures.

Table 1. Mix proportions and curing conditions of steel slag pastes

Sample type	Mix proportions			Curing conditions	
	Steel slag (g)	Glycine (g)	Water to binder ratio	0d - 1d	1d - 8d
GC0	100	0	0.2	**Hydration** 40°C	**Carbonation** CO_2 = 5% T = 20°C RH = 60%
GC1		1			
GC3		3			
GC5		5			

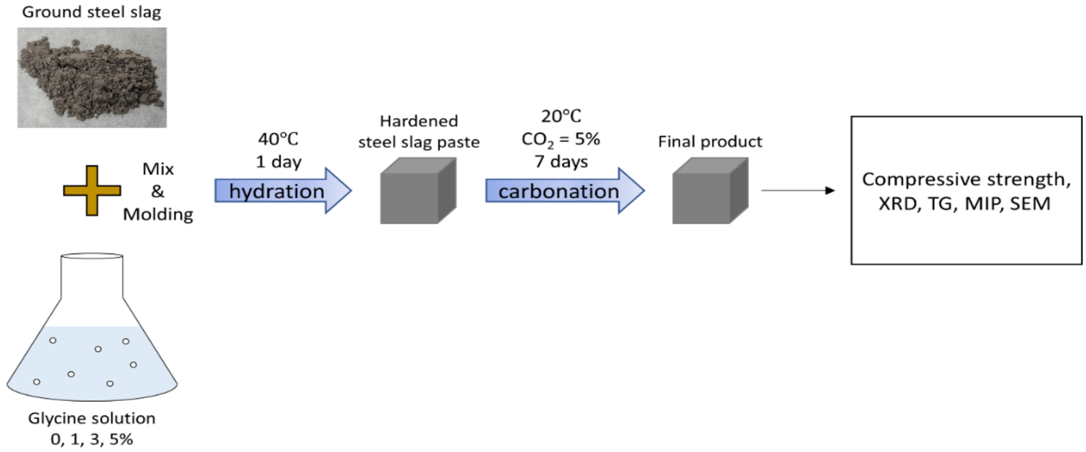


Figure 1. Schematic illustration of experimental procedures.

Results

Figure 2 illustrates the weight loss corresponding to the hydration products of steel slag pastes and decomposition temperature of $CaCO_3$ after 7 days of carbonation, as determined by thermogravimetric (TG) analysis. With increasing glycine content, the formation of hydration products such as AFm and hydrogarnet increases, while the $CaCO_3$ content decreases, accompanied by a reduction in its primary decomposition temperature.

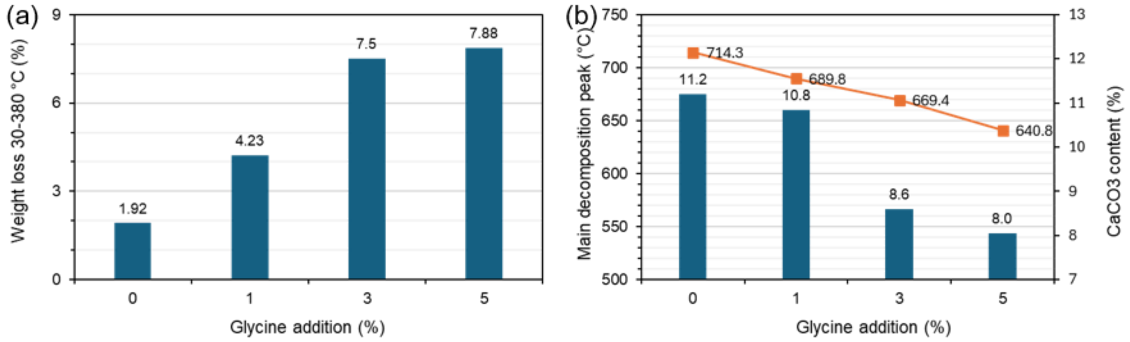


Figure 2. (a) Weight loss between 30°C to 380°C, (b) amount and decomposition temperature of $CaCO_3$.

Acknowledgement

This work was supported by the Technology Innovation Program (RS-2023-00266009, Development of admixture and blended cement using unused inorganic resource) funded By the Ministry of Trade, Industry & Energy (MOTIE, Korea).

Reference

[1] S. Kim, J. Kim, D. Jeon, J. Yang, J. Moon, Enhanced mechanical property of steel slag through glycine-assisted hydration and carbonation curing, Cement and Concrete Composites 149 (2024) 105532. https://doi.org/10.1016/j.cemconcomp.2024.105532.

IN-SITU CARBONATION OF BOFS BLENDED CEMENT PASTE VIA 13X ZEOLITE

Zhikai Wang[1], Xiaoli Wang[1], and Tung-Chai Ling[1]

[1] College of Civil Engineering, Hunan university Changsha, 410082, Hunan, China
Email: corresponding author tcling@hnu.edu.cn; tcling611@yahoo.com

Abstract

CO_2 curing of BOFS binder can improve material performance and offer environmental benefits. However, the application of this technology in BOFS faces a major challenge as the dense structure of the surface carbonation layer limits CO_2 diffusion and further carbonation. This study proposed an in-situ carbonation method to enhance the carbonation reaction within BOFS samples. Zeolite with a highly porous structure was employed as a carrier for CO_2 and incorporated in the BOFS samples.

In this study, 13X zeolite was chosen due to its high CO_2 adsorption capacity and can release the absorbed CO_2 in the presence of water. Initially, 13X zeolite particles were used to absorb CO_2 in a high-pressure reactor at 0.2 MPa. Subsequently, CO_2-13X zeolite particles were used to mix with cement and BOFS powder at a water-solid ratio of 0.4. It should be noted that cement accounting for 30% of the solid phase by weight, to achieve instant demolding. BOFS was partially replaced by pure 13X zeolite (denoted as Z) and CO_2-13X zeolite (denoted as CZ). For comparison, a control group comprising only BOFS and cement (denoted as Ref) was also prepared accordingly. After 24 h of casting, all samples were demolded and subjected to standard curing at 99% RH and 20°C until the testing day. The fresh property was assessed by flowability, hydration and heat measurement. The mineral phases and morphology changes of the pastes were characterized using X-ray Diffraction (XRD), scanning electron microscope (SEM), thermogravimetry analysis (TGA) and mercury intrusion porosimetry (MIP).

The results showed that the 13X zeolite can capture up to 18 wt.% CO_2 upon carbonation for 1 h. The flowability and finial setting time of the CZ samples decreased by about 30% and 20%, respectively, attributed to the growth of $CaCO_3$ on the particles and specific surface area by in-situ carbonation. At 1 d, the compressive strength and CO_2 uptake of Z and CZ samples were significantly improved compared with Ref. Interestingly, the 3 d compressive strength of both samples was about 8 times higher than that of Ref, indicating that 13X zeolite significantly improved the hydration between BOFS and cement. However, the 28 d compressive strength of Z decreased while that of CZ increased, probably due to the porous defects introduced by 13X zeolite and the filling effects of carbonates. In conclusion, it is feasible to utilize 13X zeolite as a CO_2 carrier to improve the early performance and carbonation of BOFS-blended cement pastes.

Keywords
In-situ carbonation; 13X Zeolite; basic oxygen furnace slag; compressive strength; CO_2 uptake

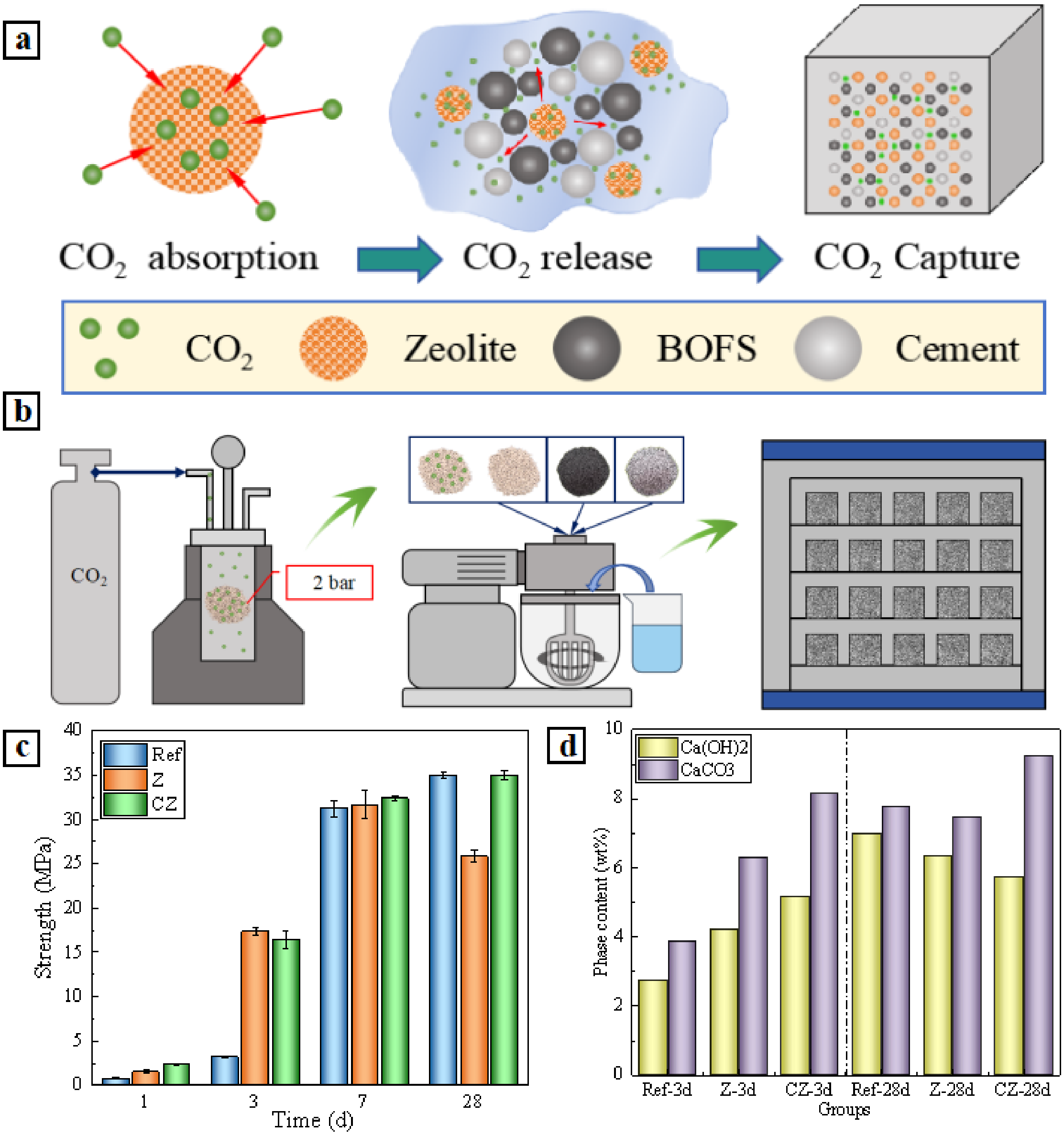


Figure 1 In-situ carbonation process (a), Sample preparation process (b), Compressive strength (c), $CaCO_3$ and $Ca(OH)_2$ content (d).

THE BALANCE BETWEEN HYDRATION AND CARBONATION WITHIN CARBONATION-ENHANCED AERATED CONCRETE: COMPARATIVE STUDY ON CURING REGIMES AND CARBONATION BINDER

Rui Sun[1], Peiliang Shen[1], and Chi-sun Poon[1]

[1]Department of Civil and Environmental Engineering & Research Centre for Resources Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hong Kong, China
Email: peiliang.shen@polyu.edu.hk; cecspoon@polyu.edu.hk

Abstract

Carbonation-enhanced aerated concrete (CAC) holds promise as the next generation of aerated concrete, offering a greener production process by substituting autoclave curing with carbonation curing. This study underscores the importance of balancing hydration and carbonation to optimize mechanical performance and CO_2-capture efficiency in CAC, with steel slag replacing conventional fly ash as a carbonation binder. Attaining a hydration degree of 35~45% of silicates in CAC is crucial for subsequent carbonation, ensuring effective pore filling with generated $CaCO_3$. This level of hydration can be achieved through 4 hours of steam curing at 60°C, initiating carbonation throughout the CAC matrix. Carbonation of C_2S promotes the formation of a rock-like dense pore wall structure. Consequently, unfilled pores in CAC with low hydration degree present challenges that carbonation only occurred on the surface of the isolated particles. While, the excessive consumption of C_2S with high hydration degree decreases the carbonates content and reduces strength.

Keywords
Steel slag; curing regime; carbonation-enhanced aerated concrete; carbonation; hydration

Session C2

CO_2 Activation of Magnesium-Containing Materials

UNDERSTANDING THE INFLUENCE MECHANISM OF MgO DOPING ON THE SINTERING AND PERFORMANCE OF TERNESITE-BASED CO_2 SEQUESTRATION BINDER

Xiaoyun Du[1], Jun Chang[1,2]

[1]School of Infrastructure Engineering, Dalian University of Technology, Dalian, 116024, China
[2]School of Civil Engineering and Architecture, Hainan University, Haikou 570228, Hainan, China
Email: duxiaoyun0202@qq.com (Xiaoyun Du); mlchang@163.com (Jun Chang)

Abstract

The use of low-lime CO_2 sequestration binder is considered to be an effective strategy to achieve greater CO_2 emission reduction in the cement industry. Ternesite has been proven to have significant competitiveness as an ultra-low lime CO_2 sequestration binder, making it worthy of industrial production. The purpose of this study is to simulate the effect of MgO content in natural limestone on the sintering and carbonation behavior of ternesite. The effects of sintering temperature and sintering duration on the mineral composition, microstructure and crystal structure of ternesite under different MgO doping levels will be studied. In addition, the carbonation behavior of different MgO doped ternesite clinkers will be further studied, including carbonation activity, CO_2 sequestration performance, mechanical properties, carbonation product differences and microscopic properties. This study will provide valuable insights into the quality control of raw materials and the prediction of product performance in the industrial production of ternesite.

Keywords
Ternesite; carbonation; compressive strength; magnesia limestone; crystal structure

MECHANICAL AND MICROSCOPIC PROPERTIES OF ORGANIC SOIL CARBONATED AND SOLIDIFIED BY REACTIVE MgO COMBINED WITH ISSA

Cai, G.H.[1], Guo, Z.Y.[2], Zhang, C.H.[1], Liu, S.Y.[2], and Poon, C.S.[3]

[1]School of Civil Engineering, Nanjing Forestry University, Nanjing 210037, P.R. China
[2]Institute of Geotechnical Engineering, School of Transportation, Southeast University, Nanjing 211189, P.R. China
[3]Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hong Kong 999077, China

Abstract

Dredging sediment has high organic matter (OM) and the weak geotechnical engineering characteristics, and it is difficult to be used directly for subgrade backfill. Among all the soil treatment technologies, the stabilization of Portland cement (PC) is the most widely-used, economical and easy-to-operate method. However, PC involves serious environmental pollution during its production, and it is not effective in treating organic soil. In the context, the carbonation technology of reactive MgO and sludge incineration bottom ash (ISSA) is used to treat organic-rich soils in this study, and a series of physical, mechanical, chemical and microscopic properties of the carbonated soils were tested under different OM contents and carbonation periods. The results showed that the unconfined compressive strength of the specimens increased with increasing binder content and carbonation time. After carbonation, mass enhancement and volume expansion were observed; moisture content values increased slightly; pH decreased while the conductivity increased. Based on XRD analysis, it was confirmed that the carbonation products such as dypingite/ hydromagnesite, and nesquehonite were the key to the strength enhancement, and a model of the reinforcement mechanism of the MgO-ISSA-treated soil was proposed. The results show that MgO and ISSA can be used as a consolidating agent for organic matter soils.

Keywords

Organic soil; reactive magnesia; incinerated sewage sludge ash (ISSA); mechanical properties; microscopic properties

TAILORING HIGH-MAGNESIUM CEMENTS FOR ENHANCED CARBONATION HARDENING AND CO_2 SEQUESTRATION

Donglin Li[1], Pengjie Rong[1], Songhui Liu[1], Peiliang Shen[2], Jian-xin Lu[2], Chi-Sun Poon[2]

[1]Henan Key Laboratory of Materials on Deep-Earth Engineering, School of Materials Science and Engineering, Henan Polytechnic University, Jiaozuo, 454003, China
[2]Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong
Email: liusonghui@hpu.edu.cn

Abstract

The reduction of CO_2 emissions from the cement industry remains a major challenge worldwide. This study investigates the feasibility of using high-magnesium limestone to prepare high-magnesium and low-calcium cementitious materials (HM-LCCs) with improved carbonation reactivity and CO_2 sequestration capacity. The carbonation reactivity, hardening properties, phase assemblage, microstructure, and pore distribution of HM-LCCs synthesized with varying proportions of high-magnesium limestone were characterized by XRD, TGA, FT-IR, SEM, and LF-NMR. It is found that with increasing substitution of high-magnesium limestone, the main mineral phases in clinker transformed from C_2MS_2 to C_3MS_2 and eventually to β-C_2S and MgO, along with markedly enhanced grindability. The carbonation reactivity and degree increased progressively with increasing substitution of high-magnesium limestone. Properly tailored HM-LCCs exhibited excellent strength development up to 94.56 MPa after carbonation, dense microstructure, and significantly higher CO_2 uptake. This study provides new insights into developing low-carbon cement by utilizing high-magnesian raw materials. The carbonation hardening and CO_2 sequestration capacity of cement can be optimized by tailoring the Mg/Ca ratio and calcium silicate phase assemblage.

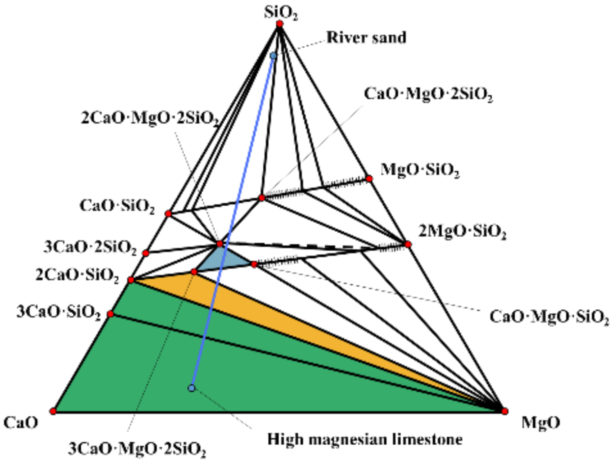


Fig. 1 Phase diagram of the MgO–CaO–SiO_2 system

Keywords
High-magnesium cement; carbonation hardening; CO_2 sequestration; mineral composition; microstructure

EFFECT OF TEMPERATURE AND CO_2 PRESSURE ON THE CARBONATION EFFICIENCY AND PRODUCTS OF MAGNESIUM SLAG

Jingang Han[1], Siyu Duan[1], Xinxing Zhou[1], Guangjun Lu[1], Zhibin Ma[1]

[1] Institute of Resources and Environmental Engineering, Shanxi University, Taiyuan 030006,
Shanxi, China
Email: mazhibin@sxu.edu.cn

Abstract

Magnesium metal is primarily produced using the Pidgeon process. For every ton of magnesium metal produced, 8-10 tons of magnesium slag (MS) are generated[1]. Approximately 8 million tons of MS are discharged annually in China, but they are presently under-utilized[2]. Currently, almost all of the MS are still dealt with through stockpiling and landfilling, leading to soil salinization and water pollution. Therefore, it is of great significance to find an effective way of using the MS to protect the local ecological environment.

The content of CaO in the MS generally exceeds 50%, and the CaO is mainly in the form of γ-dicalcium silicate (γ-C_2S). The γ-C_2S is inert in aqueous solution, but it can react with CO_2 in aqueous solution to generate $CaCO_3$[3]. Therefore, the key to study the carbonation of MS is to study clearly the carbonation process of C_2S in MS. Currently, most of the studies on the carbonation of γ-C_2S and β-C_2S are limited to their use as cementitious materials.[4] The effects of temperature and pressure during natural or accelerated carbonation on the amount of CO_2 fixation by C_2S and the mechanical properties of concrete are investigated.[5] However, the study of the whole carbonation process of powdered C_2S in solution system, including the effect of reaction conditions on carbonation efficiency as well as carbonation products and the whole kinetic process, is lacking at this stage. This study can be of great significance for the further development of C_2S-based materials.

This study proposed a novel method for preparing $CaCO_3$ with different micromorphologies and crystal kinds by the carbonation of MS. The effects of reaction time, temperature, and CO_2 pressure on the carbonation efficiency and the product characteristics were investigated. The kinetics of the carbonation reaction and the formation process of the $CaCO_3$ were revealed. Results show that the reaction temperature and CO_2 pressure are the most important factors affecting the carbonation efficiency of the MS and the product kinds. When the temperature is lower than 80 °C, the carbonation efficiency is less than 50% under atmospheric pressure, and a large number of unreacted γ-C_2S remains in the MS. Carbonation of MS under atmospheric pressure cannot achieve directional control over the crystal form of the product. Therefore, high temperature and pressure methods are required to further improve the carbonation efficiency of MS and achieve directional control over the product's crystal form.

The temperature has a large effect on the micromorphology of the product. The γ-C_2S and β-C_2S in the MS are completely transformed to $CaCO_3$ when the temperature is 140 °C or above. The needle-like aragonite with good crystallinity is obtained at 140 °C under pressurized condition. Needle-like aragonite crystals uniformly disperse with smooth surfaces, range in length from 1.52 μm to 12.83 μm, diameter from 0.8 μm to 1.24 μm, and aspect ratio between 5.71 and 16.04. The QXRD results for this condition indicate an aragonite percentage of 82.1%.

An increase in temperature facilitates the transformation from needle-like aragonite to block-like calcite. The block-like calcite dominates in the products when the temperature reaches 160 °C or above. When the temperature reaches 200 °C, the percentage of block-like calcite in the carbonation products reaches 95.8%. The micromorphologies of aragonite and calcite are shown in Figure 1 (a and b).

The kinetic fitting results of MS carbonation are shown in Figure 2. The formation process of $CaCO_3$ under pressurized carbonation can be described well by the surface coverage model, and the activation energy (E_a) for this reaction is approximately 34.36 kJ/mol. These indicate that the rate-controlling step for the carbonation process is the CO_2 diffusion. The optimal carbonation efficiency of the MS reaches approximately 89.8%, and the maximum CO_2 fixation amount reaches 459.4 kg/t. In the carbonation process, the SiO_4^{4-} in the C_2S reassembles into an amorphous SiO_2 with a three-dimensional network structure dominated by Q^4 and Q^3 structure.

The whole process is as follows: tiny grains of $CaCO_3$ are first formed on the surface of the MS particles. Under the pressurized conditions, CO_2 can pass through the $CaCO_3$ product layer into the particle and continue to react with γ-C_2S to form $CaCO_3$ crystals. Under the action of crystal growth stress, the formation of the inner $CaCO_3$ crystals causes the outer $CaCO_3$ product layer to fall off from the particle surface. These are conducive to the further carbonation reaction of the γ-C_2S inside the particles, thus greatly improving the carbonation efficiency. The whole carbonation process of the MS is similar to peeling an onion.

Keywords

Magnesium slag; CO_2 fixation; aragonite; calcite; carbonation reaction

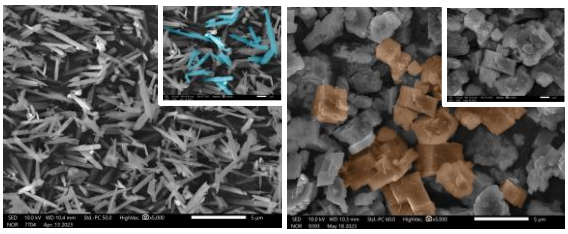


Figure 1. SEM images of the products obtained at the different temperatures. (a) 140 °C; (b) 200 °C.

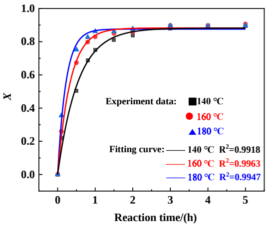


Figure 2. Fitting results based on the surface coverage model.

References

(1) Zhang, C.; Liu, S.; Tang, P.; Guan, X.; Shi, C. Enhancing the Hardening Properties and Microstructure of Magnesium Slag Blocks by Carbonation-Hydration Sequential Curing. *Journal of Building Engineering* **2023**, *76*, 107414. https://doi.org/10.1016/j.jobe.2023.107414.

(2) Zhong, K.; Huang, S.; Liu, Z.; Wang, F.; Hu, S.; Zhang, W. CO_2 Absorbing 3D Printable Mixtures for Magnesium Slag Valorization. *Construction and Building Materials* **2024**, *436*, 136894. https://doi.org/10.1016/j.conbuildmat.2024.136894.

(3) Yang, X.; Dong, F.; Zhang, X.; Li, C.; Gao, Q. Review on Comprehensive Utilization of Magnesium Slag and Development Prospect of Preparing Backfilling Materials. *Minerals* **2022**, *12* (11), 1415. https://doi.org/10.3390/min12111415.

(4) Zhao, S.; Liu, Z.; Wang, F.; Hu, S.; Liu, C. Effect of Extended Carbonation Curing on the Properties of γ-C_2S Compacts and Its Implications on the Multi-Step Reaction Mechanism. *ACS Sustainable Chem. Eng.* **2021**, *9* (19), 6673–6684. https://doi.org/10.1021/acssuschemeng.1c00200.

(5) Jiang, T.; Cui, K.; Chang, J. Development of Low-Carbon Cement: Carbonation of Compounded C_2S by β-C_2S and γ-C_2S. *Cement and Concrete Composites* **2023**, *139*, 105071. https://doi.org/10.1016/j.cemconcomp.2023.105071.

THE ROLE OF INTERNAL MOISTURE CONTENT (IMC) IN THE CARBONATION EFFICIENCY OF NATURAL FIBERS REINFORCED REACTIVE MAGNESIA CEMENT (NFs-RMC)

Bo Wu[1], **Jishen Qiu**[1]

[1] Department of Civil and Environmental Engineering, Hong Kong University of Science and Technology, Clear Water Bay, Hong Kong, China
Email: wubo@ust.hk

Abstract

The porous microstructure of natural fibers (NFs) enables the high-efficiency diffusion of CO_2 in reactive magnesia cement (RMC) regardless of the carbonation-induced densification of outer matrix. However, the high porosity and hydrophilicity of NFs make it very sensitive to the moisture, resulting in a high moisture absorption nature (e.g., 110%-240% of sisal fiber [1]). As shown in **Fig. 1**, the NFs can quickly absorb surrounding moisture to fill its lumens and micropores within 30 mins. As the CO_2 diffusivity in water is significantly lower than it is in air (2.1×10^{-5} vs. 0.14 cm^2/s [2]), the internal moisture content (IMC) largely determines the carbonation efficiency of NFs-RMC.

This work investigates the effect of IMC on strength development, hydration and carbonation products, microstructure, and CO_2 sequestration efficiency of NFs-RMC composites. Prior to elevated CO_2 curing, the NFs -RMC specimens are dried to different levels of nominal IMC (0% to 100%). Following the curing, the matrix composition was characterized with varying techniques (QXRD, TGA, FTIR, and acid digestion) for quantifying the CO_2 sequestration; the compressive strength at different depths was determined with uniaxial compressive test; the microstructure was examined with SEM/ESEM. The results show that the carbonation depth is increased by three to four times by reducing the IMC from 100% to 0%. The effects of varying IMC on local carbonation and strength are distinct in the shallow (0-20 mm) and middle/deep region (20-100 mm). In the shallow region, the CO_2 is easily accessible, hence the presence of moisture for CO_2 dissolution and RMC hydration determines the carbonation process, and the performances of matrix decreased with the decreasing IMC. By contrary, the CO_2 availability is the limiting factor in the deeper region, a reduction in IMC facilitates the inward diffusion of CO_2, thereby improving the matrix performances. These observations suggest that controlling the IMC is important to improve the CO_2 sequestration efficiency and mechanical performance, and shorten the curing time of NFs-RMC.

Keywords

Reactive magnesia cement (RMC); natural fibers; internal moisture content (IMC); carbonation products; CO_2 sequestration

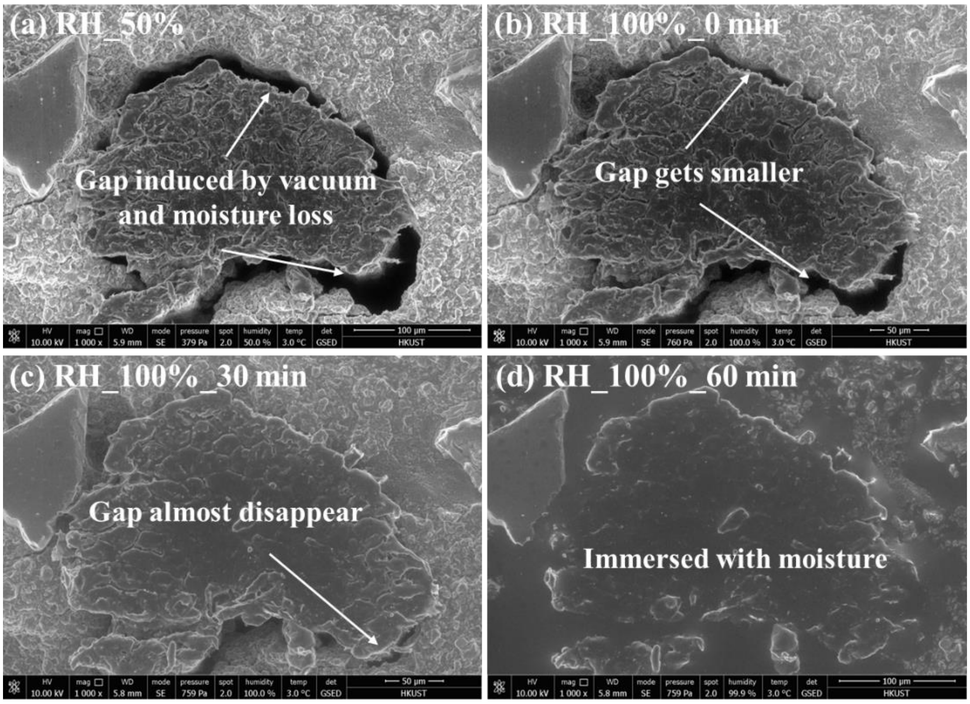


Figure 1. Environmental SEM images of sisal fiber cross-section in RMC matrix under different relative humidity. The sisal fiber can quickly absorb surrounding moisture to fill its lumens and micropores within 30 mins.

References

[1] N. Benmansour, B. Agoudjil, A. Gherabli, A. Kareche, A. Boudenne, Thermal and mechanical performance of natural mortar reinforced with date palm fibers for use as insulating materials in building, Energy and Buildings 81 (2014) 98-104.

[2] C. Hou, S. Gheorghiu, V.H. Huxley, P. Pfeifer, Reverse engineering of oxygen transport in the lung: adaptation to changing demands and resources through space-filling networks, PLoS Computational Biology 6(8) (2010) e1000902.

PREPARATION OF Mg-MODIFIED CO_2 SEQUESTRATION BINDER AND ITS CARBONATION BEHAVIORS

Peng, Yajuan[1], **Hang, Meiyan**[2], **Huang, Yongbo**[1], **Wang, Shoude**[1]*, **Chengxin**[1]

[1]Key Laboratory of Building Materials Preparation and Testing Technology, University of Jinan, Jinan250022, China
[2]Civil Engineering, Inner Mongolia University of Science and Technology, Baotou 014010, China

Email: 13589047192@163.com

Abstract

To convert solid wastes that contained magnesium into cementitious materials, a novel Mg-modified CO_2 sequestration binder (Mg-CSB) was prepared via Ca-stimulated olivine. Mg-CSB consists of a carbonizable composite mineral phases, such as the CaO-MgO-SiO_2-based minerals (shorted as $kC_{mn}S_n \cdot CMS$ (k=0, 2; m=0, 1, 2; n=0, 1)) and low-lime calcium silicates (CS, C_3S_2, and C_2S). The investigation involved the formation mechanism of $kC_{mn}S_n \cdot CMS$, as well as its effect on carbonation behaviors and performance of Mg-CSB in stirring and curing carbonation under ambient conditions. The results indicated that the $kC_{mn}S_n \cdot CMS$ would exist as $Ca_7Mg(SiO_4)_4$, $Ca_5Mg(SiO_4)_3$, $Ca_2MgSi_2O_7$, $Ca_3Mg(SiO_4)_2$, $CaMgSiO_4$, and $CaMgSi_2O_6$ by controlling the Ca/Mg ratio, which has good carbonation reactivity of that from high to low followed the aforementioned order, even the carbonation activity of $CaMgSi_2O_6$ is similar to that of γ-C_2S. This is due to a synergistic effect on carbon sequestration of calcium and magnesium, thereby the CO_2 uptake capacity of $kC_{mn}S_n \cdot CMS$ is up to 25.6% and 36.0% in three and twelve hours, respectively. The Mg-CSB has presented a good cementing property of compressive strength above 50 MPa, using solid wastes like magnesium slag for carbonation under ambient conditions.

Keywords
Ca-stimulated olivine; carbonation behaviors; CO_2 sequestration; binder; ambient conditions

CHARACTERISING REACTION PROCESSES OF SODIUM SULPHATE-ACTIVATED SLAG PASTE WITH MgO

Zhanhui Lu[1], Bo Li[1], Yung-Tsang Chen[1] and Fangying Wang[2]

[1]Department of Civil Engineering, University of Nottingham Ningbo China, 199 Taikang Road, Ningbo 315100, China
[2]Department of Civil Engineering, University of Nottingham, Nottingham NG7 2RD, United Kingdom
Email: bo.li@nottingham.edu.cn

Abstract

Sodium sulphate (SST)-activated slag shows better sustainability than the traditional sodium hydroxide and/or sodium silicate-activated slag due to its natural existence and low alkalinity. However, it has the shortcoming of slow mechanical performance development, limiting its application in the construction industry. Therefore, magnesium oxide (MgO) has been added to improve the mechanical properties of SST-activated slag by promoting hydration reaction and generating hydrotalcite phases to fill the pores. Simultaneously, adding MgO tends to increase the autogenous shrinkage since it accelerates the hydration reaction and consumes more internal water to increase capillary pore pressure, which causes early cracking of the components made with SST-activated slag. However, the reaction mechanism dominating the autogenous shrinkage of the SST-activated slag system has been not well understood yet, which necessitates further studies. Therefore, this paper investigates the reaction process of SST-activated slag paste with MgO to interpret its shrinkage behaviour.

The properties and autogenous shrinkage of SST-activated slag pastes were studied, including fluidity, setting time, compressive strength, hydration heat, and autogenous shrinkage. An accelerated hydration process with the addition of MgO is observed by hydration heat results, which also shortens their setting time. As a result, the SST-activated slag paste with a higher MgO content could achieve higher compressive strength (e.g., max. 33.91 MPa at 7 days). Increasing the SST concentration also increases the compressive strength of the paste. In addition, increasing the MgO content from 4% to 10% significantly increases the autogenous shrinkage of SST-activated slag. The autogenous shrinkage of SST-activated slag can be divided into slow and accelerated developing stages, which are highly influenced by MgO content. Increasing the MgO content from 4% to 8% shortens the slow developing stage by 49.6%, but increases the accelerated developing stage by 66.7%. This is potentially related to the expansive phase formation during the hydration process, which requires further studies to elaborate their mechanisms.

Keywords

Sodium sulphate; alkali-activated slag; magnesium oxide; autogenous shrinkage

INVESTIGATING THE CARBONATION-INDUCED VOLUME CHANGE IN REACTIVE MAGNESIA CEMENT

Quiatchon, P.R.[1], **Qiu, J.**[1]

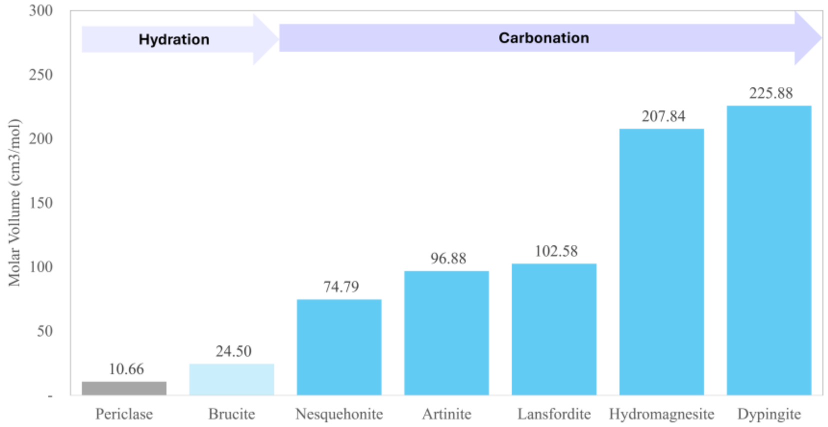
[1]Department of Civil and Environmental Engineering, The Hong Kong University of Science & Technology
Email: cejqiu@ust.hk; prjquiatchon@connect.ust.hk

Abstract

Reactive magnesia cement (RMC) hardens through hydration and carbonation, where it sequesters CO_2 from the air. The hydration and carbonation of RMC is an intrinsically volume-expansion process. A sample calculation for the changes in molar mass and molar volume during hydration and carbonation is provided below. The hydration product, brucite undergoes carbonation and forms hydrated magnesium carbonates (HMCs), which is the primary contributor to the strength of carbonated cement. Figure 1 shows the increase in molar volume from Periclase (MgO), to brucite ($Mg(OH)_2$), and to HMCs.

Hydration	$MgO + H_2O \rightarrow Mg(OH)_2$ $40.3 + 18 \rightarrow 58.32\ (mass)$ $10.66 + 18 \rightarrow \mathbf{24.5}\ (volume)$
Carbonation	$Mg(OH)_2 + CO_2 + 2H_2O \rightarrow Mg(CO_3)\cdot 3H_2O\ (Nesquehonite)$ $58.32 + 44.01 + 2(18) \rightarrow 138.36\ (mass)$ $24.5 + g + 2(18) \rightarrow \mathbf{74.79}\ (volume)$

Figure 1. Change in Molar Volume of RMC from Hydration to Carbonation



This work studies and quantifies the effect of carbonation on the volume change of RMC. Reactive magnesia cement (RMC) paste, without any additives, typically undergoes setting in 2 hours or less. The length at final setting was used as the reference point for measuring length change. After demolding at final setting, the RMC paste was cured in a high humidity chamber for 24 hours before undergoing accelerated carbonation (10% CO2, 23C, 80%RH). The length of the RMC specimens was measured at days 1, 3, 5, 7, 14, and 28 to quantify the volume change due to carbonation. The effect of factors such as Water-to-cement ratio, Water Reducer, Silica Fume (SF), Hydration Agent, and Natural Fiber was also examined.

Table 1. Mix Design

Factor	Mix ID	Proportions by mass of RMC					HA
		RMC	Water	WR	SF	NF	
Water-to-cement ratio	RMC-0.7	1	0.7				
	RMC-0.8		0.8				
	RMC-0.9		0.9				
Water Reducer (WR): Sodium Hexametaphosphate-$(NaPO_3)_6$	WR-1%	1	0.7	0.01			
	WR-2%		0.7	0.02			
	WR-3%		0.7	0.03			
	WR-4%		0.7	0.04			
Silica Fume (SF)– RMC	10%SF – 90% RMC	1	0.93		0.11		
	20%SF – 80% RMC		1.09		0.25		
	30%SF – 70% RMC		1.30		0.43		
	40%SF – 60% RMC		1.57		0.67		
Hydration Agent (HA): Magnesium Acetate	0.8MA0.05	1	0.8				0.05M
	0.8MA0.10		0.8				0.1M
Natural Fiber (NF) - Sisal Fiber	Sisal-0.5%	1	0.7	0.03		0.5	
	Sisal-1%		0.7	0.03		1	
	Sisal-1.5%		0.7	0.03		1.5	
	Sisal-2%		0.7	0.03		2	

Keywords

Reactive magnesia cement; carbonation; unrestrained length change

EFFECTS OF CO_2 CARBONATION ON THE PROPERTIES OF CSA-STEEL SLAG COMPOSITE CEMENTITIOUS MATERIALS

Xujiang Wang[1,2], Jialin Song[1,2], Yanghao Chen[1,2], Jingwei Li[1], Wenlong Wang[1]

[1]National Engineering Laboratory for Reducing Emissions from Coal Combustion, Shandong University, Jinan, Shandong, 250061, China
[2]Shenzhen Research Institute of Shandong University, Shenzhen, Guangdong, 518000, China
Email: x.wang@sdu.edu.

Abstract

China produces nearly a billion tons of steel slag annually. The CO_2 sequestration potential of steel slag can reach 99 to 135 kg per ton. However, the low reactivity of calcium silicate in steel slag leads to slow hydration-carbonation reaction rates, thereby limiting the improvement of product performance. Combining steel slag with cementitious materials contributes to the enhancement of strength and stability of steel slag carbonation products, while simultaneously increasing production efficiency and reducing production costs. Calcium sulfoaluminate cement (CSA), as a low-carbon cement, possess advantages of low energy consumption and rapid early strength development. If synergistically utilized for carbonation curing with steel slag, they would hold tremendous potential for applications. This study utilizes CSA cement and steel slag to prepare composite cementitious materials, investigating the effects of different ratios and carbonation curing conditions on the material properties and carbon sequestration capacity.

The results indicate that the extension of carbonation time exhibits a trend of initially decreasing and then increasing the compressive strength of the samples. Early carbonation curing can disrupt the AFt crystal structure, leading to a decrease in material strength. However, as the reaction progresses, the carbonation product calcium carbonate fills the pores, enhancing material density and promoting strength development. Increasing curing pressure can enhance the compressive strength of the samples to some extent, but the effect on improving the degree of carbonation is relatively limited. This may be related to the densification effect of carbonation products on material pores, blocking the CO_2 transport channels. Results of rresponse surface experiments indicate that the CSA content has the most significant effect on material strength, followed by curing temperature, and lastly, pre-curing time. Curing temperature is the most crucial factor affecting the degree of carbonation, followed by CSA content, with pre-curing time having the least impact. Ultimately, the optimized ratio and curing conditions for the composite cementitious material are determined to be: curing temperature of 70°C, pre-curing time of 12 hours, and CSA content of 26%. Under the optimal ratio and conditions, the compressive strength of the neat paste specimen reaches 24.7 MPa after 1 day, with a high carbonation degree of 37.86%. During subsequent natural curing, there is minimal change in the mineral phases of the specimens, but the crystallinity of carbonation products such as calcite and vaterite gradually increases, accompanied by a reduction in porosity. The microscopic structure becomes denser, leading to a continuous improvement in both strength and carbonation degree.

In summary, CSA-steel slag composite cementitious materials demonstrate excellent performance and significant carbon sequestration capability. Leveraging the advantages of CSA in enhancing early strength and reactivity, along with optimizing curing conditions and adjusting mineral composition, effectively enhances the performance of composite cementitious materials and expands their application scope. This provides technical support for the low-carbon development in the prefabricated components and modular construction sectors.

Keywords
Sulfoaluminate cement; steel slag; carbonization; pore structure; minerals

ENHANCED AQUEOUS CARBONATION OF BOFS FINE AGGREGATES EFFICIENCY USING Na_2CO_3

Haodong Lin[1], Qifeng Song[1], and Tung-Chai Ling[1]

[1]College of Civil Engineering, Hunan University, Changsha, 410082, Hunan, China
Email: tcling@hnu.edu.cn; tcling611@yahoo.com

Abstract

Accelerated carbonation is considered as a feasible method to mitigate volumetric instability of basic oxygen furnace slag (BOFS). However, BOFS with size of few millimeters is extremely inefficient to be carbonated due to the dense surface structure. Therefore, this study aims to enhance the carbonation efficiency of BOFS fine aggregates in aqueous route using Na_2CO_3.

The concentration of Na_2CO_3 solution was optimized at different carbonation temperature and duration to examine the $CaCO_3$ deposition depth development and properties of BOFS with different particle sizes.

The carbonation characteristics were compared among three groups: only CO_2 injection (0.3 L/min) (0.3CO), only Na_2CO_3 solution (0.5 mol/L) (0.5NC), and CO_2 injection in Na_2CO_3 solution (0.5NC+0.3CO). The results showed that the 0.3CO group showed lowest carbonation efficiency, the 0.5NC group 's efficiency gradually decreased due to the decline in CO_3^{2-} concentration, while the 0.5NC+0.3CO group achieved significantly higher carbonation efficiency and degree than the other two groups (see Fig. 1a). Na_2CO_3 solution not only enhanced CO_2 dissolution which maintains the CO_3^{2-} concentration, but also retains the solution pH above 8, providing a sustainable carbonation environment. Through a 2-hour continuous carbonation process, it was found that 0.5NC+0.3CO group reached its maximum CO_2 uptake within 60 minutes. Conversely, the CO_2 uptake of the 0.3CO group, despite ongoing increases, remained a significantly lower CO_2 uptake even after 120 minutes carbonation compared to the 0.5NC+0.3CO group after just 10 minutes (see Fig. 1c). Subsequently, the effect of temperature was investigated (see Fig. 1d). The 40 ℃ group demonstrated a higher carbonation rate and peak compared to the 20 ℃ group. However, the 80℃ group showed a turning point at 30 minutes, where prolonged carbonation was detrimental. Considering these factors and environmental considerations, the optimal carbonation condition was to inject CO_2 to 0.5 mol/L Na_2CO_3 solution at a flow rate of 0.3 L/min for 60 min at 40℃. Under such carbonation condition, the positive effect was become less obvious with increasing particle size of BOFS fine aggregates, yet the CO_2 uptake can still be increased to over threefold that of the 0.3CO group (see Fig. 1e). The experiment results also demonstrate the viability of recycling the Na_2CO_3 solution, which maintains its effectiveness after 3 cycles. Finally, the water absorption of the BOFS fine aggregates was reduced upon carbonation regardless of its particle sizes (see Fig. 1f).

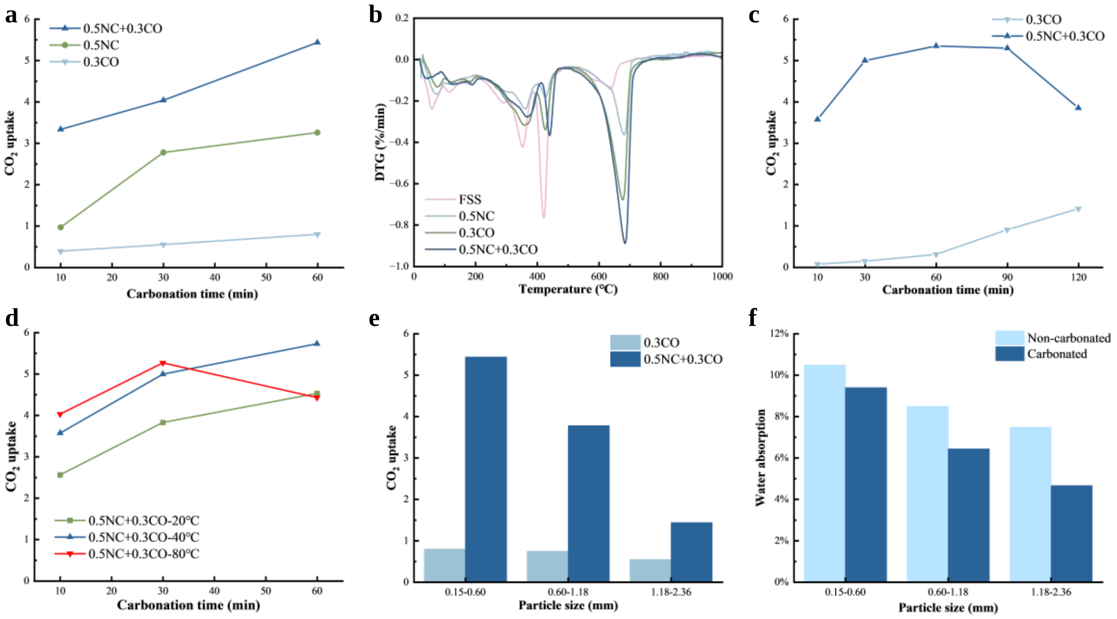


Fig 1. Results of the experiment: (a) The effect of carbonate initial concentrations; (b) TGA of fresh and carbonated BOFS fine aggregate; (c) The effect of carbonation duration; (d) The effect of carbonation temperature; (e) The effect of particle size; (f) Water absorption of non-carbonated and carbonated BOFS fine aggregate.

Keywords

BOFS; fine aggregate; aqueous carbonation; cement mortar; Na_2CO_3

BONDING AND MICRO-MECHANICAL PROPERTIES OF STEEL SLAG CARBONATION

Jie Li[1], Qifeng Song[1], and Tung-Chai Ling[1]

[1]College of Civil Engineering, Hunan University, Changsha 410082, China
Email: tcling@hnu.edu.cn; tcling611@yahoo.com

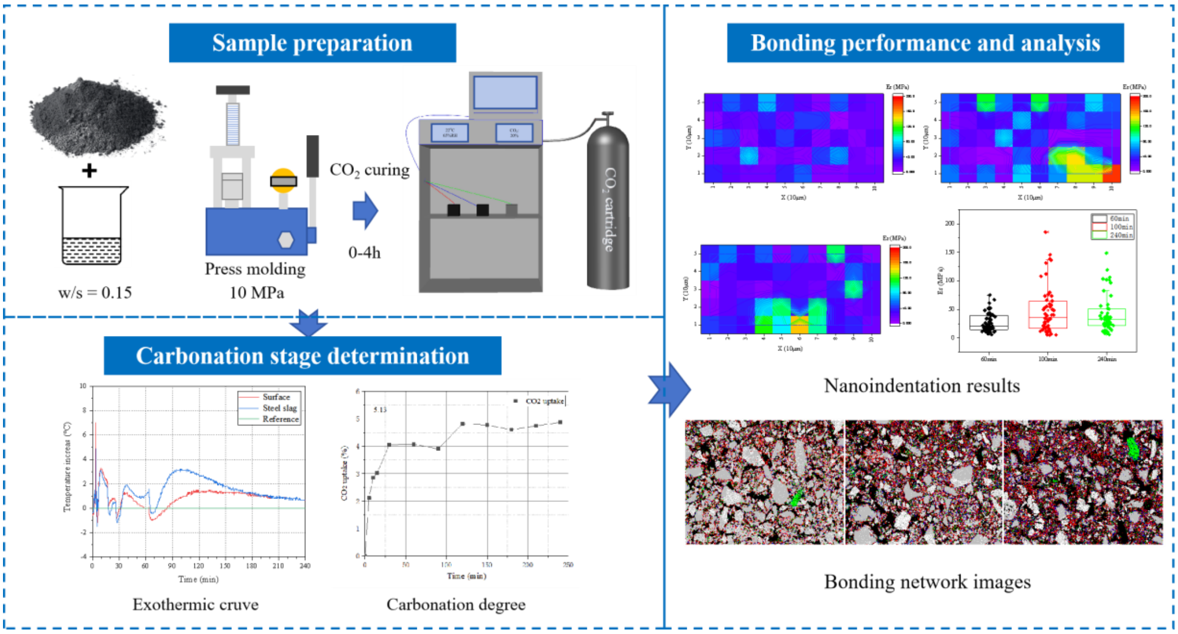
Abstract

The integration of steel slag with CO_2 for the production of construction materials offers a dual benefit of upcycling industrial waste and CO_2 generated from steel making industry. Studies demonstrate that the carbonation of steel slag compact products can achieve compressive strength as high as 69.9 MPa in 6 hours, and mainly related the strength development to the formation of $CaCO_3$. They claimed that small carbonate particles could fill the pores and densify the microstructure matrix. However, the bonding mechanism within carbonated steel slag remains unclear. This study aims to investigate the micro-mechanical properties changes and bonding mechanism of steel slag particles upon carbonation curing.

Carbonation stages were identified based on continuous exothermic monitoring and TGA methods. The micro-mechanical properties at each stage were then studied using nanoindentation. In combination with XRD, FTIR, and BSE examinations, the micro-mechanical properties of steel slag carbonation were elucidated. The results showed that carbonation could improve the contact modulus of steel slag particles. However, prolonged the carbonation time the contact modulus was gradually decreased. This phenomenon can be attributed to the characteristics of products network obtained by BSE. It could be found that $CaCO_3$ network was rapidly formed during carbonation, but the bonding within steel slag particles was relatively weak. The bonding of steel slag particles was mainly governed by the formation of Ca-modified silica gel network, which can effectively emerge the bonding of steel slag particles with the formed $CaCO_3$, leading to a stronger micro-mechanical property. However, the Ca-modified silica gel can also be carbonated with a continuous carbonation reaction, resulting in the decrease of Ca/Si ratio of the silica gel network. This eventually causes the decalcification of the silica gel network and poor micro-mechanical strength in the matrix. In summary, the development of bonding properties of steel slag particles is mainly related to the structure of the connecting network of carbonation products, and highlighting the importance of dynamic interplay between the $CaCO_3$ and the silica gel network in steel slag carbonation.

Keywords
Steel slag; carbonation curing; bonding mechanism; element analysis; nanoindentation

Graphical Abstract



FUNCTIONAL BIOCHAR FOR PERMANENT CAPTURE OF CO_2 AND ITS USE IN MORTAR BLOCK: A STEP TOWARDS CO_2 EMISSION TO CAPTURE

Razia Sultana[1], Shipeng Zhang[1], and C.S. Poon[1]

[1]Department of Civil and Environmental Engineering, The Hong Kong Polytechnic
University, Hong Kong SAR
Email: razia.sultana@connect.polyu.hk

Abstract

The construction industry faces significant challenges in achieving carbon neutrality, notably due to the high carbon dioxide (CO_2) emissions associated with cement production. Additionally, solid waste generation from Spent Coffee Grounds (SCG) is responsible for greenhouse gas emissions. A sustainable solution to handle these challenges lies in the innovative use of biochar. However, biochar production is not without its CO_2 emissions, which occur during biomass pyrolysis. This work captured and reintegrated the CO_2 released during functionalization into the biochar's structure. Initially, two varieties of pristine biochar (B300 and B450) were produced at temperatures of 300 ℃ and 450 ℃, respectively, from SCG. Subsequently, two types of functional biochar (B300 Ca and B450 Ca) were produced using a functionalising agent ($Ca(OH)_2$), which reacts with its emitted CO_2 inside a horizontal tube furnace to form $CaCO_3$ at 400℃. Through Thermogravimetric Analysis (TGA) and Quantitative X-ray Diffraction (Q-XRD) analyses, it was observed that a greater quantity of $CaCO_3$ formed at lower pyrolysis temperature and higher functionalisation temperature due to the subsequent CO_2 release from the SCG at this temperature. The pristine and functionalised biochar, at 3 wt.% and 6 wt.%, were then used as partial cement replacements to produce mortar blocks. The study revealed that the inclusion of both pristine and functionalised biochar increased the heat of hydration compressive strength and decreased the workability, wettability, and water absorption of the mortar block. Mortar blocks containing biochar produced at higher pyrolysis temperatures and lower functionalisation temperatures exhibited reduced compressive strength. The optimal conditions identified in this work were a pyrolysis temperature of 300℃ and a biochar dosage of 6%. It was also determined that maintaining a lower pyrolysis temperature relative to the functionalisation temperature is crucial for effective CO_2 capture and enhancing the compressive strength of the mortar blocks.

Keywords

Solid waste; CO_2 capture; functional biochar; sustainable construction; waste management

STUDY THE POZZOLANIC REACTIVITY OF Si-Al GEL WITH DIFFERENT Si/Al RATIO

Shunmin Xiao[1], Yi Jiang[1], Zihan Ma[1], Long Jiang[1], Jionghuang He[1], Huiling Wang[1], Peiliang Shen[1,2], Chi-sun Poon[1,2]

[1] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hong Kong, China
[2] Research Centre for Resources Engineering towards Carbon Neutrality,
The Hong Kong Polytechnic University, Hong Kong, China
Email: peiliang.shen@polyu.edu.hk

Abstract

Aluminosilicate gel is an important component of carbonated RCF (Recycled concrete fines) and steel slag and can provide reactivity to building materials. The nature of aluminosilicate gel produced through carbonation from different raw materials varies and affects the properties of the materials containing them. This study focuses on the pozzolanic reactivity of aluminosilicate gel with different Si/Al. Using a two-step carbonation process followed by HCl washing, aluminosilicate gels with varying Si/Al were synthesized from RCF and steel slag and characterized through FTIR, NMR, and BET analyses. The pozzolanic reactivity of these gels was assessed by examining the exothermic reaction with $Ca(OH)_2$ and analyzing the product compositions. Findings indicate that an increased Si/Al ratio enhances silicon polymerization within the gels, leading to greater specific surface area and porosity. The hydration products of Gel-0.004, Gel-0.166, and Gel-0.131 were primarily composed of C-(A)-S-H, with their 7-day hydration heat correlating positively with silica content. Gel-0.004 exhibited the fastest exothermic and hydration rates, while Gel-0.433, with the highest aluminum content, predominantly formed stratlingite and demonstrated higher 7-day hydration heat than Gel-0.131. These insights offer a theoretical foundation for the application of carbonated solid waste in building materials.

Keywords
Carbonation; recycled concrete fines; steel slag; aluminosilicate gel; pozzolanic activity

Session C3

Carbonation Additives

APPROPRIATE TECHNOLOGIES TO STIMULATE HYDRATION OR CARBONATION OF INDUSTRIAL BY-PRODUCTS

Juhyuk Moon[1]

[1]Department of Civil and Environmental Engineering, Seoul National University, Seoul
08826, Republic of Korea
Email: juhyukmoon@snu.ac.kr

Abstract

Conventional ordinary Portland cement (OPC) is one of most widely used manufactured materials which has been especially developed to maximize its hydraulic ability. However, due to the CO_2 crisis, the use of OPC should be reconsidered for achieving the net zero by 2050. There are actually many technologies of green cement but only a few of them have been successfully implemented. Main reason is due to the exceptionally cheap price of OPC. It is probably the cheapest manufactured materials on earth. Therefore, in this talk, appropriate technologies will be introduced which targets to replace OPC completely or partially by new types of industrial by-products. Direct utilization of CO_2 can be another option to stimulate its carbonation ability to permanently store CO_2 in the case where hydration compatibility of certain industrial by-product with OPC is poor. In this talk, the use of bottom ash or steel slag will be covered. These two industrial by-products are very abundant in South Korea as well as globally. With customized grinding agent for each industrial by-products, its reactivity index was similar to fly ash type F which is currently being recycled as supplementary cementitious materials globally. For the case of steel slag, single grinding step may be not enough to fully activate its hydraulic behavior so chemical additive of glycine was proposed to stimulate its carbonation behavior under CO_2 curing. With these appropriate technologies, its reactivity as construction materials can be engineered and contributes to the CO_2 reduction.

Steel slag, which contains reactive minerals such as C_2S and C_2F, has the potential to be a substitute for OPC. However, due to its intrinsic low reactivity, ground steel slag has not been recycled as supplementary cementitious materials like fly ash or ground blast furnace steel slag (GGBFS). Recent studies are focusing on enhancing the reactivity of C_2F with chelating agents and leveraging the carbonation reactivity of C_2S. This study explores the use of glycine as an activator for steel slag and examines its mechanical and microstructural properties under carbonation curing. Left Figure 1 shows the compressive strength development of carbonated steel slag (Kim et al., Enhanced mechanical property of steel slag through glycine-assisted hydration and carbonation curing, *Cement and Concrete Composites*, 2024).

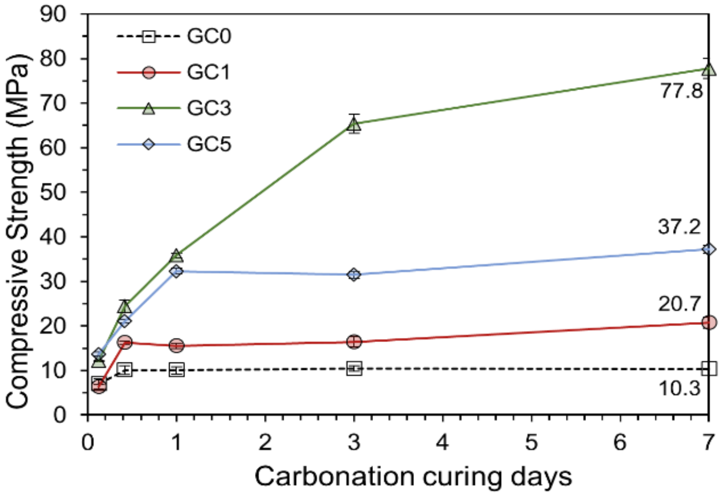


Figure 1. Compressive strength of carbonated steel slag pastes.

The compressive strengths were tested after the 40°C hydration curing and continued through the carbonation curing period. The use of 3% glycine showed a consistent increase in compressive strength up to the 7th day, while all other samples exhibited minimal strength changes after 1 day.

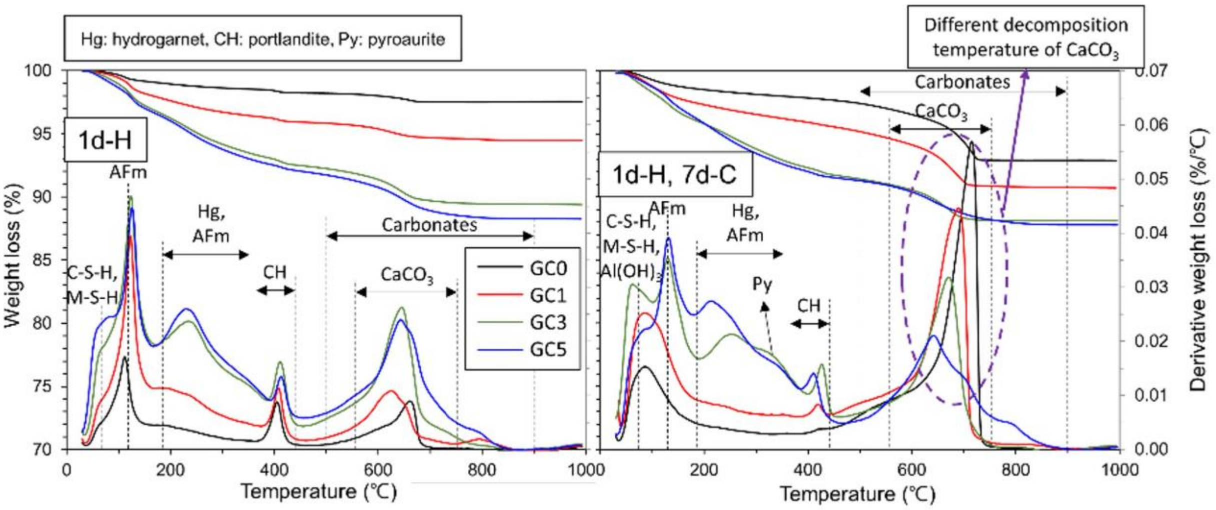


Figure 2 TG results after hydration for 1 day and subsequent carbonation for 7 day

From the XRD analysis, below observation was confirmed. While C_2S was consumed both during the hydration and carbonation, C_2F was consumed only during the hydration. As the glycine dosage increased, the amount of hydrated C_2S and C_2F significantly increased. In particular, most of the C_2F was consumed within one day with the use of glycine. Figure 2 shows the TG results of steel slag pastes during the curing process. As a result of the C_2F consumption, substantial amounts of C-S-H, AFm, hydrogarnet, portlandite, and $CaCO_3$ were precipitated in the hydration (Figure 2 ‘1d-H’). On the other hand, during the carbonation process, the carbonation degree of C_2S and AFm decreased with increasing glycine dosage. This resulted in reduced amounts of generated $CaCO_3$ and decreased decomposition temperatures, as shown in the ‘1d-H, 7d-C’ results in the Figure 2.

As an example of appropriate technology to reduce CO2 foot print in construction materials, this study introduces as mentioned carbonation curing. Also, more details regarding the technology to transform non-SCM to have reactivity of conventional SCM will be givein in the talk. Grinding agent is typically utilized for improving grinding efficiency such as energy saving. Customized grinding agent can have a huge potential to transform non-SCM to SCM. Considering the typical dosage of grinding agent of less than 0.1%, it has more space to be added and designed further for each non-SCM such as bottom ash or steel slag. In this sense, understanding the chemistry of hydration behavior and characterization of raw materials is crucial to confirm its performance and reproducibility.

Keywords

Bottom ash; steel slag; cementless binder; carbonation; grinding agent

Acknowledgment

This work was supported by the Technology Innovation Program (RS-2023-00266009, Development of admixture and blended cement using unused inorganic resource) funded By the Ministry of Trade, Industry & Energy (MOTIE, Korea).

ENHANCING EFFECT OF β-CYCLODEXTRIN ON CARBONATION PROPERTIES OF STEEL SLAG

Jinglong Huang[1], Yilian Li[1], Yunhua Zhang[1]

[1]School of Materials and Chemical Engineering, Hubei University of Technology, Wuhan, China
Email: yhzhang@whut.edu.cn

Abstract

In order to improve the carbonation properties of steel slag, the carbonation reaction process of steel slag and the formation of carbonation products were regulated by the interaction between multiple hydroxyl groups within β-cyclodextrin (β-CD) molecule and calcium ions leached from steel slag. Through a meticulous experimental design, varying contents of β-CD, from 0.0% to 5.0%, were mixed with steel slag to investigate their impact on carbonation kinetics, mechanical strength, and microstructural evolution. The results indicate that β-CD significantly accelerates the carbonation process of steel slag, with a particular boost to the carbonation of β-dicalcium silicate (β-C_2S), a key mineral contributing to the carbonation activity. At an optimal β-CD concentration of 2.5%, the compressive strength of the treated slag reached a peak of 138.5 MPa, marking a 13.4% improvement over the control sample. Similarly, the CO_2 uptake was optimized at 18.3%, increasing by 16.5%. Mechanism analysis reveals that β-CD not only facilitates carbonation but also refines the resulting carbonation products, leading to a denser microstructure and enhanced mechanical properties. However, at a β-CD content of 5%, the carbonation process was hindered by excessive viscosity. These pivotal findings propose an innovative strategy to augment the efficiency of carbonation and the inherent material properties of steel slag, thereby fostering a more sustainable approach to the utilization of industrial byproducts.

Keywords
Steel slag; β-cyclodextrin; accelerated carbonation; enhancing mechanism

BIOMOLECULAR REGULATED CARBONATION TO PROCESS CALCIUM-RICH ALKALINE INDUSTRIAL WASTES INTO SUPPLEMENTARY CEMENTITIOUS MATERIALS

Sen Zhang[1], Xi Xiao[1], Cong Shen[1], Jialai Wang[2], and Hongyu Zhou[1]

[1]Department of Civil and Environmental Engineering, University of Tennessee Knoxville
[2]Department of Civil, Environmental, and Construction Engineering, University of Alabama Tuscaloosa
Email: hzhou8@utk.edu

Abstract

Concrete as the most used manmade material on planet earth, provides tremendous opportunity to sequestrate CO_2. Current production process of OPC is extremely energy and carbon intensive, accounting for ~8% of total CO_2 emission worldwide. CO_2 mineralization, which sequestrates CO2 via reaction with oxides of alkaline or alkaline-earth metals to form carbonates, is an effective method curb atmospheric greenhouse gas emissions for climate change mitigation. This research presents a low-cost and eco-friendly biomolecule-regulated carbonation, namely BioCarb, pathway that converts calcium-rich, alkaline construction and industrial wastes – i.e., cement kiln dust (CKD), and high calcium fly ash into carbon-negative and carbonate-rich supplementary cementitious materials (SCMs) and permanently stores CO_2 in the produced SCMs. Unlike other CO_2 mineralization process in aqueous environment, the new method uses a low-cost biomolecule as a small doze additive to facilitate the carbonation and regulate the formation of $CaCO_3$ to produce metastable calcium carbonate. This produced carbonate can react with ordinary Portland cement (OPC) or its hydration products, triggering a new pozzolanic reaction. Strength improvement can be achieved by the concrete made with this novel SCM over those made with existing carbonation methods. Additionally, the proposed technology brings vast environmental benefits by reducing/ eliminating multiple waste material streams from several industry sectors.

INNOVATIVE USE OF AMINES TO ENHANCE CO_2 MINERALISATION IN STEEL SLAG FOR ECO-FRIENDLY CONSTRUCTION MATERIALS

Yogarajah Elakneswaran[1], Kanta Takebe[1], Ryoma Kitagaki[1], Ryosuke Saito[2], Keiichi Yano[2], Yuya Yoda[2], Masato Tsujino[2], Akira Nishida[2], Hisanori Senboku[1], Koji Fushimi[1], Masatoshi Sakairi[1]

[1] Faculty of Engineering, Hokkaido University
[2] Shimizu Corporation, Shimizu Institute of Technology
Email: elakneswaran@eng.hokudai.ac.jp

Abstract

Global warming has become an urgent issue due to the increasingly severe CO_2 emissions into the atmosphere. The steel industry significantly contributes to these emissions and generates steel slag as an alkaline industrial waste. Utilising steel slag to absorb CO_2 from flue gas can be an effective strategy for both reducing CO_2 emissions and recycling industrial waste. Amines are commonly used in post-combustion CO_2 capture due to their ability to form carbamates or bicarbonates, which are crucial for CO_2 capture. Amines that form bicarbonates enhance calcite formation, while those forming carbamates do not contribute to calcite formation during CO_2 capture. While extensive research has focused on the carbonation of cement-based materials and various methods to accelerate mineralisation [1-3], the incorporation of amines into cement-based materials for enhancing CO_2 capture introduces a novel approach. Despite our prior investigation [4] focusing solely on analysing the CO_2-capturing ability of a tertiary amine (MDEA) in cement-based materials, identifying a superior amine for enhancing CO_2 mineralisation and application to alkaline materials is necessary. This study aims to enhance CO_2 mineralisation in steel slag using amines, with the goal of developing sustainable construction materials.

The investigation examined three amines: N-methyldiethanolamine (MDEA) as a tertiary amine, 2-(methylamino)ethanol (MAE), and 2-amino-2-methyl-1-propanol (AMP), in combination with blast furnace slag fine powder (BFS). Ordinary Portland cement (OPC) was also added as an additive. The slag leaching process was conducted in a sample solution containing the amine, water, and slag, with and without cement. CO_2 was introduced for carbonation at an air concentration of 5 vol% and a flow rate of 2 L/min for 24 hours. After carbonation, the solid and liquid phases were separated using suction filters, and a comprehensive analysis was performed to identify and quantify the carbonation products. Additionally, slag cement paste, both with and without amines, was prepared and carbonated at air and 5 vol% for 28 days, after which the compressive strength was measured.

Calcium leaching leads to slag carbonation, but the addition of amines alone does not enhance this process (**Figure 1**). However, the overall carbonation reaction is significantly accelerated by the addition of cement. Higher $CaCO_3$ conversion rates were also observed with the inclusion of amines. Among the amines tested, AMP yielded the highest $CaCO_3$ content at 11.07%, indicating that the carbonation reaction was accelerated by approximately 3.5 times compared to the control sample and 1.5 times compared to the control sample with 5% cement. This suggests that the formation of calcium silicate hydrate (C-S-H), due to the addition of cement and its subsequent carbonation, enhances calcite formation.

Figure 2(a) shows the compressive strength of each sample carbonated at 0.04% and 5% CO_2 concentrations, while **Figure 2**(b) displays the carbonation conversion. The addition of MDEA was found to increase compressive strength. As the CO_2 concentration increased from 0.04% to 5%, both the control and MDEA samples exhibited increased strength and higher $CaCO_3$ formation. Conversely, a slight decrease in strength was observed in samples with MAE and AMP, which did not correlate with $CaCO_3$ formation. The relationship between carbonation and strength is complex with amines, involving factors such as porosity, subsequent density changes, and alterations in crystal structure due to carbonation.

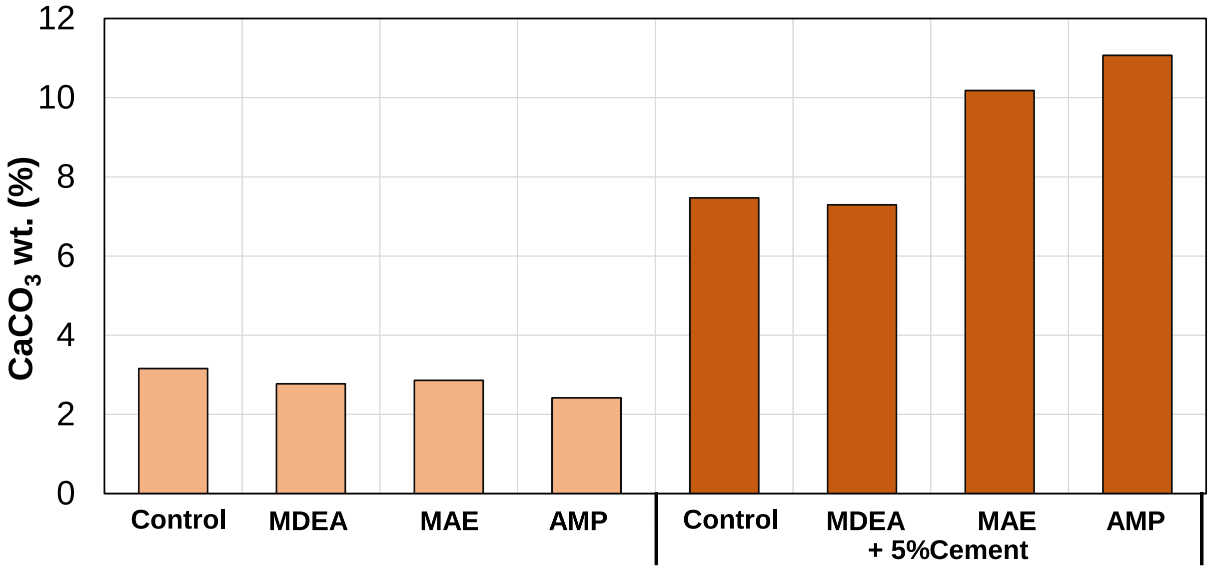


Figure 1: Produced $CaCO_3$ after wet carbonation process

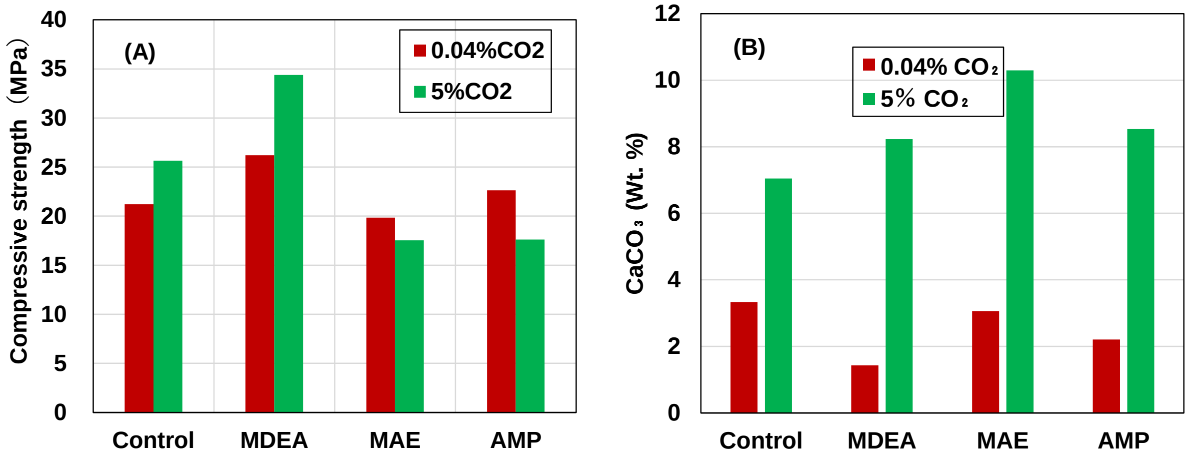


Figure 2: (A) Compressive strength; (B) formed $CaCO_3$ after slag cement paste exposed to air and 5 % CO_2 environment

Keywords
Amines; CO_2 emission; cement; calcium silicate hydrate (C-S-H); calcite

References

[1] M. Boumaaza, P. Turcry, B. Huet, A. Aït-Mokhtar, Constr. Build. Mater. 253 (2020).
[2] L. Liu, Y. Liu, X. Tian, X. Chen, Constr. Build. Mater. 346 (2022) 128364.
[3] D. Wang, J. Xiao, Z. Duan, Constr. Build. Mater. 314 (2022) 125646.
[4] K. Kopitha, et al., Chem. Eng. J. 475 (2023)

INTERNAL-EXTERNAL SYNERGISTIC CO_2 SEQUESTRATION OF CEMENT-BASED MATERIALS USING AMINO ACID SALTS

Chen, T.[1], **Guo, M.**[1], **and Gao, X.**[1]

[1]School of Civil Engineering, Harbin Institute of Technology
Email: chentf@hit.edu.cn

Abstract

This study presents a new CCUS method, which uses potassium glycine (KG) solution to capture CO_2 derived from industrial flue gases and sequesters the CO_2 in cement-based materials by mineralization-desorption, thus achieving the cyclic regeneration of KG absorbent and the activation of cement-based materials. The results confirm the favorable absorption capacity and desorption efficiency of this method, with the CO2 cyclic capacity of KG absorbent up to 0.47 mol/mol. The addition of CO_2-rich KG solution increases 7-d and 28-d compressive strength by 29.8% and 16.2%, as well as achieving an overall CO_2 uptake of 4~11% in cement paste. However, at higher KG concentrations, the rapidly generated amorphous calcium carbonate covers the surface of cement particles, which shortens the setting time and inhibits early-age hydration. The presence of carbamate in CO_2-rich KG solution improves the stability of metastable $CaCO_3$ polymorphs and prolongs the period of dissolution-recrystallization. Calcite generated by the slowed recrystallization within 14 days having a filling and binding ability in the cement matrix, significantly reducing porosity and providing a secondary strengthening effect.

Keywords
CO_2 sequestration; cement-based materials; potassium glycine; metastable $CaCO_3$

IMPROVING CARBON DIOXIDE SEQUESTRATION IN CONCRETE WASTE THROUGH POLYMER MODIFICATION

Kravchenko, E.[1], **Besklubova S.**[2] **and Privizentseva, D.**[1]

[1]Southern Federal University, Russia
[2]University of Hong Kong
Email: lanabess@hku.hk

Abstract

The expansion of the global population has driven a surge in construction activities, which, while boosting the global economy, also contributes significantly to environmental challenges such as greenhouse gas (GHG) emissions and resource depletion. The production of cement is particularly impactful, generating about 0.9 tonnes of CO_2 per tonne of cement and contributing 5-7% of global GHG emissions (Chaudhury et al., 2023). Additionally, construction and demolition activities produce large amounts of waste, including waste concrete powder (WCP), which is often relegated to landfills, leading to further environmental degradation (Besklubova et al., 2023). Research indicates that WCP can sequester CO_2, but traditional methods often require high temperatures and long carbonation times, increasing the carbon footprint (Kravchenko et al., 2024). This study explores using polyvinyl alcohol (PVA), a eco-friendly polymer, to enhance WCP's CO_2 sequestration capacity.

In this study WCP was sourced from the South East New Territories Landfill in Hong Kong. Commercially available PVA powder has a maximum viscosity of 45 mPa•s, 85% purity and 6.0% volatile substances. The molecular weight is 105×10^3 (Mw) with a 99% degree of hydrolysis. PVA powder was dispersed in water at 20°C, then heated to 70°C and stirred to dissolve fully. After cooling to room temperature, the PVA solution was mixed with dry WCP at a water-to-solid ratio of 0.23. The mixture was processed in a construction mixer until homogeneous, then layered into a metal mold and compressed at 12.5 MPa using a press machine equipped with a digital tensiometer for load control. PVA-treated WCP samples, sized 200 mm×100 mm×60 mm, were suitable for paving block manufacturing (ETWB(W) 810/83/02, 2004). These compacts were then placed in a carbonation chamber with CO_2 gas at 0.15 MPa for 2, 4, and 6 hours. A detailed diagram of the carbonation chamber setup and the testing procedure is provided in the work of Kravchenko et al (2023).

Figure 1 shows the thermogravimetric (TGA) and differential thermogravimetry (DTG) curves. The initial mass loss (30–130°C) in TGA is due to the vaporization of free water and decomposition of C–S–H and hydrated aluminates. The second mass loss at 435°C is from portlandite decomposition, with a DTG peak at 400–450°C due to $Ca(OH)_2$ dehydration. The most mass loss occurred in samples carbonized for 6 hours. Raw PVA degradation below 300°C primarily produces water, with complete OH group cleavage at 360°C. PVA-treated WCP showed greater mass loss due to $CaCO_3$ decomposition. At 800°C, non-carbonized PVA-treated WCP had a 14.8% mass loss, increasing to 17.6%, 19.7%, and 20.1% after 2, 4, and 6 hours of carbonation, respectively, indicating more low- and high-crystallinity $CaCO_3$ with longer carbonation. Carbon absorption calculations from mass gain and TGA mass loss curves indicated that the optimal carbonation time was 4 hours. During this period, WCP compacts absorbed up to 53.7 kg of CO_2 per m^3 of finished compacts.

Compression tests were conducted on 127 mm×40 mm specimens to assess the impact of carbonation curing and PVA treatment on strength. The results reveal that increased carbonization time enhances compressive strength due to a higher concentration of calcium carbonate. PVA consistently strengthens all samples, boosting strength by 13%, 15%, and 28% at 2, 4, and 6 hours of carbonation, respectively, compared to untreated WCP. PVA improves mechanical properties by filling pores and enhancing particle bonding.

Incorporating PVA not only boosts CO_2 absorption during carbonation but also enhances the strength of recycled WCP concrete by forming calcium carbonate and creating a polymer film around WCP particles. These findings suggest a sustainable method for utilizing construction waste and reducing the carbon footprint of concrete production, contributing to more eco-friendly building practices.

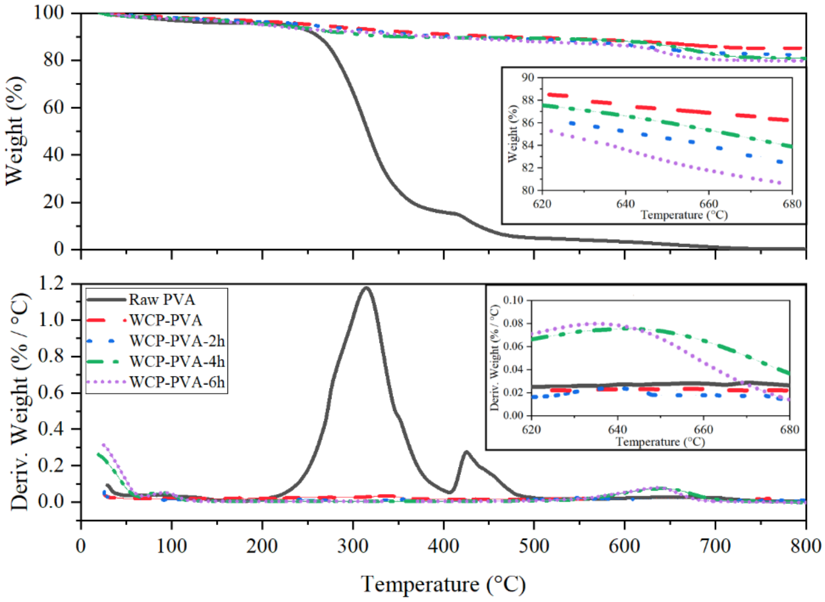


Figure 1. TGA and DTG curves

Keywords
Carbon sequestration; polyvinyl alcohol; concrete recycling; carbon uptake

Acknowledgment
The authors are grateful for the financial support provided by the 'Priority 2030' program of the Southern Federal University, Russia.

References

Besklubova, S., Kravchenko, E., Tan, B. Q., & Zhong, R. Y. 2023. A feasibility analysis of waste concrete powder recycling market establishment: Hong Kong case. *Environmental Impact Assessment Review*, *103*, 107225.

Chaudhury, R., Sharma, U., Thapliyal, P., & Singh, L. 2023. Low-CO2 emission strategies to achieve net zero target in cement sector. *Journal of Cleaner Production*, 137466.

Kravchenko, E., Qin, C., Lin, Z., & Ng, C. W. W. 2023. Effect of polyvinyl alcohol on the CO2 uptake of carbonated steel slag. *Construction and Building Materials*, *375*, 130761.

Kravchenko, E., Sauerwein, M., Besklubova, S., & Ng, C. W. W. 2024. A comparative life cycle assessment of recycling waste concrete powder into CO2-Capture products. *Journal of environmental management*, *352*, 119947.

THE ROLE OF ADDITIVE ON CARBONATION BEHAVIOR AND MECHANICAL PROPERTIES OF LOW CALCIUM SILICATE MATERIALS

Junil Pae[1], **Juhyuk Moon**[1,2]

[1] Department of Civil and Environmental Engineering, Seoul National University, Seoul 08826, Republic of Korea
[2]Institute of Construction and Environmental Engineering, Seoul 08826, Republic of Korea
Email: juhyukmoon@snu.ac.kr

Abstract

This study examined the effects of adding admixture to carbonation-cured pseudowollastonite pastes on their microstructure, compressive strength, and mineral carbonation using TGA, XRD, FT-IR and micro-CT. Pseudowollastonite was successfully produced on a lab scale and utilized to make pastes without the need for energy-intensive procedures. The outcomes showed a substantial difference in the effects of additive at concentrations of 0.5% and 1%. As shown in Figure 1, the addition of 0.5% additives was the single factor that increased compressive strength to 25.8 MPa, representing a 24.0% increase over the control. The phase boundary guided the first stage of the carbonation reaction of the pastes with or without 0.5% additives, whereas product layer diffusion drove the second stage (Figure 2).

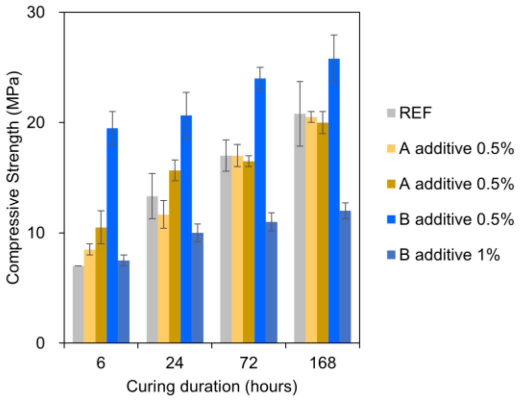


Figure 1. The compressive strength development of carbonated pseudowollastonite pastes.

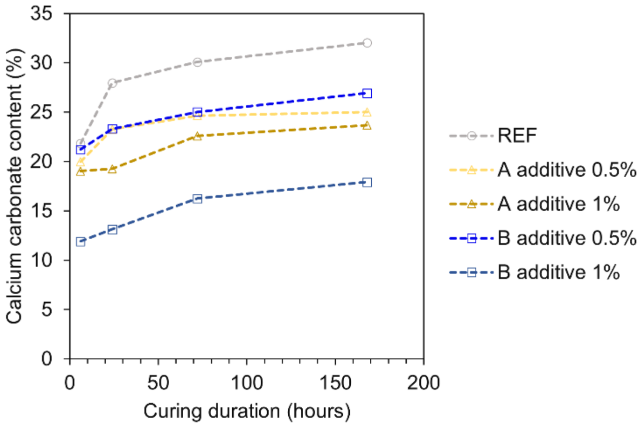


Figure 2. Amount of calcium carbonate precipitated during the carbonation curing.

The calcium carbonate phases found in the pastes were determined to be amorphous calcium carbonate (ACC), vaterite, and calcite. In the phase boundary stage of carbonation reaction, the addition of 0.5% additives facilitated the formation of ACC. The ACC nearly fully recrystallized into calcite in the subsequent product layer diffusion stage. The inclusion of additives also increased internal carbonation and prevented the development of macrocracks, which improved the pastes' compressive strength. It was established that the volume percentage of cracks controls both the compressive strength and the extent of carbonation reaction of the pastes containing additives.

Intriguing conclusion of this study is that aspartic additives are a highly effective additive that improves the carbonated pseudowollastonite paste's mechanical performance. The improved compressive strength of the pastes containing additives was speculated to be due to the creation of an organic/inorganic hybrid material, greater internal carbonation, inhibition of macrocrack formation, and the recrystallization of ACC into calcite. Therefore, integrating additives into pseudowollastonite may provide a promising method for creating a CO_2 sequestration binder.

Keywords

Pseudowollastonite; calcium silicate; additives; CO_2 curing; calcium carbonate

Acknowledgements

This work was supported by the Industrial Strategic technology development program-Development of manufacturing technology of hardened cement with carbonation curing (RS-2022-00155662, Development of manufacturing and application technology of 1,000 ton/year class hardened cement with carbonation curing) funded By the Ministry of Trade, industry & Energy (MOTIE, Korea).

ENHANCED HYDRATION OF GROUND STEEL SLAG WITH CHEMICAL ACTIVATOR

Ahyeon Lim[1], **Juhyuk Moon**[1,2]

[1]Department of Civil and Environmental Engineering, Seoul National University, Seoul 08826, Republic of Korea
[2]Institute of Construction and Environmental Engineering, Seoul National University, Seoul 08826, Republic of Korea
Email: ahyunlim@snu.ac.kr

Abstract

Steel slag is a by-product generated in the iron industry like ground granulated blast-furnace slag, but it has not been generally used as a supplementary cementitious material due to its low reactivity [1]. Steel slag is typically composed of C2S, C4AF, RO phase, and other inert phases, so the reactivity of steel slag is much lower than other supplementary cementitious materials [2]. In this study, chemical admixture was adopted to improve the mechanical properties of steel slag.

The steel slag used in this study was provided by POSCO Co. Ltd., Korea. It was ground for 3 hours and steel slag powder with a particle size of 600 µm or less was used in this experiment. 0, 0.5, and 1.0% of chemical admixture was added to steel slag and mixed with 0.2 of w/b. The admixture was fully dissolved in water by a magnetic stirrer for at least 2 hours before mixing. TG and XRD analyses were conducted to investigate the hydration properties of steel slag after 3 and 28 days of curing.

Error! Reference source not found. shows the TG results of hydrated steel slag at 3 and 28 days. In the presence of chemical admixture, chemically bound water (CBW) which is the weight loss between 30 ℃ and 500 ℃ was increased and this indicated that the degree of hydration was enhanced from 3 days of curing. Meanwhile, without activation, steel slag was almost unreacted until the 28 days of curing. The addition of chemical admixture mainly contributed to the formation of C-S-H, AFm phases, and calcite. C-S-H and AFm phases were intensively precipitated until 3 days of curing, and calcite was increased for the overall curing period. In particular, 1% use of activator was effective for the productions of C-S-H and AFm phases. Furthermore, portlandite was consumed by reacting with chemical admixture and produced the calcite. At 28 d, a significant increase in calcite was observed with the addition of the activator.

XRD patterns of hydrated steel slag at 3 and 28 days are presented in **Error! Reference source not found.**. In the presence of the chemical activator, the reaction of C2S was significantly enhanced. As shown in TG results, C-S-H was increased with the addition of the activator, which was generated by the reaction of C2S. The main hydration products of steel slag identified in the XRD pattern were AFm phases and calcite. At 3 d, without chemical admixture (SS0), hydration products were hardly observed. However, a remarkable increase of AFm phases was observed with 1% of admixture which is well matched with TG results. Otherwise, there was no significant difference in the formation of calcite according to the addition of the chemical activator at 3 d. At 28 d, a small amount of AFm phases were produced in SS0 and

SS0.5, whereas, AFm phases were increased in SS2. In addition, more calcite was produced with the addition of 1% of the chemical activator.

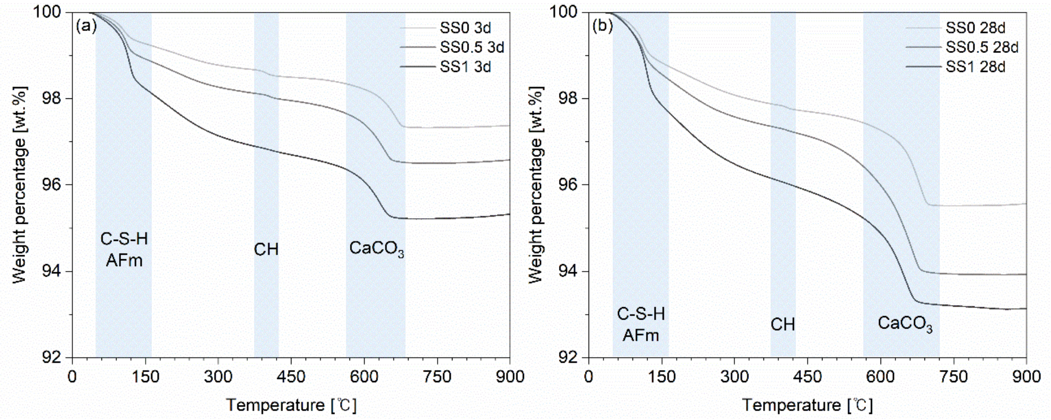


Figure 1. TG results of steel slag at (a) 3 d, and (b) 28 d.

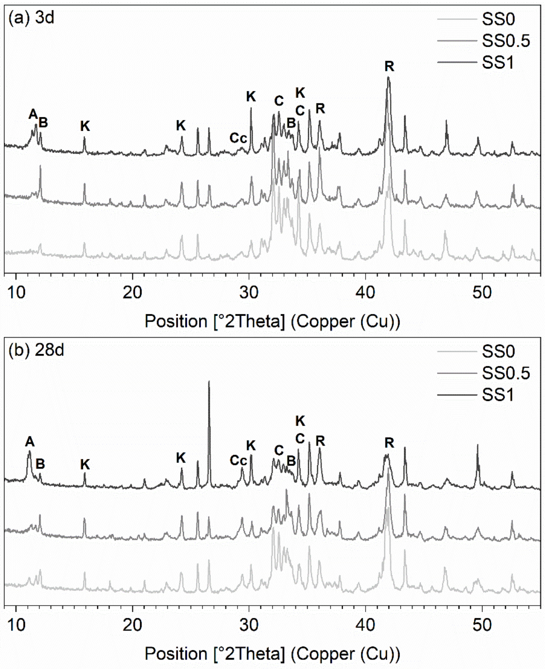


Figure 2. XRD patterns of steel slag at (a) 3 d, and (b) 28 d.

Keywords

Steel slag; alkali activator; hydration; compressive strength; carbonation

References

1. Yildirim, I. Z., & Prezzi, M. (2011). Chemical, mineralogical, and morphological properties of steel slag. Advances in civil engineering, 2011.
2. Yi, H., Xu, G., Cheng, H., Wang, J., Wan, Y., & Chen, H. (2012). An overview of utilization of steel slag. Procedia Environmental Sciences, 16, 791-801.

CONTROLLABLE $CaCO_3$ SYNTHESIS FROM SOLID WASTE BY AN “ALL-IN-ONE” AMINO ACID-IN STRATEGY

Xuan Zheng[1], Long Ji[1], Kangkang Li[2], Liang Li[3], Hai Yu[3] and Shuiping Yan[1]

[1] College of Engineering, Huazhong Agricultural University, Number 1, Shizishan Street, Wuhan 430070, China
[2] Department of Energy and Resources Engineering, College of Engineering, Peking University, Beijing100871, China
[3] CSIRO Energy Centre, Mayfield West, Newcastle, NSW 2304, Australia
Email: long.ji@mail.hzau.edu.cn; yanshp@mail.hzau.edu.cn

Abstract

Solid wastes carbonation suffers from extensive consumption of acidic/alkaline reagents due to the limited kinetics, which necessitates the development of an atom/energy-efficient process. To address such an issue, an innovative amino acid-mediated Ca leaching-CO_2 mineralization cycle process was proposed which enabled simultaneous CO_2 capture and recovery of valuable products (i.e. $CaCO_3$ and supplementary cementitious material (SCM)). In this research, amino acid, in this all-in-one strategy, acts as a multifunctional reagent that can simultaneously achieve promising Ca leaching from alkaline waste, appreciable CO_2 absorption, and tunning of high-valued $CaCO_3$. This study explored the interactions between amino acid-species and Ca-bearing minerals/Ca^{2+}, $CaCO_3$ pre-nucleation, nucleation, and growth by investigating the evolution of active compounds from the perspectives of thermodynamic equilibrium analysis and experimental study including evolution of Ca concentration, solution turbidity and morphology in complex coal fly ash (CFA). Thermodynamic equilibrium analysis showed that the active Ca-bearing minerals of CFA varied in the systems of ammonium salt, acid, and amino acid, e.g., the active phases in glycine (Gly) were portlandite ($Ca(OH)_2$), lime (CaO), and gehlenite ($CaO \cdot Al_2O_3 \cdot SiO_2$). High Ca^{2+} concentrations of 0.26 M, 0.14M, and 0.1 M were achieved after 3 minutes of leaching assisted by amino acids of Gly, L-Alanine (Ala), and L-Arginine (Arg), respectively, which demonstrated the kinetics and thermodynamical feasibility. Such phenomenon was contributed to the fact that Gly and Ala had minimal pH buffering capacity in the early leaching stage. The deprotonated forms of Gly and Ala with a higher affinity with Ca^{2+} increased from 0.03% and 0.04% to 33.9% and 12.88%, respectively, thereby promoting Ca^{2+} leaching. Moreover, the leaching residue with reduced Ca content was used as a SCM and the compressive strength reached 33.4 MPa after 28 days of curing. These amino acids ultimately led to an increased $CaCO_3$ production as follows: Gly (114.8 g/kg) > Ala (78.2 g/kg) > Arg (54.5 g/kg) from CFA. Gly leachate remained transparent (1.23 NTU) in the first 3 minutes of CO_2 bubbling even though the solution was supersaturated and then got turbid suddenly (6508.75 NTU in 10 minutes) as continued CO_2 bubbling, indicating the inhibition of Gly on $CaCO_3$ nucleation. Observations using Cryo-SEM verified that the presence of abundant $CaCO_3$ nanoparticles in the early stages of mineralization, which later assemble into vaterite crystal particles with a sub-structure and coalesce into larger particles in Gly and Ala. Besides, Arg inhibited calcite growth along the C-axis, leading to the formation of thin, blocky-like particles. Furthermore, 0.010~0.027 mmol/g amino acids were occluded into the $CaCO_3$, stabilizing the metastable vaterite and imparting thermal stability, which appeared to be associated with this multistep pathway involving a transient prenucleation cluster, nanoparticle precursor formation, and particle-particle coalescence. To sum up, the present study took a step

forward for a better understanding of the production of $CaCO_3$ from alkaline waste by carbonation of solid wastes to minimize energy and materials consumption.

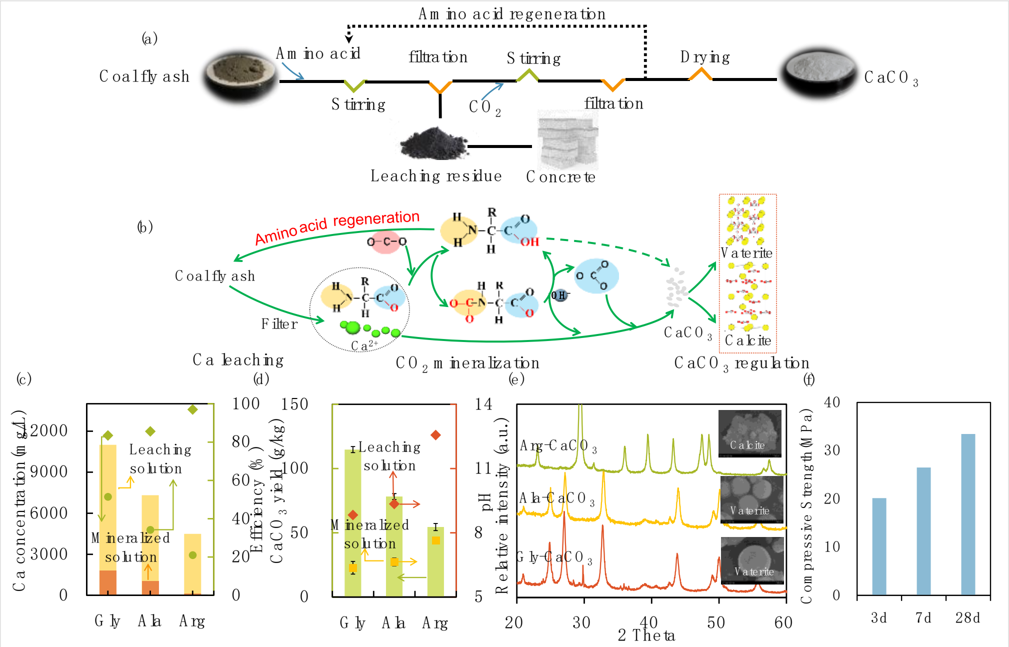


Figure 1. Ca^{2+} leaching-CO_2 mineralization process using coal fly ash. (a) The flow diagram. (b) The details of the proposed pathway. (c) $[Ca^{2+}]$ and corresponding efficiency before and after CO_2 mineralization. (d) Production of various $CaCO_3$ induced by amino acids and pH of leaching and solution. (e) XRD and FE-SEM images of $CaCO_3$ induced by amino acids. And (f) Strength development of cement paste.

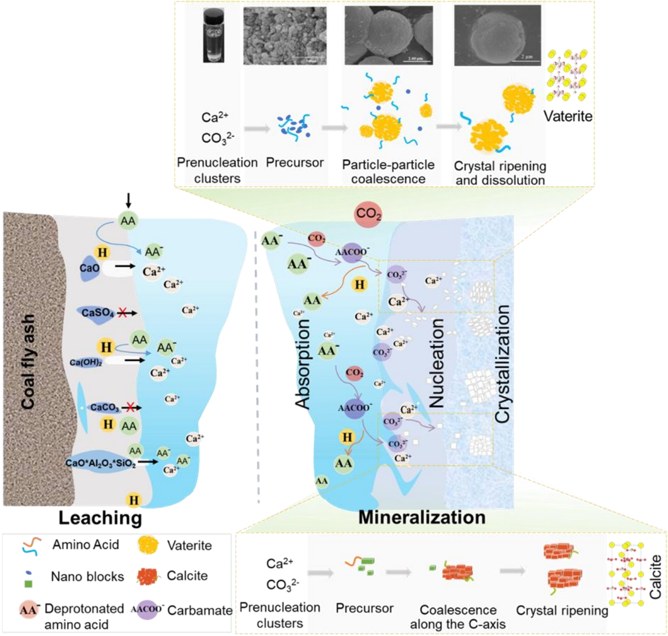


Figure 2. Schematic diagram of the possible formation mechanism of Ca selective leaching, spherical vaterite and step-like calcite $CaCO_3$ from coal fly ash.

Keywords

CO_2 mineralization; amino acid; vaterite; calcite; supplementary cementitious material

IMPROVEMENT IN THE CARBON CAPTURE OF CEMENTITIOUS MATERIALS USING HYDROGEL

Tao Wang[1], **Liang Liu**[2], **Tung-Chai Ling**[1]

[1]College of Civil Engineering, Hunan University, 410082, Changsha, China
[2]Institute of Bast Fiber Crops, Chinese Academy of Agricultural Sciences, 410205, Changsha, China
Email: tcling@hnu.edu.cn

Abstract

The carbon capture is a continuing concern due to the continuous increase of carbon dioxide concentration in the atmosphere and the accompanying reality of global warming. Cementitious materials applied in construction and civil engineering, possess excellent carbon capture capabilities due to a high alkalinity. However, the dense and disconnected microstructure is one of the main limitations for the carbon capture of cementitious materials. Thus, the design and regulation of pore structure could significantly enhance the carbon capture capability of cementitious materials.

The hydrogels composed of hydrophilic polymers exhibit high absorption stability, porous structure, and adjustable mechanical properties. They have been widely applied in the fields of biomedicine, vehicle engineering, and construction engineering. Researches in the last two decades have identified that the addition of hydrogel can improve the mechanical properties and durability of cementitious materials.

Based on the hydrogels' characteristic of strong liquid absorption, high toughness, and porous structure, this study aims to reveal the impact of hydrogels on the carbon capture efficiency, strength, and microstructure of cement paste. The results showed that the hydrogel obtained by physical crosslinking did not present a significant effect on the hydration of Portland cement. Compared to pure cement paste, the hydrogel incorporated cement paste demonstrated a more significant strength gain after carbonation. Moreover, the addition of hydrogel into cement can contribute to the increase of micropore volume, thereby leading to the enhancement of carbon capture properties of hydrogel incorporated cement paste.

Keywords
Hydrogel; cementitious materials; carbon capture; microstructure

MECHANISM OF ORGANIC ADDITIVES-INDUCED CARBONATION ACTIVATORS ON AFFECTING CEMENT MORTARS

Shuangshuang Liu [1,2], **Guangqi Xiong**[1,2], **Shipeng Zhang**[1,2], **Chi Sun Poon**[1,2]

[1] Department of Civil and Environmental Engineering, The Hong Kong Polytechnic
University, Hung Hom, Kowloon, Hong Kong
[2] Research Centre for Resources Engineering Towards Carbon Neutrality, The Hong Kong
Polytechnic University, Kowloon, Hong Kong
Email: shipeng.zhang@polyu.edu.hk

Abstract

In this study, we investigated the impact of introducing an in-situ activator, produced by carbonating cement particles in an aqueous solution, on the properties of cement mortars through secondary mixing. Two organic additives were employed to enhance the leaching of calcium ions during carbonation, thereby improving carbonation efficiency. A suite of characterization techniques revealed that the presence of organic additives could refine the carbonated particles and influence the morphology. The carbonated activators generated by this process were rich in silica gel and various polymorphic forms of calcium carbonate. These components, serving as fillers and nucleation for cement hydration, significantly accelerated the hydration process of cement mortar and promoted the formation of carboaluminate in the secondary mixing process. This approach effectively decreased the porosity of the cement mortar, refined the pore structure, and enhanced the mechanical strength.

Keywords

In-situ pre-carbonation; organic additives; $CaCO_3$ polymorphs; reactive seeding; accelerated hydration

Session C4

Integration of Waste and CO_2 for Construction

MARINE STRUCTURES WITH RECYCLED CONCRETE AND NON-METALLIC REINFORCEMENTS

Tamayo, P.[1], Gomez, G.[1], Trevilla, Z.[1], Qazi, S.[1], Cimentada, A.[1], Thomas, C.[1]

[1]LADICIM (Laboratory of Materials Science and Engineering), University of Cantabria
E.T.S. de Ingenieros de Caminos, Canales y Puertos, Av./Los Castros 44, 39005 Santander, Spain
Email: carlos.thomas@unican.es

Abstract

This work addresses the critical issue of corrosion in marine structures and introduces an innovative solution through the use of recycled concrete reinforced with non-metallic elements.

The aggressive marine environment accelerates the corrosion of traditional steel-reinforced concrete, compromising the durability and safety of coastal and underwater infrastructures. To tackle this challenge, the research utilizes recycled aggregates sourced from construction and demolition wastes, incorporating them into a specially designed concrete mix. By replacing conventional steel reinforcements with non-metallic alternatives, the project aims to significantly enhance the longevity of the structures while maintaining mechanical integrity.

The work focuses on the development and implementation of these non-metallic reinforced recycled concrete in the construction of artificial reefs. It explores the mechanical properties, durability, demonstrating how this approach mitigates corrosion-related issues. Additionally, the research discusses the ecological benefits of using this sustainable material in creating habitats that foster marine biodiversity.

By providing insights into the application of recycled concrete and non-metallic reinforcements, the presentation contributes to the advancement of sustainable construction practices in marine engineering. It offers a compelling perspective on how innovative materials and techniques can overcome traditional challenges, promoting the long-term viability of marine infrastructures.

Keywords

Corrosion; recycled concrete; non-metallic reinforcements; marine structures; sustainability

MECHANISMS AND ADVANCEMENTS IN MICROWAVE-ENHANCED CO_2 MINERALIZATION OF LIGHTWEIGHT POROUS CONCRETE

Lei Yang[1], Zhuo Liu[1], Shuqiong Luo[1], Songhui Liu[1], Xuemao Guan[1], Tangwei Mi[2]

[1] Henan Key Laboratory of Materials on Deep-Earth Engineering, School of Materials Science and Engineering, Henan Polytechnic University, Jiaozuo 454003, P. R. China
[2] School of Civil and Environmental Engineering, Nanyang Technological University, 50 Nanyang Avenue, 639798, Singapore
Email: luoshuqiong@hpu.edu.cn

Abstract

Lightweight porous concrete (LPC) typically exhibits low early strength, which can be addressed by carbon dioxide (CO_2) mineralization curing to enhance early strength while achieving permanent CO_2 sequestration. The efficacy of this process relies on maintaining optimal moisture levels. This study investigates the influence of moisture content on the performance of solid waste-based lightweight porous concrete blocks (RSFAC) subjected to pretreatment with microwave heating. Moisture levels were adjusted to 10%, 20%, 30%, and 40% (initial water-to-cement ratio of 45.4%) before undergoing mineralization curing within an environment set at a temperature of 20 °C, relative humidity of 75%, and a CO_2 concentration of 40% for a duration of 0.5 hours. The impact of moisture content on RSFAC's compressive strength, carbon sequestration rate, mineralization products, and microstructure was evaluated. Results indicate that RSFAC treated with microwave heating outperforms untreated samples (Ref-0 and Ref-1) in terms of compressive strength and carbon sequestration. A peak in performance was observed at a moisture content level of 30%, with the highest compressive strength recorded at the age of 1, 3, and 7 days. The compressive strength reached its peak value at the age of 7 day with a measurement of 2.32 MPa, while the carbon sequestration rate achieved a value as high as 8.2%. This enhancement is attributed to the microwave effect facilitating rapid water migration within pore spaces while simultaneously providing suitable diffusion pathways for CO_2 during mineralization. Consequently, CO_2 mineralization is intensified, which results in a denser packing of mineralization products, effectively filling RSFAC pores and improving structural strength.

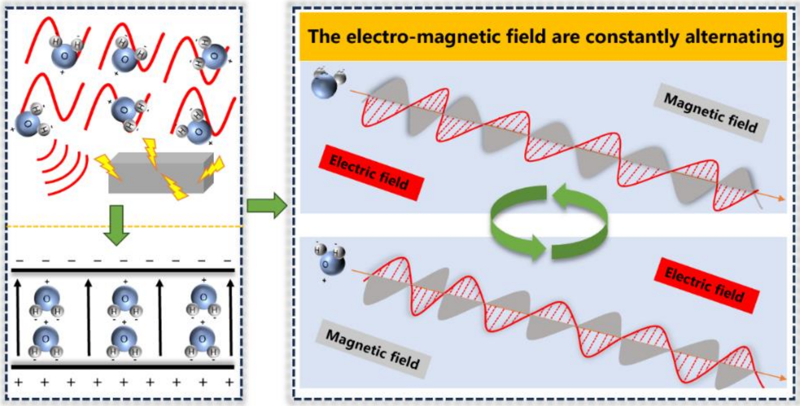


Fig. 1. Microwave interaction with water molecules: a mechanism of heat generation

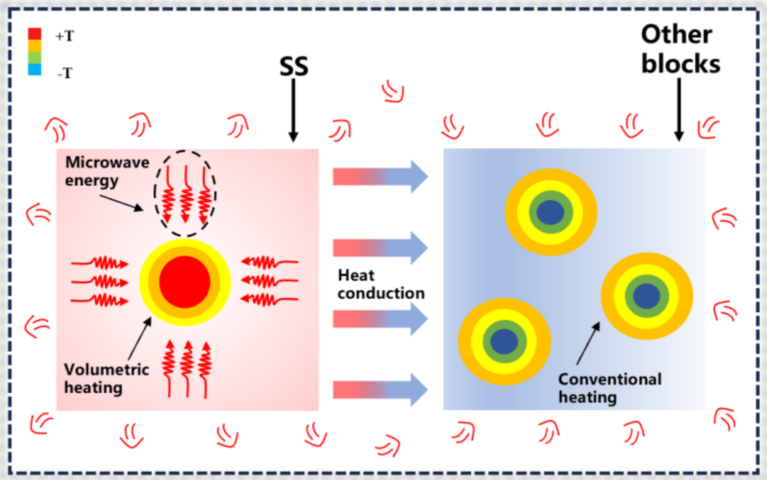


Fig. 2. Heat generation through microwave absorption by Fe_2O_3

Keywords

Solid waste; porous concrete; microwave pretreatment; CO_2 mineralization curing; crystal morphology

References

[1] LI Y, ZHOU Y, WANG R, et al. Experimental investigation on the properties of the interface between RCC layers subjected to early-age frost damage [J]. Cement and Concrete Composites, 2022, 134.

[2] PRANEETH S, GUO R, WANG T, et al. Accelerated carbonation of biochar reinforced cement-fly ash composites: Enhancing and sequestering CO_2 in building materials [J]. Construction and Building Materials, 2020, 244.

[3] HAQUE K E. Microwave energy for mineral treatment processes—a brief review [J]. International Journal of Mineral Processing, 1999, 57(1): 1-24.

[4] TANG T, CAI L, YOU K, et al. Effect of microwave pre-curing technology on carbide slag-fly ash autoclaved aerated concrete (CS-FA AAC): Porosity rough body formation, pore characteristics and hydration products [J]. Construction and Building Materials, 2020, 263.

[5] MEHDIZADEH H, JIA X, MO K H, et al. Effect of water-to-cement ratio induced hydration on the accelerated carbonation of cement pastes [J]. Environ Pollut, 2021, 280: 116914.

CO_2-DRIVEN 3D CONCRETE PRINTING TECHNOLOGIES

Shipeng Zhang[1,2], **Lucen Hao**[1], **Hengrui Liu**[1], **Kaiyin Zhao**[1], **Chi-sun Poon**[1,2]

[1]Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hong Kong, China
[2]Research Centre for Resources Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hong Kong, China
Email: shipeng.zhang@polyu.edu.hk

Abstract

In response to labor shortages, an aging workforce, construction waste generation, and low productivity, the construction industry is increasingly transitioning towards digitalization, with 3D concrete printing (3DCP) emerging as a promising solution. Unlike conventional methods, 3DCP utilizes digital controls to extrude concrete layer by layer, eliminating the need for formwork and significantly reducing construction time, costs, and waste. However, this process demands more precise control over the rheological properties of concrete, often necessitating mixes with excessive cement, which conflicts with sustainability goals due to the high carbon footprint of Ordinary Portland Cement (OPC). This talk introduces innovative strategies to reduce the embodied carbon in 3DCP by incorporating gaseous waste CO_2. The greenhouse gas CO_2 serves as a rheology modifier, facilitating a 'set-on-demand' approach that balances pumpability with buildability while ensuring permanent CO_2 sequestration within the printed concrete.

Keywords

3D concrete printing; rheology control; sustainable concrete; carbonation

VALORIZATION OF WASTED-DERIVED BIOCHAR IN ULTRA-HIGH-PERFORMANCE CONCRETE (UHPC): PRETREATMENT, CHARACTERIZATION, AND ENVIRONMENTAL BENEFITS

Jiang Du[1 2], Yuhuan Wang[1], Yi Bao[1], Dibyendu Sarkar[1], and Weina Meng[1]

[1]Department of Civil, Environmental and Ocean Engineering, Stevens Institute of Technology, Hoboken, NJ 07030, USA
[2]State Key Laboratory of Mountain Bridge and Tunnel Engineering, Chongqing Jiaotong University, Chongqing 400074, People’s Republic of China
Email: jiangdu@cqjtu.edu.cn

Abstract

UHPC is the class of advanced concrete with self-consolidating property, outstanding mechanical performance, and superior durability [1]. However, in view of global warming concerns, the world-wide applications of UHPC are restricted due to the extremely high CO_2 emission and energy consumption [2]. Biochar is a by-product of biomass during the pyrolysis progress [3], which is usually performed at a temperature ranging from 450 ℃ to 550 ℃ in the absence of oxygen, resulting in the highly carbonaceous property of biochar [4, 5]. It was reported that, the production of biochar is promising to sequester 870 kg CO_2-eq greenhouse emission for every ton of biomass feedstock [6]. In addition, it was estimated that the use of every 1 kg biochar in concrete can sequester about 2.5 kg CO_2 [7].

This study presents the feasibility of utilizing wasted-derived biochar in ultra-high-performance concrete (UHPC) to alleviate environmental effects, reduce shrinkage behaviors, and further enhance the mechanical properties, shown in **Fig. 1**. Two types of wasted-derived biochar are investigated, including the fresh biochar from pyrolysis process of vetiver grass roots and the used biochar after purifying contaminated water with heavy metals. The wasted-derived biochar is pre-saturated and then used to substitute the cement in UHPC.

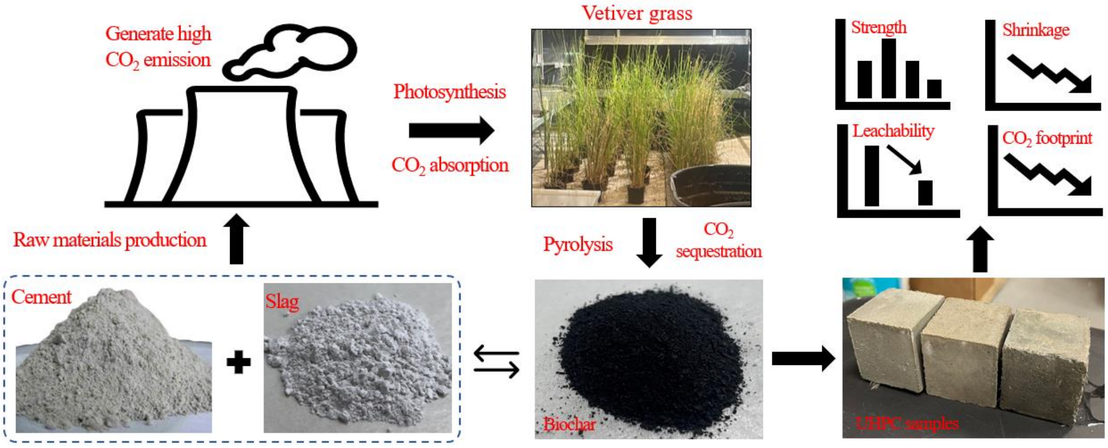


Fig. 1 Valorization of wasted biochar in producing eco-friendly UHPC

Experimental results showed that the 1% of pre-saturated biochar in UHPC enhanced the mechanical strengths (up to 20%) and reduced the autogenous shrinkage (up to 15%). The underlying mechanisms of the property enhancement are the improved hydration degree of

UHPC due to the internal curing effect. However, as non-negligible micro-defects, the utilization of over 1% biochar reduced mechanical performance instead. Moreover, as the biochar contents added to 1.0%, the carbon emissions of UHPC were reduced by 3.2% and the unit CO_2 emission per strength is reduced by 13%. This study successfully proposed a carbon sequestering admixture in UHPC production that also provides a new avenue for wasted-derived biochar recycling.

Keywords

Carbon sequestration; internal curing; wasted-derived biochar; sustainability; UHPC

Acknowledgement

This research is supported by the National Science Foundation [grant number CMMI-2046407] and New Jersey Department of Transportation: Task Order 388 – Bridge Resource Program (2021-2022), contract ID number: 21-50862. The authors also would like to thank Mr. Bill Kulish (the president of Steelike lnc) for the donation of steel fibers and Dr. Tsengming Chou of Stevens Institute of Technology for training AFM based nanoindentation test.

References

[1] Du, J., Meng, W., Khayat, K. H., Bao, Y., Guo, P., Lyu, Z., Abu-obeidah, A., Nassif, H., and Wang, H., 2021. New development of ultra-high-performance concrete (UHPC). Composites Part B: Engineering, PP: 109220.

[2] Yang, R., Yu, R., Shui, Z., Gao, X., Xiao, X., Zhang, X., Wang, Y., and He, Y., 2019. Low carbon design of an ultra-high-performance concrete (UHPC) incorporating phosphorous slag. Journal of Cleaner Production, 240, PP: 118157.

[3] Dixit, A., Verma, A., and Pang, S. D., 2021. Dual waste utilization in ultra-high performance concrete using biochar and marine clay. Cement and Concrete Composites, 120, PP: 104049.

[4] Lehmann, J. and Joseph, S., Biochar for environmental management: science, technology and implementation. 2015: Routledge.

[5] Chen, T., Zhao, L., Gao, X., Li, L., and Qin, L., 2022. Modification of carbonation-cured cement mortar using biochar and its environmental evaluation. Cement and Concrete Composites, 134, PP: 104764.

[6] Roberts, K. G., Gloy, B. A., Joseph, S., Scott, N. R., and Lehmann, J., 2010. Life Cycle Assessment of Biochar Systems: Estimating the Energetic, Economic, and Climate Change Potential. Environmental Science & Technology, 44(2), PP: 827-833.

[7] Dixit, A., Gupta, S., Pang, S. D., and Kua, H. W., 2019. Waste Valorisation using biochar for cement replacement and internal curing in ultra-high performance concrete. Journal of Cleaner Production, 238, PP: 117876.

DIFFERENT APPROACHES OF CO_2 SEQUESTRATION IN CEMENTITIOUS MATERIALS MANUFACTURING

Zhuo Liu[1], Xuyong Chen[1], and Weina Meng[2]

[1]Wuhan Institute of Technology
[2]Stevens Institute of Technology
Email: liuzhuo@wit.edu.cn

Abstract

1. INTRODUCTION

In the context of the grand goal of global carbon neutrality for combatting climate changes, the sequestration of recycled CO_2 in cementitious materials attracts attention as the wide application of concrete generates huge potential for CO_2 recycling. In our studies, we proposed three different strategies to sequester CO_2 in cementitious materials. The first approach uses CO_2 to cure fresh concrete in CO_2-rich environments. The concentration of CO_2 and pressure are kept high to let CO_2 dissolve into water and react with cement to form dense carbonates on a thin surface of concrete. The second approach injects gaseous or liquid CO_2 into fresh concrete during the mixing. The injected CO_2 dissolves into the mixing water and reacts with cement to produce $CaCO_3$. The third approach is to sequester CO_2 using solid wastes before the mixing. The treated wastes are used to prepare concrete that permanently locks CO_2 inside.

2. MATERIALS AND METHODS

First, CO_2 is sequestered in cementitious materials incorporated with waste glass by carbonation curing. Two glass mortar (GM) mixtures with 20% and 40% river sand replaced by recycled glass by volume and carbonation-cured for 0 hours, 6 hours, and 12 hours at early ages were investigated. After carbonation curing, the GM prism samples were utilized for ASR expansion tests, and GM cubic samples were further moisture-cured in a water tank until testing ages for measuring compressive strength at 1d, 7d, 28d.

Second, CO_2 is sequestered in cementitious during mixing by CO_2 injection. Nano-$CaCO_3$ suspension is firstly prepared by two methods. The first method is an injection method, denoted by CI, which injects CO_2 gas into $Ca(OH)_2$ suspension to synthesize nano-$CaCO_3$. The second method is a mixing method, denoted by CP, which mixes the commercial nano-$CaCO_3$ powder with water and stirs the mixture for dispersion of the particles. Cement past mixtures were prepared by using $CaCO_3$ suspensions with various concentrations using the CI method and CP method. The performance of specimens was compared.

Third, CO_2 is sequestered in cementitious materials before mixing by carbonating waste materials. A waste material, off-specification fly ash (OSFA), is used in concrete. A modification process by coating OSFA with nano-$CaCO_3$ (CC) particles through carbonation process is employed to enhance its properties. Experimental investigations were then carried out to compare the mechanical properties of three different types of concrete incorporated with original OSFA, original OSFA and nano-CaCO3 powders, and OSFA coated with nano-CaCO3.

3. RESULTS AND DISCUSSION

Regarding the first process, for mixtures containing recycled glass, as the carbonation curing duration increases from 0 hours to 12 hours, the compressive strength at 28 days increased by

more than 10%, and ASR-induced expansion is reduced by more than 85%. The results indicate that carbonation curing can significantly improve the strength at both early and late ages and shows significant effectiveness for ASR mitigation.

Regarding the second process, compared with the plain concrete, 28-day compressive strength was increased by 16% by the CI method at an optimal concentration of 2% and by 8% by the CP method at an optimal concentration of 4%. Porosity results revealed that the microstructures of concrete prepared with $CaCO_3$ suspension were densified, and the CI method resulted in 8%-20% less porosity than the CP method.

Regarding the third process, SEM observations show that after modification, the OSFA particles are embedded with nano-level particles $CaCO_3$ particles. Compared with mixture using original OSFA, mixture using OSFA together with nano-$CaCO_3$ powders and mixture using nano-$CaCO_3$ coated OSFA both accelerated the hydration heat, promoted the compressive strength, and densified the microstructure. The coated OSFA is more effective because the coating can reinforce the weak OSFA particle and generate well-dispersed nano-$CaCO_3$ particles on the surface behaving as nucleation sites.

4. CONCLUSION

First, carbonation curing was used at early age to mitigate deleterious alkali-silica reaction (ASR) of glass-incorporated cementitious composite. It was found that the CO_2 sequestration by carbonation curing led to a 40% increase in 28-d compressive strength of glass mortar (GM) and 85% reduction in the ASR-induced expansions. Mechanism study indicated that the carbonation curing mitigated the ASR in GM by reducing calcium hydroxide content and the volume of pore solution, densifying the microstructure, and reducing the pH and free alkali metal content in the pore solution.

Second, the well-dispersed $CaCO_3$ suspension prepared by CO_2 injection was proposed as an additive in cementitious materials. The results showed that the 28-day compressive strength of cement paste was increased by 20% using 6% $CaCO_3$ suspension. Analysis of cement hydration kinetics, thermal gravimetry, pore structure, and nano particle dispersion revealed that the nano-$CaCO_3$ suspension promoted cement hydration and densified the microstructures because of the nucleation effect by the high-level dispersion of nano-$CaCO_3$ particles, and the promotion by CO2 injection is significantly better than directly adding nano-$CaCO_3$ powders.

Third, the carbonation process was employed to coat waste/porous materials with dense nano $CaCO_3$, which is upcycling as raw materials in cementitious materials. It was found, through the carbonation coating, the waste material is transformed from a harmful additive to beneficial one to concrete. Characterization of the coating showed the carbonation coating can: (1) densify the microstructure of wastes, (2) enhance compatibility between wastes and cement matrix, and (3) promote the hydration and pozzolanic reaction by the filler effect.

All carbonation approaches are approved to be effective in sequestering considerable amounts of CO_2 and enhancing the mechanical properties and durability of the cementitious materials. They are promising to contribute to a significant fraction of global CO_2 emission reduction.

Keywords
CO_2 sequestration; carbon neutrality; CO_2 curing; CO_2 injection; waste recycling

UPCYCLING OF BAGHOUSE FINES INTO ARTIFICIAL AGGREGATE FOR PERMEABLE PAVING BRICKS

Yuguang Wang[1], Bo Li[1], Weizhuo Shi[1], and Fangying Wang[2]

[1]Department of Civil Engineering, University of Nottingham Ningbo China, 199 Taikang Road, Ningbo 315100, China
[2]Department of Civil Engineering, University of Nottingham, Nottingham NG7 2RD, United Kingdom
Email: bo.li@nottingham.edu.cn

Abstract

Permeable paving bricks are widely used in the development of sponge cities. However, their production typically requires more cementitious materials than conventional concrete paving bricks. When cold-bonded artificial aggregates are used, cementitious materials are needed for both the artificial aggregates and the permeable bricks. This further increases carbon emissions and the production costs of the permeable paving bricks.

To tackle this issue, this study proposes a novel technology for producing permeable paving bricks using freshly produced artificial aggregates. The cementitious material is only needed for artificial aggregate production. These aggregates are made from baghouse fines, primarily composed of tuff and basalt powder, generated from the asphalt concrete mixing process. Moreover, alkali-activated slag, a low-carbon binder, is utilised in the production of the artificial aggregate. This study investigates the effects of aggregate grading and targeted porosity on the mechanical properties and structural integrity of the permeable bricks.

The results show that the compressive strength of the permeable paving bricks using freshly produced artificial aggregates is primarily influenced by targeted porosity, while their permeabilities are significantly affected by the artificial aggregate grading. Specifically, increasing the porosity from 10% to 15% decreases the compressive strength from 19.22 MPa to 11.11 MPa. The permeability of the bricks made from coarser artificial aggregate (12.5-16 mm) is around four times that of bricks made from fine aggregate. Overall, the bricks incorporating freshly produced artificial aggregates exhibit a balanced compressive strength of 19.22 MPa and a permeability coefficient of 2.60×10^{-2} cm/s, meeting the requirements in the national standard. This study proposes a new strategy for transforming industrial waste into sustainable construction products, contributing to both waste reduction and carbon footprint mitigation.

Keywords
Baghouse fines; artificial aggregate; permeable paving bricks

DIRECT INK WRITING OF NON-SINTERED CERAMIC WITH BIOMIMETIC CELLULAR STRUCTURE

Huang, K.[1,2], **Liu, Z.**[1,2], **Wang, F.**[2], **and Hu, S.**[2]

[1]School of Materials Science and Engineering, Wuhan University of Technology, Wuhan, 430070, China
[2]State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan, 430070, China
Email: liuzc9@whut.edu.cn

Abstract

This work reports a novel approach for fabricating 3D printed cellular ceramics under ambient conditions using CO_2-cured γ-C_2S ink via direct ink writing. The study investigates the carbonation process, microstructural evolution, mechanical performance, and failure mechanisms of hexagonal, triangular, and square cellular structures. Exposure to a CO_2-rich environment transforms the printed green body into a dense matrix of $CaCO_3$ and silica gel, eliminating high-temperature sintering. These non-sintered cellular ceramics achieve high compressive and specific strength comparable to sintered counterparts. Notably, the square-shaped structure demonstrates superior performance, reaching 104 and 171 MPa for in-plane and out-of-plane compressive strength, respectively. Further, the in-situ analysis reveals distinct deformation modes and crack propagation patterns across structures, providing insights into structure-property relationships to guide the design of high-strength non-sintered cellular ceramics. This innovative method circumvents common issues associated with high-temperature sintering (such as shrinkage, precision loss, and degradation of functional additives), which paves the way for designing high-strength non-sintered cellular ceramics.

Keywords

Cellular structure; biomimicry; CO_2 solidification; calcium silicate

ELUCIDATING HOW CO_2 INFLUENCES RHEOLOGICAL TIME-VARYING BEHAVIOR OF CEMENTITIOUS SYSTEM BASED ON IMPROVED PARTICLE LINKAGE (IPL) THEORY

Hengrui Liu[1], Shipeng Zhang[1], and Chi Sun Poon[1]

[1]Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong
Email: Shipeng.zhang@polyu.edu.hk

Abstract

In order to explain the complex physicochemical mechanism of CO_2's influence on rheological time-varying behavior, the Improved Particle Linkage (IPL) theory was proposed, which offers a comprehensive framework by considering the strength, type, and quantity of linkages within the colloidal network structure of the paste. This theory effectively distinguishes between the weak contact(α_{weak}) region, strong contact(β_{strong}) region and inherent characteristic($\gamma_{inherent}$) region, each playing distinct roles throughout the hydration process. The results indicate that the increase in rheological parameters of the carbonation paste is attributed to the increase in the quantity and strength of weak(J_1) and strong(J_2) linkages within the colloidal network, The increase in the number of J_1 and J_2 linkages within the colloidal network structure is due to the CO_2 promoting the transformation of free water to capillary water, gel water, CSH interlayer water, and chemically bound water. Furthermore, changes in ionic strength, zeta potential, and particle spacing enhance the microscale interactions between particles, such as van der Waals forces, electrostatic repulsion, and CSH adhesion forces, leading to an increase in the strength of J_1 and J_2 linkages. In a carbonized cementitious system, the α_{weak} region primarily enhances network robustness continuously during hydration, whereas the β_{strong} region becomes significant in the later hydration stages, and the $\gamma_{inherent}$ region predominantly influences the initial hydration stages.

Keywords

Particle linkage; carbonization rheology; weak contact(α_{weak}) region; strong contact(β_{strong}) region; inherent characteristic($\gamma_{inherent}$) region

EVALUATION OF VISCOELASTIC PROPERTIES IN FRESH CEMENT PASTE WITH CO_2 MIXING

Kaiyin Zhao[1], Shipeng Zhang[1], and Chi Sun Poon[1]

[1]Department of Civil and Environmental Engineering, Research Centre for Resources Engineering toward Carbon Neutrality, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong
Email: shipeng.zhang@polyu.edu.hk

Abstract

This study monitored in situ the development of viscoelasticity in fresh paste with CO_2 mixing in situ by small amplitude oscillation shear (SAOS) within 2 h. Two different stages can characterize the development of the viscoelasticity of paste over time. Isothermal microcalorimetry, Zeta potential l(ZP), X-ray diffraction (XRD), Thermal gravimetric analysis (TGA) and low field 1H nuclear magnetic resonance (NMR) measurements were used to investigate the underlying mechanisms behind these two stages. It was found that the first stage is governed by the increase of solid volume fraction and electrostatic attraction between the particles, while the rigidification of calcium carbonate networks controls the second stage. CO_2 mixing has the potential to shorten percolation time and enhance the rate of rigidity development in the slurry. Moreover, cement paste with CO_2 mixing exhibits a more pronounced effect on the yielding strain compared to the critical strain. The results provide a new insight into the microstructure build-up of carbonized cement paste at an early age.

Keywords

Small amplitude oscillation shear; CO_2 mixing; NMR relaxometry; critical strain; viscoelasticity

INNOVATIVE DEVELOPMENT OF CARBON-SINK HIGH-STRENGTH FOAM CONCRETE FOR CARBON NEUTRAL APPLICATIONS

Dingqiang Fan[1], Jian-Xin Lu[1], Chi Sun Poon[1]

[1]Department of Civil and Environmental Engineering & Research Centre for Resources
Engineering Towards Carbon Neutrality (RCRE), The Hong Kong Polytechnic University,
Hong Kong, Hung Hom, Kowloon, Hong Kong, China
Email: jxinlu@polyu.edu.hk; cecspoon@polyu.edu.hk

Abstract

The world is currently facing a significant environmental challenge due to the substantial release of CO_2. This study innovatively developed a carbonation-sink high-strength foam concrete (HSFC) for maximum CO_2 sequestration and utilization. The HSFC was produced by incorporating a fine and stable CO_2 foam into a dense paste matrix designed by the particle packing theory. After that, the carbonation curing regime was adopted to enhance the performance of HSFC, as well as realize CO_2 sequestration. A series of micro- and macro- tests were conducted to understand the synergetic mechanisms of internal and external carbonation within the HSFC system. The results revealed that the physical foam with 20% CO_2 concentration was able to produce HSFCs with excellent performance, including high strength (about twice that of normal foam concrete), excellent durability and low thermal conductivity. Moreover, CO_2 foam could trigger internal carbonation effects to further enhance the HSFC performance. On the other hand, external carbonation could also improve the performance of HSFC materials. The compressive strength and microhardness of HSFCs were improved through carbonation curing. Also, the designed HSFC exhibited low water sorptivity and high electrical resistivity, implying excellent durability of HSFC compared to conventional concretes. This was because the carbonation products could fill the micro-cracks and micro-pores within the matrix, thus improving the microhardness and durability of the HSFC. Finally, the developed HSFC also exhibited excellent carbon capture capabilities, sequestering 90 kg of CO_2 through carbonation curing and 1.1 m^3 of flue gas via the CO_2 foam by one m^3 of concrete. The heat conduction simulation demonstrated significant energy-saving potential, with the designed HSFC potentially reducing electricity consumption or carbon emissions by 20%. As a result, this innovative concrete material presents a promising strategy for advancing sustainable building practices, ultimately contributing to the objective of carbon neutrality.

Keywords
High-strength foam concrete; carbon reduction; carbonation enhancement; CO_2 foam; material performance

Session D1

Advanced Low-Carbon Technologies

APPLICATION OF “WASTES” IN GREEN ULTRA-HIGH PERFORMANCE CONCRETE: MECHANISM AND PROSPECT

Yu, Rui[1], Wu, Chiqiu[2], Lv, Wei[2], and Shui, Zhonghe[1]

[1]State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan, China
[2]Hubei Changyao New Materials Co., Ltd, Yichang, China
Email: r.yu@whut.edu.cn

Abstract

Ultra-high performance concrete (UHPC) is a relatively new building materials, which has superior durability, ductility and strengths compared to Normal Strength Concrete (NSC) and Fiber Reinforced Concrete (FRC) due to its extremely low porosity, dense matrix, high homogeneous system and the inclusion of various fibers. However, in recent years, the high material cost and significant environmental impact of UHPC have been identified as major disadvantages that limit its further development and application, especially as more attention is given to low carbon footprint development and cleaner products. To address these issues, a series of approaches have been proposed. In this study, based on the particle packing theory, various “wastes” are efficiently utilized as natural recourse to produce green UHPC, including carbonated recycled aggregates, carbonated recycled concrete fines, phosphogypsum aggregates and so on. Furthermore, the detailed mechanism for the properties and microstructure development in the developed green UHPC are discussed. Finally, based on extensive experimental results and analysis, the low carbon footprint development trend for UHPC in the near future is suggested.

Keywords

Ultra-high performance concrete (UHPC); wastes; optimized design; particles packing; carbon footprint

MORPHOLOGIC CONSTROL OF GRAPHITIC CARBON NITRIDE FOR PHOTOCATALYTIC HYDROGEN PRODUCTION

Huiqing Fan[1], Xiaobu Wu[1], Lin Lei[1], and Weijia Wang[1]

[1]State Key Laboratory of Solidification Processing, School of Materials Science and
Engineering, Northwestern Polytechnical University, Xi'an, 710072, China
Email: hqfan@nwpu.edu.cn

Abstract

Hydrogen is regarded as clean and sustainable energy sources due to its advantages of pollution-free byproducts, renewability and good combustion performance. Photocatalytic water splitting employing visible light irradiation is one of most promising approaches to produce hydrogen. Graphitic carbon nitride (g-C_3N_4), as a metal-free polymeric and visible-light active semiconductor photocatalyst, has attracted widespread research interest owing to its facile synthesis, thermal stability, environmental benign, suitable band gap (2.7 eV) and fascinating electronic property. Nevertheless, the shortcomings of pristine g-C_3N_4 photocatalyst including small surface area, insufficient light absorption, slow electron transfer rate of charge carriers and high recombination rate of electron-hole pairs, weaken its photocatalytic activity. Recently, various engineering strategies such as elements doping, constructing multiple types of nanostructures, modification with functional groups with semiconductor materials are adopted for elevating the HER and AQE of g-C_3N_4. In this work, g-C_3N_4 photocatalysts with excellent hydrogen production performance were prepared based on the construction of multi-dimensional (1D, 2D, 3D) nanostructures. Combined with cocatalyst modification and in-plane electronic structure regulation, the synergistic effect is maximized to improve the performance.

Fabrication of two-dimensional thin carbon nitride nanosheet photocatalyst by chemical vapor deposition method is deeply investigated. It is found that the 2D porous structure significantly improves the surface area, light absorption, and separation efficiency of photogenerated electron-hole pairs. Furthermore, a moderate and economical in-air CVD method was successfully developed to prepare ordered and ultra-long g-C_3N_4 nanotubes (CN-NT) photocatalysts *via* SiO_2 nanofiber templates synthesized by electrospinning technique. The increase of specific surface area, faster electron transfer pathway, enhancement of light absorption capacity, as well as the low recombination rate of photogenerated electron-hole pairs lead to the advanced photocatalytic hydrogen evolution performance. The ultimate HER of the as-prepared CN-NT can reach 4605.2 $\mu mol \cdot h^{-1} \cdot g^{-1}$, which was 33.4 times higher than that of the pristine CN. The present work highlights a facile route to develop high-performance CN-NT with excellent optical and electrical properties, which opens up a new angle of view for the synthesis of a multi-dimensional photocatalyst with 1D tubular in length and 3D in architectural structure.

Keywords

Graphitic carbon nitride; carbon ring; multi-dimensional; photocatalysis; hydrogen evolution

CARBON REDUCTION STRATEGIES FOR HIGH-PERFORMANCE LIGHTWEIGHT CONCRETE TOWARDS MODULAR INTEGRATED CONSTRUCTION APPLICATIONS

Jian-Xin Lu[1], Dingqiang Fan[1], Xuesen Lv[1], Weiyi Ji[1], Yujie Huang[1], Chi Sun Poon[1]

[1]Department of Civil and Environmental Engineering & Research Centre for Resources Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hung Hom, Hong Kong, China
Email: jianxin.lu@polyu.edu.hk; cecspoon@polyu.edu.hk

Abstract

Lightweight concrete is usually limited to non-loadbearing blocks, developing low-carbon high performance lightweight concrete (HPLC) capable of loading bearing purposes is essential to meet the demanding requirements of modern construction. Incorporating lightweight materials into sustainable ultra high-performance concrete (UHPC) can produce durable HPLC, characterized by low density, ultra high strength and superior durability. The utilization of waste glass in forms of powder and cullet was able to make a desirable UHPC binder for HPLC. The adoption of tailored high-strength core-shell lightweight aggregates contributed to developing low-carbon HPLC, which has been successfully applied in the modular integrated construction units. Moreover, combining the waste glass with other solid wastes, such as incinerated sewage sludge and bottom ashes, can develop high-strength lightweight aggregates for the HPLC production. Apart from solid wastes, waste flue gas (CO_2) bubbles were also introduced into cement paste for preparing a high-strength foam concrete. In addition, a superhydrophobic structure was designed to further improve the durability of foamed HPLC. This study provides substantial solutions for reducing carbon emissions and enhancing the performance of lightweight concrete, which would be promising for practical applications in energy-saving modular integrated constructions.

Keywords

Low-carbon lightweight concrete; carbon reduction; lightweight aggregates; foam concrete; durability

RHEOLOGICAL BEHAVIOR AND STRUCTURAL EVOLUTION OF BLAST FURNACE SLAG-BASED ALKALI-ACTIVATED PASTE WITH SAP

Beibei Sun[1] and Dengwu Jiao[2]

[1]Magnel-Vandepitte Laboratory, Department of Structural Engineering and Building Materials, Ghent University, 9052 Ghent, Belgium
[2]Department of Architecture and Civil Engineering, City University of Hong Kong, Kowloon Tong, Hong Kong SAR, China
Email: dengjiao@cityu.edu.hk

Abstract

The rapid reactivity of Alkali-activated slag (AAS) is accompanied by a series of challenges, such as volumetric stability, inadequate microstructure development, and shrinkage. Utilizing superabsorbent polymers (SAP) in AAS can promote internal curing and remarkably alleviate the shrinkage, while it also exerts significant impact on the workability of the material. As rheological properties play a pivotal role in characterizing the flowability and predicting placing process, investigating the rheology of AAS with varying SAP dosages is crucial for ensuring a balance between enhanced internal curing and the preservation of desirable workability and mechanical properties. However, there remains a scarcity of reports on the flowability and rheological characteristics of SAP-incorporated AAS. To facilitate the widespread utilization of SAP in AAS, it becomes crucial to investigate how the workability changes when the SAP dosage falls below or exceeds the required level for internal curing.

To address these problems, this research conducts a series of experiments to clarify the rheological characteristics, structural evolution, setting, and hydration behavior of AAS containing SAP with various particle sizes and contents. The results revealed that the addition of SAP increases yield stress and plastic viscosity but reduces early structural build-up rate of AAS paste. After resting for more than 30 min, however, AAS pastes with SAP exhibit an increased structural build-up rate. The second hydration peak of AAS is delayed and reduced after incorporation of SAP, despite achieving more thorough hydration at 7 d. Comparing with larger-sized SAP, AAS with smaller-sized SAP exhibits more elastic characteristics and a more challenging attainment of equilibrium. Besides, it has a longer delay and reduction in the second hydration peak, along with higher 7-day cumulative hydration heat. This study provides a theoretical support for the practical applications of SAP in AAS, specifically in terms of rheological considerations.

Keywords

Alkali-activated slag; rheology; structural evolution; setting time; hydration; SAP

A BRIEF TALK ON THE INORGANIC NANOPARTICLE ADDITIVES IN SULPHOALUMINATE CEMENT FROM SOLID WASTE-TAKING TiO_2 AND $CaCO_3$ AS EXAMPLES

Fangjie Pang[1], **Zengmei Wang**[1]

[1]School of Materials Science and Engineering, Jiangsu Key lab. of Construction Materials,
Southeast University, Nanjing 211189, PR China
Email: 101011338@seu.edu.cn; pangfangjie11@163.com

Abstract

Utilizing urban industrial solid waste to prepare sulphoaluminate cement materials has successfully addressed the challenges in urban industrial solid waste disposal. Simultaneously, it overcomes the issue of high raw material costs associated with sulfur-aluminate cement, truly achieving the concept of "turning waste into treasure." The prepared solid waste-based sulphoaluminate cement possesses advantages such as rapid setting, early strength, and resistance to sulfate erosion. Additionally, it has low CO_2 emissions, making it a focal point in current research on low-carbon green building materials. However, the porous structure and low alkalinity of solid waste-based sulphoaluminate cement make it susceptible to carbonation, leading to deterioration in mechanical properties and reduced stability, limiting its application. To address these issues, this study utilizes high-surface-area nanoscale calcium carbonate and nanoscale titanium dioxide to control the hydration process, optimize pore structure, increase the density of the cement matrix, and enhance stability. The nanomaterials also exhibit a filling effect, further optimizing the pore structure and hindering the transmission of CO_2. Additionally, nanomaterials can improve the selectivity of hydration products, generating more amorphous gels that encapsulate calcium aluminate. This reduces the decomposition of calcium aluminate due to carbonation, thereby improving the mechanical and carbonation resistance properties of the cement. By introducing water-reducing agents with surfactant functionality to regulate the adsorption degree on the surfaces of different clinker mineral phases, selective control of the carbonation of clinker mineral phases is achieved. This work provides a feasible strategy for enhancing the mechanical properties and improving the carbonation resistance of solid waste-based sulphoaluminate cement, offering important guidance for the engineering application of sulphoaluminate cement.

Keywords
sulphoaluminate cement; solid waste; ettringite; nanomaterials; carbonization process

PARTICLE PACKING MODEL OF HIGH CONTENT PHOSPHOGYPSUM CEMENTITIOUS MATERIALS WITH DENSE STRUCTURE AND HIGH SURFACE ACTIVE REACTION

Zhengkang Yu[1], **Zhonghe Shui**[1], **Chiqiu Wu**[2], **Wei Lv**[2], **Jian-Xin Lu**[3], **Xudong Zhao**[3]

[1]State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan 430070, China
[2]Hubei Changyao New Materials Co., Ltd, Yichang 443100, China
[3]Department of Civil and Environmental Engineering & Research Centre for Resources Engineering Towards Carbon Neutrality, The Hong Kong Polytechnic University, Hong Kong, China
Email: 2937703365@qq.com

Abstract

Aimed to address the low utilization rate of phosphogypsum (PG) and its challenge in resource utilization in China, this study developed phosphogypsum cementitious materials made by high-content (more than 70%) PG together with a small amount of supplementary cementitious materials (SCMs) such as cement, metakaolin granulated blast furnace slag (GBFS). According to the particle size and morphology characteristics of solid particles in cementitious material system, a new packing model is proposed. The key parameters of the model are determined by Box-Behnken design method. On this basis, the dense packing states of PG and SCMs are obtained, and the accurate ratio of two segments are confirmed. Thus, the optimal mix ratio of high-volume phosphogypsum hydraulic cementitious materials can be calculated. In addition, on the basis of theoretical analysis, this paper studies the influence of the content of SCMs on the mechanical properties and microstructure of cementitious materials. The physical properties, mechanical properties (compressive strength) and pH value of leaching solutions of high-volume phosphogypsum hydraulic cementitious materials. Besides, detailed analyses on their microstructures are made via XRD, TG, LF-NMR technologies. The experimental results show that the designed cementitious material contributes high packing density, and a layer of hydration products mainly composed of ettringite and C-S-H gel are formed on the surface of phosphogypsum particles. By changing the content of SCMs, the formation of hydration products such as ettringite are promoted, and the pore structure is optimized, and the mechanical properties of cementitious materials are improved. The design of mix proportion based on the established particle packing model is of great significance for the mass utilization of PG and the obtaining of cementitious materials with better performance.

Keywords

Phosphogypsum; particle packing model; microstructure; strength

PREPARATION OF IN SITU NANO-SILICA WITH CONTROLLED YIELD PERCENT FOR IMPROVING CEMENT PASTES PERFORMANCE

Saqib Iqbal[1,2,3], Ganghua Pan[1,2,3], and Haining Meng[3]

[1]School of Materials Science and Engineering, Southeast University, Nanjing, 211189, PR, China
[2]Jiangsu Key Lab of Construction Materials, Southeast University, Nanjing, 211189, PR, China
[3]Jiangsu Zhenjiang Research Institute of Building Science Group Co., Ltd., Zhenjiang, 212000, PR China
Email: ghpan@seu.edu.cn (G. Pan); mhningcz@163.com (H. Meng)

Abstract

The inherent agglomeration tendency of nano-silica (NS) thwarts its complete usage in cement-based products. Agglomeration concerns can be mitigated by the in-situ growth of NS directly on the surface of supporting materials. This work employed a technique to produce highly monodispersed NS particles with a uniform particle size distribution and a controllable yield percent by growing in-situ NS (NS-I) on the surface of acid-modified fly ash (FA). Surface modification characteristics of modified FA were used to avoid agglomerations of NS-I instead of using any surfactant. The morphology and dispersion of NS-I were investigated by SEM and TEM. The yield percent of NS-I was regulated, and the highest yield percent achieved was used to prepare the cement pastes. The synthesized NS-I exhibited superior fresh and dense microstructure due to its excellent dispersion, outperforming the commercial NS (NS-C) in terms of nano reinforcements in cementitious materials. This work may offer fresh approaches to the modification of mineral admixtures for improved NPs preparation and dispersion with controlled yield percent.

Keywords
Commercial NS; original FA; acidic-surface modification; dispersion; cement pastes

THE SOLIDIFICATION OF Pb, Zn AND Cd BY SPONTANEOUS COMBUSTION GANGUE GEOPOLYMER AND THE DEPOLYMERIZATION AND RECONSTRUCTION MECHANISM

Xiao Han[1], Baomin Wang[1]

[1]School of Civil Engineering, Dalian University of Technology, Dalian 116024, PR China
Email: 17861500110@163.com

Abstract

MSWI fly ash is classified as hazardous wastes (HW18) in the National Hazardous Waste List, because it is rich in heavy metals (such as Cd, Pb, Cr and Zn etc.), and requires proper treatment before further landfill disposal. In addition, Coal gangue (CG) is one of the largest amounts of industrial wastes in the world, and spontaneous combustion is likely to occur when it reaches an ignition point. This produces a large number of toxic and harmful gases such as CO, CO_2, SO_2 and NOx, which are discharged into the atmospheric environment, and heavy metal elements such as arsenic, mercury, fluorine, lead and selenium are released into the soil and groundwater. In this paper, the physical properties and chemical properties of spontaneous combustion gangue and MSWI fly ash were were tested and analyzed. And the types, contents, leaching toxicity, occurrence forms and chemical valence of heavy metals in MSWI fly ash were analyzed. Zn, Pb and Cd in MSWI fly ash were determined as the key immobilization objects of this research. Then, the central composite design experiment of response surface was carried out under the two chemical activators of $K_2O{\cdot}nSiO_2$+KOH and $Na_2O{\cdot}nSiO_2$+NaOH with the setting time, fluidity and compressive strength as the objective function, and the optimum ratio was determined. According to the total content of Pb^{2+}, Zn^{2+} and Cd^{2+} in MSWI fly ash, Pb^{2+}, Zn^{2+} and Cd^{2+} were added to the geopolymer of spontaneous combustion gangue in proportion. The effects of Pb^{2+}, Zn^{2+} and Cd^{2+} on hydration heat, hydration products, molecular structure characteristics, microstructure and interfacial bonding in the depolymerization and reconstruction of spontaneous combustion gangue geopolymer are tested by isothermal calorimeter, XRD, TG-DTA, FTIR, RAMAN, XPS, SEM.

Results shows that: the compressive strength of spontaneous combustion gangue geopolymer prepared with $K_2O{\cdot}nSiO_2$+KOH were higher that of $Na_2O{\cdot}nSiO_2$+NaOH. And The significance of the influencing factors both were sodium silicate modulus＞Alkali/ CG＞W/CG. The best when the content of Pb, Zn and Cd is 0.18%, 1.22% and 0.09%, and the curing rate of 90 d reaches 92.13%, 96.96% and 99.81% respectively. In addition, as shown in Fig.1,the chemical form of heavy metals is dominated by bound to carbonate at 3d, and gradually concentrated in bound to Fe-Mn oxides and residue with the increase of age. From Fig.2- Fig.4, the crystal peak in the geopolymer of spontaneous combustion coal gangue is mainly SiO_2, amorphous peak appeared in the range of 20 ° ~ 40 ° (2θ), and the flocculent amorphous gel was also observed in SEM. The higher the compressive strength, the wider the stretching vibration peaks of Si-O and Al-O at 1200-950cm^{-1}. And Na^+ and K^+ have a synergistic effect, and it is difficult for K^+ to move out because of its large radius, which can continuously and effectively promote the Polycondensation between SiO_4 and AlO_4 tetrahedron. The main structural bonds of spontaneous combustion coal gangue are Si-O and Al-O, which exist in the form of $[SiO_4]^{4-}$ and $[AlO_4]^{5-}$, respectively, and SiO_2 and Al_2O_3 have strong solubility. The polymerization process of spontaneous combustion coal gangue can be expressed by the following chemical equations:

$$2SiO_2+6Ca(OH)_2+aq \longrightarrow CaO\cdot 2SiO_2\cdot nH_2O+3Ca(OH)_2 \quad (1\text{-}1)$$
$$Al_2O_3+3Ca(OH)_2+3CaSO_4+23H_2O \longrightarrow 3CaO\cdot Al_2O_3\cdot 3CaSO_4\cdot 31H_2O \quad (1\text{-}2)$$
$$Al_2O_3+Ca(OH)_2+CaSO_4+9H_2O \longrightarrow 3CaO\cdot Al_2O_3\cdot CaSO_4\cdot 12H_2O \quad (1\text{-}3)$$

Under the action of alkaline solution, the covalent bonds of Si-O-Si and Al-O-Al are depleted and broken and forming ions into the alkaline solution, so that the structure of coal gangue was disintegrated. With the continuous reaction in formula (1-3) and (1-2), Al^{3+} occupy the position of Si^{4+}, and $[SiO_4]^{4-}$ and $[AlO_4]^{5-}$ are connected by tetragonal oxygen atoms in the structure to form a three-dimensional network structure, which is called a three-dimensional polyaluminate structure. Because Al^{3+} are trivalent cations with negative charge around, the alkali metal cations such as Na^+ and K^+ with positive valence are filled in the gap of amorphous gel in the way of equilibrium charge, thus a relatively stable gel structure is obtained.

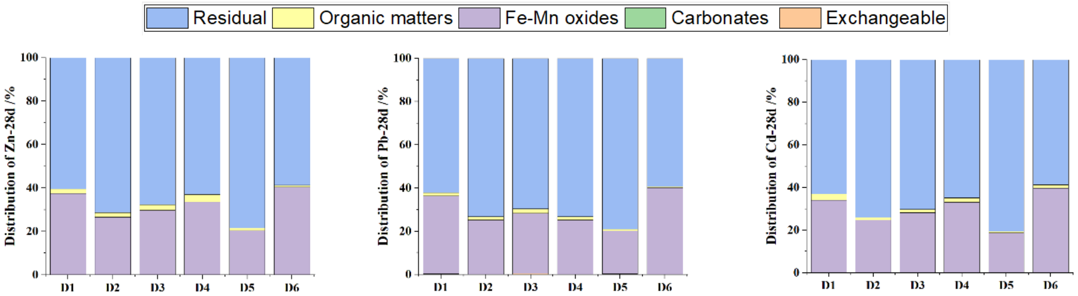


Fig.1 The chemical form of Pb, Zn and Cd in spontaneous combustion gangue geopolymer

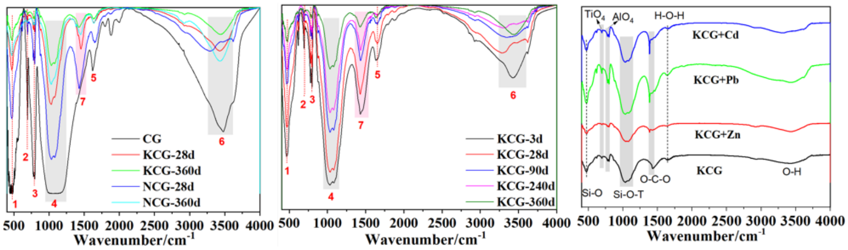


Fig.2 FTIR of Pb, Zn and Cd in spontaneous combustion gangue geopolymer

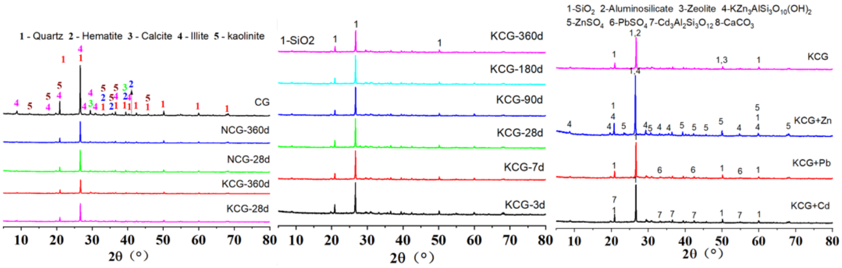


Fig.3 XRD of Pb, Zn and Cd in spontaneous combustion gangue geopolymer

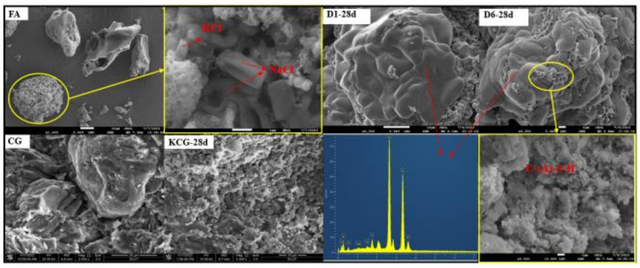


Fig.4 SEM and EDS of Pb, Zn and Cd in spontaneous combustion gangue geopolymer

Keywords

Spontaneous combustion gangue; heavy metals; solidification mechanism; geopolymer

SYNTHESIS OF VATERITE VIA WIND-SUSPENDED CARBONATION

Kuizhou Liu[1], **Tung-Chai Ling**[1]

[1]College of Civil Engineering, Hunan University, Changsha 410082, Hunan, China
Email: tcling@hnu.edu.cn; tcling611@yahoo.com

Abstract

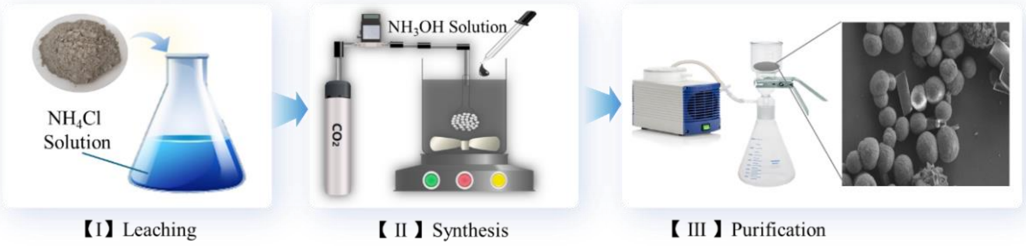
Steel slag (SS) is an industrial solid waste produced in the process of steel making, and the stockpile of steel slag in China is over 150 million tons per year. A large amount of stockpiled steel slag not only encroach on the land, but also cause serious environmental pollution such as soil alkalization. Most of the steel slag in China is converter slag, containing a large amount of CaO (34% ~ 48%) MgO (2.5% ~ 10%) and other alkaline oxides, and can easily react with CO_2 to form stable calcium carbonate.

Vaterite as one of the three crystal types of $CaCO_3$ crystals possess a higher value that calcite and aragonite for most industrial applications. This is because vaterite has good dispersibility, fluidity and high specific surface area, and has important applications in plastics, rubber and paper industries. Morever, in the field of biomedicine and pharmacology, vaterite can be used for the preparation of hydroxyapatite, which can enhance the mechanical strength of bone and dental implants, promote the transformation and proliferation of cells to osteoblasts as well as a drug carrier. Therefore, SS as a source of Ca for the preparation of vaterite is a high value-added utilization pathway.

Current studies usually adopt leaching-carbonation approach to prepare vaterite (Fig. 1), and the main steps involved are as follows: [I] leaching: using NH_4Cl solution to leach Ca^{2+}; [II] synthesis: using CO_2 gas to mineralize the calcium ions in the solution ; [III] Purification: filtering and drying to obtain high purity of vaterite. In the process [II], crystal shape modifiers need to be added to regulate the crystal shape, and common crystal shape modifiers include $NH_3.H_2O$, amino acids and etc. This method involves the consumption of a significant amount of NH_4Cl solution during the leaching of Ca^{2+}, leading to a production process that is not resource-efficient.

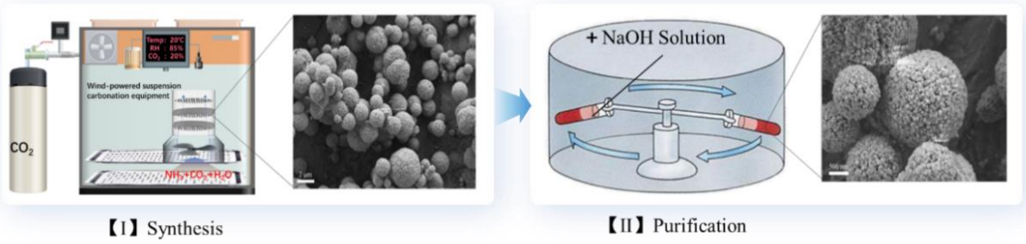
In this paper, we propose a process for the preparation of vaterite by wind-powered suspension carbonation (Fig. 2). The specific steps are as follows, [I] Synthesis: using as-received SS as carbon precursor, the SS is suspended by wind power in a CO_2 rich atmosphere. With presence of NH_3 and moisture, contact efficiency and carbonation reaction rate can be greatly enhance to form vaterite. [II] Purification: dissociating the silica network structure with low concentration NaOH solution and release the agglomerated vaterite. Finally, a high purity vaterite can be obtained by using high speed centrifugation and drying.

The optimal synthesis parameters were determined by orthogonal tests, with $NH_3.H_2O$ addition of 0.2 ml/g, wind speed of 100 m^3/h, and carbonation duration of 30 min. The resulting vaterite were of good sphericity, with particle sizes of 6-10 μm, and purity of up to 90%, and the long-term hydro-stability of sub-stable vaterite was strengthened by a crystalline control agent. In comparison to the traditional method, the wind-suspended carbonation method for vaterite preparation streamlines the process, reduces leaching material enhance vaterite synthesis efficiency, and enables integrated vaterite preparation.



【I】 Leaching 【 II 】 Synthesis 【 III 】 Purification

Fig. 1 Leaching – Carbonation



【I】 Synthesis 【II】 Purification

Fig. 2 Wind-suspended carbonation

Keywords
Steel slag; wind-suspended carbonation; vaterite; carbon sequestration

DEVELOPMENT OF VALUE-ADDED AEROGEL FROM WASTE GLASS BY CO_2 EXTRACTION FOR LIGHTWEIGHT INSULATING CONCRETE: TOWARDS ENERGY CONSERVATION IN BUILDINGS

Xudong Zhao[1], Jian-Xin Lu[1], and Chi Sun Poon[1]

[1]Department of Civil and Environmental Engineering & Research Centre for Resources Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hong Kong
Email: 24053742r@connect.polyu.hk

Abstract

The construction industry is pivotal in global energy consumption and greenhouse gas emissions. With the growing global interest in sustainable practices and energy conservation, lightweight and thermal insulative concrete has attracted increasing interest in the energy efficiency of buildings. However, to achieve high-performance applications in green and near-zero energy buildings, it becomes imperative to further reduce the density and thermal conductivity of these materials. Meanwhile, managing post-consumer beverage glass containers has become an acute waste management issue in Hong Kong. A considerable amount of waste glass containers is generated (211 tonnes/daily in 2022), but the recycling rate is very low at only 20% due to the lack of local glass industry. Therefore, how to provide viable outlets for the collected waste glass has become a critical element of the waste management framework.

This study explores an innovative approach to develop a high-value-added aerogel material from waste glass by CO_2 extraction to encompass the reduction of energy consumption in buildings, heightened utilization of waste glass, and enhanced CO_2 capture. For this purpose, a new process will be developed to convert waste glass into aerogels. The resulting aerogels are characterized by low density, high porosity and impressive thermal insulation properties. By incorporating these aerogels into lightweight insulating concrete, thermal conductivity and density can be further reduced, energy efficiency can be increased, and the overall performance of building materials can be improved. This approach promotes the reutilization of waste glass by diverting it from landfills, thereby mitigating the environmental repercussions associated with waste glass. The CO_2 extraction process simultaneously facilitates greenhouse gas capture, aligning with global climate change mitigation efforts. Moreover, integrating glass aerogel into lightweight insulating concrete has the potential to improve the mechanical properties and durability of concrete due to the extremely high surface area and fine particle size of aerogel, which will accelerate the hydration of cement and the pozzolanic reaction of calcium hydroxide.

The development of this aerogel lightweight concrete exhibits complex physicochemical processes and interdisciplinary research involving aerogel synthesis, cement chemistry, CO_2 sequestration, life cycle assessment, and energy efficiency evaluation. The challenges lie in the mechanism exploration of silica gel extraction from waste glass by CO_2 catalyst, hydration kinetics and aerogel-binder interface characterization of aerogel lightweight concrete. Furthermore, the mechanical properties, durability, thermal insulation and energy-saving potential of aerogel-based lightweight concrete will be thoroughly investigated.

Keywords
Lightweight concrete; waste glass; silica aerogel; CO_2 sequestration; energy efficiency

Session D2

Mineral Carbonation Materials as SCMs

ENHANCING INTERNAL CURING IN ALKALI-ACTIVATED SLAG WITH SUPERABSORBENT POLYMER AND LIMESTONE POWDER

Xinyan Liu[1], Bo Li[1], Yung-Tsang Chen[1], Weizhuo Shi[1], and Fangying Wang[2]

[1]Department of Civil Engineering, University of Nottingham Ningbo China, 199 Taikang Road, Ningbo 315100, China
[2]Department of Civil Engineering, University of Nottingham, Nottingham NG7 2RD, United Kingdom
Email: bo.li@nottingham.edu.cn

Abstract

Alkali-activated slag (AAS) has been recognised as a green binder possessing excellent mechanical properties and chemical resistance. Meanwhile, the AAS materials show higher autogenous shrinkage than ordinary Portland cement (OPC) materials, which increases the cracking tendency and limits their wide applications in the construction industry. It is well-known that the autogenous shrinkage of AAS materials can be effectively mitigated by internal curing using superabsorbent polymer (SAP). However, there are lack of studies focusing on the effects of SAP properties and curing liquid diffusion on the internal curing effectiveness of AAS materials. Therefore, the paper first investigates the effects of SAP dosage, particle size, addition method, and absorption status on the internal curing effectiveness of AAS pastes. Afterwards, the pore structure of AAS pastes is modified by adding limestone powder (LP) to enhance the internal curing effectiveness.

The results indicate that incorporating SAP reduces the autogenous shrinkage of AAS pastes, particularly for those with more SAP particles and larger-size SAPs. The gradually released activator from SAP promotes the geopolymerisation of the paste around the SAP particles, contributing to the reduction of autogenous shrinkage driving force. This mitigation is achieved by the increased local restraint and reduced fraction of pores that can drive autogenous shrinkage. Moreover, the large SAP particles achieve a farther travel distance, enhancing the internal curing effectiveness for AAS paste. The appropriate addition of limestone powder in the pastes with SAP increases the internal curing effectiveness by reducing the fraction of mesopores in the paste with SAP. Furthermore, adding SAP maintains the moisture supply in the mixtures and increases the size of minimum pores that can drive autogenous shrinkage, collectively contributing to mitigating the autogenous shrinkage of the pastes. Overall, this study offers strategies on enhancing the internal curing effectiveness for AAS pastes with SAP, specifically through the optimisation of SAP properties and the pore structure of AAS pastes.

Keywords

Alkali-activated slag; autogenous shrinkage; internal curing; limestone powder; superabsorbent polymer

DEVELOPMENT OF LOW-CARBON CONCRETE MATERIALS BY REUSING THE FINE FRACTION OF CONCRETE WASTE

Wenwen Lian[1], Danni Qu[1], Jiefeng Liang[1], Zhikang Xia, Xiaoliang Fang[1]

[1]School of Civil & Environmental Engineering and Geography Science, Ningbo University,
Ningbo, Zhejiang, China
Email: fangxiaoliang@nbu.edu.cn

Abstract

Concrete waste derived from construction and demolition waste during the urban renewal consumes landfill capacity and causes a series of environmental issues of major cities. The enhancement and reuse of recycled concrete aggregate improved remarkably over the past decade, but the fine fraction of the concrete waste is still lack of practical upcycling solution. This study utilized the concrete waste with particle sizes less than 5 mm to produce both cementitious materials and fine aggregates for a type of low-carbon concrete.

The 0.15-5 mm fractions was treated by a wet carbonation process and used to replace river sand as fine aggregate. The investigation of the ITZs between the adhered old mortar and the new mortar showed a reduction in porosity and increases in microhardness and hydration products, which suggested enhanced ITZs after wet carbonation.

The powder fraction (<0.15 mm) was used to produce cementitious materials through carbonation and thermal treatment procedures. The combination of the two activation approaches provided SCMs to replace Portland cement and fillers to improve the hydration.

A series of experiments, microscopic observations and comparative analysis were conducted, including Isothermal Conduction Calorimetry (ICC), XRD, XRF, FTIR, TGA, FTIR and SEM. The results indicated that the fine fraction of concrete waste can be recycled to produce low-carbon concrete through carbonation.

Keywords

Recycled fine aggregate; carbonation; low-carbon concrete; supplementary cementitious materials; concrete waste

HYDRATION BEHAVIOR OF CARBONATED WASTE PASTE CALCINED CLAY CEMENT

Qing Liu[1], Yu Yan[1], Yuchen Hu[1], Qiang You[1], Guoqing Geng[1]

[1]Department of Civil and Environmental Engineering, National University of Singapore,
117576, Singapore
Email: liuqing@nus.edu.sg

Abstract

Concrete-based building materials offer significant potential for carbon capture, utilization and storage due to their alkaline nature. Herein, we combine the CO_2 mineralization in the waste cement paste (WCP) with calcined clay cement to develop a novel low-carbon cement—carbonated waste paste calcined clay cement (CWPC^3). 1 kg WCP can mineralize 0.278 kg CO_2 within 2 h, as well as producing silica-alumina gel. The early hydration of cement is promoted due to the nucleation effect and the high pozzolanic reactivity of CWP. There is a competition between metakaolin and silica-alumina gel, with the latter being more apt to react with portlandite. Silica-alumina gel distributes randomly in the matrix in the form of a cluster and reacts fast, resulting in high early compressive strength. CWPC^{3}50 demonstrates a greater environmentally friendly compared to LC^{3}50 in reducing CO_2 emissions. Hopefully, our work can shield light on developing a novel low-carbon cement.

Keywords

Carbonation; carbonated waste paste; hydration; low-carbon cement; compressive strength

Acknowledgements

This work was financially supported by the Singapore MoE Tier1 project (A-0009302-01-00), Singapore Energy Centre Innovation project (A-8000053-01-00), Jointly Cultivating Doctoral Students by the National Foundation for Overseas Studies project (202108340063).

CORRELATION BETWEEN STRENGTH AND NON-DESTRUCTIVE ULTRASONIC MEASUREMENTS ON EARLY AGE CARBONATED BOF SLAG

Franco Santos, W.[1], Brouwers, H.J.H.[1]

[1]Department of the Built Environment, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, the Netherlands
Email: w.franco.santos@tue.nl

Abstract

Basic oxygen furnace (BOF) slag is a by-product of steel making. Its utilization as a cementitious binder is limited by its low hydraulic properties. The disposal of BOF slag (BOFS) at landfills wastes valuable resources causing environmental pollution. The direct replacement of BOFS in Portland cement (PC) without any treatment decreases the strength of the final product. Researchers have found that carbonation can improve the properties of BOFS. In the present paper carbonation treatment is carried out to improve the hydration process of BOFS with the aim to study the effects of carbonation on the early-age fresh state and strength development to better understand its performance. It highlights a correlation between a destructive test, compressive strength, and a non-destructive test, an ultrasonic wave transmission test (UWTT), investigated for up to 7 days. Carbonated BOFS-cement composite was designed according to the European standard EN196-1 with 25 wt% replacement in PC. In addition, the changes in particle size and phase analysis of the BOF slag before and after carbonation by thermogravimetry are also presented. As the relation between the destructive compressive strength test and the non-destructive UWTT is now established, the results can be used to assess the structural integrity of carbonated-BOFS composites in the fresh state.

Keywords

BOF slag; carbonation; strength development; ultrasonic testing; pulse velocity

TURNING MSWI BOTTOM ASH INTO VALUABLE SCM VIA A COMBINATION OF ALKALINE ACTIVATION AND EARLY-AGE AMBIENT-PRESSURE CARBONATION CURING

Yu, Zhe[1], **Xian, Xiangping**[1]

[1] Department of Architecture and Civil Engineering, City University of Hong Kong, Hong Kong SAR, China
Email: xiangping.xian@cityu.edu.hk

Abstract

The combination of alkaline activation and early-age carbonation curing presents great potential to enhance the properties of blended cement with municipal solid waste incineration (MSWI) bottom ash as supplementary cementitious materials (SCMs). In this study, alkalis including calcium hydroxide and sodium hydroxide were used as both chemical pretreatment agents and accelerators, aiming to finally improve the quality of paste samples. The result showed that pretreatment by removing metallic aluminum and zinc is necessary to minimize the risk of detrimental effects including volume expansion and strength loss of the blended cement matrix. Besides, despite sodium hydroxide being more effective in the removal of metallic aluminum and zinc than calcium hydroxide, both alkalis could reduce the setting time. In terms of preconditioning, blended cement paste with calcium hydroxide exhibited the highest water loss rate than other batches, which would affect the CO_2 uptake and compressive strength of paste samples. Multiple microstructural techniques including qualitative X-ray diffractometer (QXRD), thermogravimetric analysis (TGA), and so on were also conducted to characterize the combination effect of alkaline activation and carbonation curing on the enhanced properties, which was attributed to the generation and precipitation of carbonates and their intermingling effect with other phases. In total, calcium hydroxide could be considered an effective accelerator to turn waste into valuable construction products combining the introduction of early-age ambient-pressure carbonation.

Keywords

Carbonation curing; MSWI bottom ash; cement; calcium hydroxide; sodium hydroxide

COMPARISON OF EVALUATION METHODS FOR POZZOLANIC REACTIVITY OF STEEL SLAG WITH AND WITHOUT CARBONATION

Yuanyuan Shen[1], **Tung-Chai Ling**[1]

[1]College of Civil Engineering, Hunan University, Changsha 410082, Hunan, China
Email: corresponding_ tcling@hnu.edu.cn; tcling611@yahoo.com

Abstract

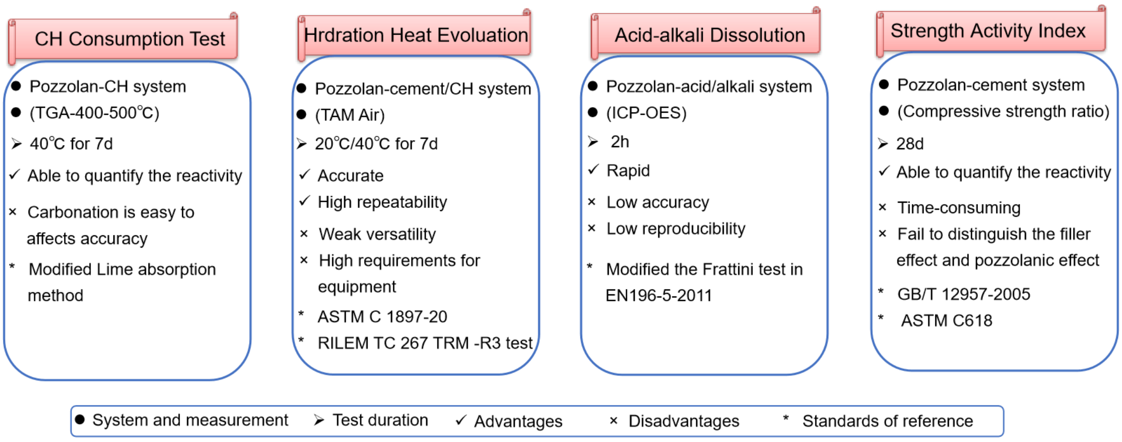
The utilization of steel slag powder (SSP) and carbonated steel slag powder (C-SSP) as supplementary cementitious materials (SCMs) can enhance the sustainability of the cement industry. However, compared to traditional SCMs, the reactivity of these SCMs on the physicochemical properties of blended cement pastes remains unclear. Therefore, it is important to identify suitable or optimal characterization methods for SSP and C-SSP as SCMs to better evaluate their performance evolution in blended cement systems.

This study aims to comprehensively compare four widely used pozzolanic reactivity evaluation methods to assess the reactivity of SSP and C-SSP in blended cement pastes. These methods include calcium hydroxide (CH) consumption, hydration heat evolution, acid-alkali dissolution, and strength activity index. As shown in Figure 1, the characteristics, advantages, and disadvantages of these methods, along with the test instruments and reference standards, are shown based on a review of literature data. Basic oxygen furnace slag (BOFS) and electric arc furnace slag (EAFS), both before and after carbonation (C-BOFS and C-EAFS), were used to replace part of Portland cement (PC) for blended pastes evaluation.

Figure 2 shows the results of the pozzolanic reactivity for SSP and C-SSP using four test methods. The CH consumption test results showed that the CH consumption ratios of C-BOFS and C-EAFS were 30.32% and 14.54%, respectively, with a standard deviation of less than 5% for triplicate measurements. However, for slag containing CH and hydrate components, the CH consumption method cannot solely used as a quantitative evaluation method to determine their pozzolanic reactivity. The reaction potential of the C-SSP-PC system and C-SSP-CH system were evaluated by monitoring the reaction heat release, and to establish the hydration kinetic model of SSP-PC. The acid-alkali dissolution method results showed that the concentration of SiO_2 and Al_2O_3 ions gradually decreased after carbonation, indicating that carbonation inhibited the leaching of Si and Al ions in SS. This is mainly due to the formation of a barrier layer on the SSP surface. The activity rate $K\alpha$ values were calculated by the formula to be 16.17%, 12.70%, 6.53%, and 4.62% for BOFS, C-BOFS, EAFS, and C-EAFS, respectively. The results of the strength activity index indicated that the activity indices of BOFS and C-BOFS were 88.54% and 92.80% respectively.

These comprehensive measurements quantified the pozzolanic reactivity of SSP and C-SSP, elucidating the potential reaction mechanisms that govern their behavior in cementitious systems. Notably, the effects of accelerated carbonation treatment on the reactivity of steel slag were revealed. The insights gained contribute to a better understanding of how the physical and chemical properties and performance of cement are influenced by the inclusion of SS and C-SS as SCMs. This knowledge paves the way for optimizing steel slag utilization to enhance sustainability and tailor the performance characteristics of blended cement.

Keywords
Steel slag powder (SSP); carbonated steel slag powder (C-SSP); pozzolanic reactivity; supplementary cementitious materials (SCMs); hydration



CH Consumption Test

- ● Pozzolan-CH system
- ● (TGA-400-500℃)
- ➢ 40℃ for 7d
- ✓ Able to quantify the reactivity
- × Carbonation is easy to affects accuracy
- * Modified Lime absorption method

Hrdration Heat Evoluation

- ● Pozzolan-cement/CH system
- ● (TAM Air)
- ➢ 20℃/40℃ for 7d
- ✓ Accurate
- ✓ High repeatability
- × Weak versatility
- × High requirements for equipment
- * ASTM C 1897-20
- * RILEM TC 267 TRM -R3 test

Acid-alkali Dissolution

- ● Pozzolan-acid/alkali system
- ● (ICP-OES)
- ➢ 2h
- ✓ Rapid
- × Low accuracy
- × Low reproducibility
- * Modified the Frattini test in EN196-5-2011

Strength Activity Index

- ● Pozzolan-cement system
- ● (Compressive strength ratio)
- ➢ 28d
- ✓ Able to quantify the reactivity
- × Time-consuming
- × Fail to distinguish the filler effect and pozzolanic effect
- * GB/T 12957-2005
- * ASTM C618

● System and measurement ➢ Test duration ✓ Advantages × Disadvantages * Standards of reference

Figure 1 Comparison of four methods for evaluating pozzolanic reactivity of SSP and C-SSP

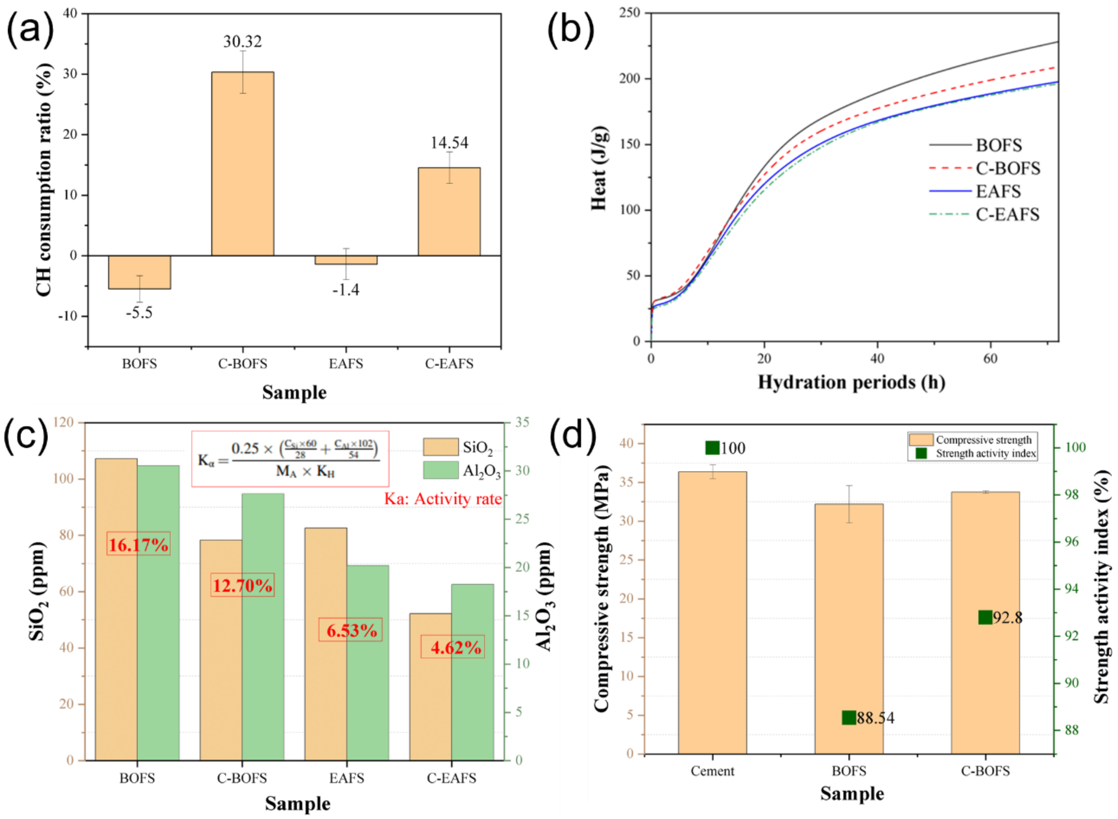


Figure 2 (a) CH consumption test results; (b) Cumulative heat release curves; (c) Acid-alkali dissolution results; (d) Strength activity index

A NOVEL INTERNAL CARBONATION METHOD FOR UTILIZATION OF STEEL SLAG-BASED BINDER: STRENGTH, MICROSTRUCTURE, AND IN-SITU CARBON MIGRATION

Weiwei Chen[1], Peiliang Shen[1], Qinglong Qin[1], Yong Tao[1], Faqian Liu[2], Chi Sun Poon[1]

[1]Department of Civil and Environmental Engineering, Research Centre for Resources Engineering toward Carbon Neutrality, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong
[2]School of Chemical Engineering and Technology, Sun Yat-sen University, Zhuhai 519082, China
Email: peiliang.shen@polyu.edu.hk and cecspoon@polyu.edu.hk

Abstract

A novel internal carbonation method is proposed in this study for synthesizing high-strength steel slag-based binder, using carbonated recycled concrete fines (CRCF) as calcium carbonate carriers and sodium meta-aluminate (SMA) solution as a chemical activator. The results demonstrate that the internal carbonation accelerates hydration and densifies microstructure of steel slag paste, reducing setting time by 84% and enhancing compressive strength by 5.4 times. During the process, the CRCF is rapidly dissolved upon activation of SMA solution, releasing carbonate ions to react with katoite and portlandite first and then the larnite in steel slag. After the internal carbonation, the main carbonated phases are Mc and calcite. The resulting C-A-S-H gels is less polymerized than that in the SMA-activated steel slag due to the incorporation of released Ca^{2+} ions from CRCF. Compared to accelerated carbonation curing, this approach is not constrained by product size, offering greater flexibility for field applications.

Keywords

Internal carbonation; steel slag paste; recycled concrete fines; calcium carbonate carriers; chemical activator; field applications

Acknowledgement

The authors wish to gratefully thank the financial support of the Innovation and Technology Fund and the Guangdong-Hong Kong-Macao Joint Innovation Field Research Foundation (2023A0505010011). The equipment support from the University Research Facility on Chemical and Environmental Analysis (UCEA) at the Hong Kong Polytechnic University is also acknowledged.

DEVELOPMENT OF REACTIVE CARBONATE-CALCINED CLAY-CEMENT (C4) COMPOSITES THROUGH SYNCHRONIZING ALUMINATE-CARBONATE REACTION: TOWARD HIGH COMPRESSIVE STRENGTH AND LOW CARBON EMISSION

Yingliang Zhao[1], Yong Zheng[1], Kai Cui[1], Peiliang Shen[1], Chi Sun Poon[1]

[1]Department of Civil and Environmental Engineering & Research Centre for Resources Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hong Kong, China
Email: peiliang.shen@polyu.edu.hk; cecspoon@polyu.edu.hk

Abstract

Limestone calcined clay cement (LC3) offers a promising strategy for producing cement with significantly reduced CO_2 emissions. However, it generally exhibits limited early-age compressive strength due to the uncoordinated reaction between limestone and aluminates, which is a consequence of the low reactivity of limestone. Moreover, further reducing the CO_2 emissions of LC3 is a challenges. This study presents an innovative method for producing carbon-fixing calcium carbonates (Cc) with varied reactivities and highly reactive Si-Al gels via the mechano-carbonation of recycled concrete fines (CRCF), aimed at replacing limestone in LC3 production. This approach seeks to develop a high compressive strength and low-carbon cement composite, designated as Reactive **C**arbonate-**C**alcined **C**lay-**C**ement (***C4***). The results demonstrate that ***C4*** shows a sustained improvement in compressive strength, with increases of over 30% and 17% at 1 d and 28 d, respectively, compared to the LC3. The superior early-age performance of ***C4*** is attributed to a pivotal "reaction window" between the highly reactive Cc and Si-Al gels, which facilitates the generation of monocarbonate (Mc) due to the synchronized aluminate-carbonate reaction. The early precipitated Mc and calcium-silicate-hydrate (C-S-H) gels from the pozzolanic reaction of Si-Al gels serve as seedings, facilitating a higher cement reaction degree and the early strength development. Moreover, the moderately reactive Cc in CRCF still exhibits higher reactivity than limestone, enabling a more synchronized reaction with calcined clay at the later stages. This contributes to the formation of a more compact microstructure, thereby favoring the continued strength development. Moreover, ***C4*** offers an additional advantage of reducing CO_2 emissions by over 50% in comparison to LC3, thus contributing significantly to environmental sustainability.

Keywords

Limestone calcined clay cement; carbonation; reaction window; Si-Al gels; synchronous reaction

DEVELOPMENT OF HIGHLY ACTIVE CALCIUM CARBONATE THROUGH ANHYDROUS CARBONATION: INFLUENCE ON THE PERFORMANCE AND HYDRATION OF SULFOALUMINATE CEMENT-BASED MATERIALS

Kai Cui[1], Yong Zheng[1], Yingliang Zhao[1], Peiliang Shen[1], and Chi Sun Poon[1]

[1] Department of Civil and Environmental Engineering & Research Centre for Resources Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hong Kong, China
Email: peiliang.shen@polyu.edu.hk; cecspoon@polyu.edu.hk

Abstract

Calcium carbonate-based materials, when used as supplementary cementitious materials (SCMs) can enhance the performance of sulfoaluminate cement (SAC), and this enhancement is closely related to the activity of calcium carbonate. This study introduced a novel method named anhydrous carbonation (AC). This method involves the introduction of CO_2 during the grinding stage of cement production, leading to the in situ generation of highly active metastable calcium carbonate on the surface of cement clinker. Subsequently, the mechanical properties, hydration kinetics, and microstructural evolution of SAC after AC were investigated. The results indicate that AC accelerated the hydration process of SAC samples and significantly enhanced both early and later compressive strength. Specifically, compared to the reference sample (Ref), the 1-day compressive strength of AC-0.5h, AC-1h, AC-2h, and AC-3h increased by 10.4%, 17.9%, 28.0%, and 27.6%, respectively. Additionally, the 28-day compressive strength increased by 7.7%, 15.4%, 27.2%, and 20.9%, respectively. AC altered the hydration kinetics of SAC. During the early hydration stage (1 day), with sufficient gypsum present, $C_4A_3\check{S}$ reacted with water and formed AFm. Simultaneously, the in situ generated metastable calcium carbonate reacted with $C_4A_3\check{S}$ and produced monocarbonate (Mc), hemicarbonate (Hc). In the later hydration stage (28 days), AFm reacted with metastable calcium carbonate, forming Mc and Hc, further stabilizing AFt. The enhancement of SAC performance after AC is primarily attributed to several factors: the provision of nucleation sites for hydration products by metastable calcium carbonate, its participation in hydration reactions to form Mc and Hc, the indirect stabilization of AFt, the filling effect of metastable calcium carbonate as an inert filler, the refinement of the pore structure, and the refinement in cement clinker particle.

Keywords

Anhydrous carbonation; metastable calcium carbonate; hydration kinetics; accelerated hydration

SYNERGISTIC EFFECT OF POZZOLANIC ACTIVITY OF RECYCLED RED BRICK FINES AND CARBONATED RECYCLED CONCRETE FINES AS HIGHLY ACTIVE SUPPLEMENTARY CEMENTITIOUS MATERIAL

Yong Zheng[1,2], **Kai Cui**[1], **Yingliang Zhao**[1], **Peiliang Shen**[1], **Chi Sun Poon**[1]

[1]Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong
[2]Research Center of Space Structures, Guizhou University, Guiyang 550025, China
Email: peiliang.shen@polyu.edu.hk; cecspoon@polyu.edu.hk

Abstract

Recycled hybrid fines (RHF) consisting of recycled concrete fines (RCF) and red brick fines (RBF) from construction & demolition wastes (C&D) are limited its resource utilisation due to its low activity. In this study, the wet carbonation technique was employed to enhance the pozzolanic activity of RHF to promote its utilisation as a highly active supplementary cementitious material (SCM) in concrete production, which not only saves costs but also promotes the resources utilisation of C&D. The evolution of phase assemblages and microstructures, and the carbonation kinetics of RHF during wet carbonation were comprehensively investigated. As the content of RBF increases, the volcanic ash activity of carbonated RHF (CRHF) is kept between 65% and 70%, and its water consumption, to achieve the standard consistency, is gradually reduced from 87% to 48%. There are four significant stages in the carbonation process of RHF. From the first to the fourth stage, the carbonation reaction is controlled by the rate of CO2 dissolution, the rate of calcium dissolution in the hydration products, hydration products dissolution, and the diffusion rate of calcium in the principal layer from decalcified C-S-H, respectively. The primary products of RHF after complete carbonation are calcite, amorphous silica gel, alumina gel, and silica-alumina gel, in which calcite gradually fills the pores and covers the surface of RBF particles during the carbonation process. The CRHF is employed as an SCM, and its pozzolanic activity can further enhance concrete's mechanical properties and durability by acting synergistically in the early and late stages of cement hydration. The positive findings from the current study have demonstrated the feasibility of efficiently producing carbonated RHF as a highly active SCM, paving the way for future industrial applications.

Keywords

Recycled concrete fines; red brick fines; carbonation kinetics; wet carbonation; pozzolanic activity

Session D3

Durability of CO_2 Enabled Concrete

CO_2-INDUCED CORROSION AND BONDING STRENGTH EVOLUTION OF THE STEEL-CONCRETE INTERFACE EXPOSED TO CO_2 UP TO 1000 kPa PARTIAL PRESSURE

Liwei Zhang[1,2], Quan Xue[3], Manguang Gan[1,2], Yan Wang[1], Yue Yin[1,2], Hanwen Wang[1,2]

[1]State Key Laboratory of Geomechanics and Geotechnical Engineering, Institute of Rock and Soil Mechanics, Chinese Academy of Sciences, Wuhan, Hubei 430071, China
[2]University of Chinese Academy of Sciences, Beijing 100049, China
[3]State Key Laboratory of Eco-hydraulics in Northwest Arid Region of China, Xi’an University of Technology, Xi’an, Shaanxi 710048, China
Email: lwzhang@whrsm.ac.cn

Abstract

CO_2 leakage from natural CO_2 reservoirs and geologic CO_2 storage formations may expose subsurface reinforced concrete structures to high pressure CO_2. In order to investigate the effect of CO_2 pressure and water on bonding strength evolution of the steel-concrete interface, this study investigated the carbonation process at the steel-concrete interface of the composite samples in dry CO_2 gas, pure water with dissolved CO_2, and 70% relative humidity (RH) environments with 100 kPa, 500 kPa and 1000 kPa CO_2 partial pressures. The results of 1000 kPa CO_2 showed that CO_2-induced corrosion at the steel-concrete interface was significant, which was very different from previously reported results with low CO_2 pressures. The formation of aragonite and $Fe(OH)_3$ at the interface had a negative effect on the bonding strength, while the formation of calcite at the interface had a positive effect on the bonding strength. Also, the reaction environment had a noticeable effect on Fe-bearing corrosion products formed at the interface. In summary, if the concentration and the pressure of CO_2 reach a certain level (i.e., 100% concentration and 1000 kPa partial pressure or higher) and abundant water is present, the CO_2-induced carbonation of concrete and corrosion of steel can eventually lead to large damage at the steel-concrete interface.

Keywords

High pressure CO_2; reinforced concrete; steel-concrete interface; bonding strength; carbonation

THE IMPROVEMENT OF CORROSION RESISTANCE OF WET-CAST CONCRETE SUBJECTED TO EARLY-AGE AMBIENT PRESSURE CARBONATION CURING

Xiangping Xian

Department of Architecture and Civil Engineering, City University of Hong Kong,
Hong Kong
Email: xiangping.xian@cityu.edu.hk

Abstract

This presentation will mainly discuss the possibility of developing early-age ambient pressure carbonation curing to lower the corrosion risk and improve the resistance by avoiding deep CO_2 penetration and chemically and physically densifying the concrete surface layer. In addition to superior CO_2 uptake and compressive strength, the effectiveness of ambient pressure carbonation on corrosion resistance of concrete was evaluated by two methods, an accelerated impressed current method and long-term ponding methods (ASTM G109). Due to improved physical properties, carbonation-cured concrete had a lowered current reading, a reduced rebar mass loss, a lessened total or free chloride content, and a decreased chloride diffusivity. Similarly, in the natural ponding test, a lower macrocell current and a lower total corrosion value were observed in the carbonation-cured concrete during the 33-month wet-dry cyclic exposure to a chloride solution. In conclusion, ambient pressure carbonation-cured concrete is corrosion resistant while still having the capacity to sequestrate carbon dioxide, which could contribute to sustainability and a circular carbon economy.

EFFECT OF CARBONATED FINE RECYCLED CONCRETE AGGREGATE ON DURABILITY

Lye, C.Q.[1], **and Pang, S.D.**[1]

[1] Department of Civil and Environmental Engineering, National University of Singapore
Email: ceepsd@nus.edu.sg

Abstract

The construction industry's pursuit of sustainability has significantly increased the interest in alternative construction materials, particularly fine recycled concrete aggregates (FRCA) as a viable replacement for natural sand in concrete and mortar. The carbonation of FRCA, due to the adhered cement paste, has been shown to enhance the properties of fine RCA, expanding its potential applications. This study explores a relatively untapped area of research concerning carbonated FRCA, focusing on their impact on mortar durability. This investigation is motivated by concerns about potential thaumasite sulfate attack, which may occur due to the presence of carbonates from the carbonation process of fine RCA, combined with moisture exposure. Additionally, the study examines resistance to acid attack and chloride ingress to provide a thorough understanding of the long-term performance of carbonated FRCA in various aggressive environments.

Periodic evaluations of the mortar mixes include monitoring changes in the strength and mass of mortar cubes, as well as the length of mortar bars. Furthermore, XRD analysis of samples taken from deteriorated specimen surfaces offers insights into their mineralogical compositions. This research provides valuable insights into the long-term performance implications of using carbonated FRCA. By contributing to the dialogue on sustainable construction practices, it highlights the need for further investigation in this promising field.

Keywords

Carbonated fine recycled aggregates; sulfate attack; acid attack; durability; sustainability

DURABILITY INVESTIGATION OF CONCRETE CONTAINING CARBON CAPTURE AND UTILIZATION (CCU) MATERIALS

Avadh K.[1], **Mori K.**[1], **Torichigai T.**[1], **Watanabe K.**[1] **and Sakai G.**[1]

[1]Kajima Corporation, Tokyo, Japan
Email: avadh@kajima.com

Abstract

As the threat of climate change continues to grow, it has become imperative for researchers worldwide to collaborate and develop technologies that enable the reduction and upcycling of waste materials. Concrete, being one of the most widely used man-made materials, is associated with a high degree of carbon emissions along its supply chain. Therefore, there is an evolving need to process and explore ways to upcycle waste materials generated during concrete application back into concrete. Given the complexity of the construction industry's supply chain, which involves various stakeholders such as cement manufacturers and suppliers, SCM producers, on-site constructors, pre-cast concrete manufacturers, construction equipment manufacturers, etc., there is a need for a platform that facilitates collaboration among them. The Green Innovation Project Fund serves as such a platform, supporting the development of low-carbon technology while addressing the concerns of each stakeholder and overcoming potential barriers that may arise during the implementation of this technology. As part of the technology being developed under the umbrella of this project, this study introduces a waste collection, processing, and upcycling system that has been successfully applied to the production of pre-cast concrete poles in Japan.

The factory employs centrifugal forming to manufacture high-strength concrete poles and piles. During the centrifugal forming process, cement particles and water seep out of the formwork called concrete sludge. The presence of cement particles in the concrete sludge, results in highly alkaline effluent that cannot be directly disposed of according to Japanese standards necessitating treatment. The concrete sludge containing cement particles is collected in an elution tank, it is then diluted to allow hydration to progress and for Calcium ions to leach into the water while continuously mixing, forming a cement slurry. Subsequently, the cement slurry undergoes filter pressing to separate it into a highly alkaline liquid rich in Calcium ions and sludge cake consisting of solid particles. The liquid part is wet carbonated using boiler exhaust gas generated in the factory, resulting in the production of highly stable synthetic calcium carbonate (SCC). On the other hand, the sludge cake is air-dried and crushed to produce two categories of CSP (i) Coarse CSP(0~2mm) and (ii) Fine CSP (0~100μm). Both are further subjected to carbonation at 200℃ and 100% CO_2 concentration for 8 hours, leading to the formation of carbonated sludge powder (C-CSP). Since each of the above-mentioned materials has been produced by utilizing waste CO_2 gas which otherwise would have harmed the environment, the material will be cumulatively called Carbon Capture and Utilization (CCU) materials. The current recycling and carbonation system can produce approximately 60 tons/year of SCC, along with a total of 400 tons per year of CSP and C-CSP. The production amount is expected to increase by many folds in the coming years.

The objective of this study is twofold. Firstly, it involves the characterization of each of the CCU materials obtained from the concrete sludge treatment process. Secondly, it aims to investigate the performance of concrete when 50 kg/m^3 of sand is replaced with these CCU

materials. The materials are characterized using the X-ray diffraction (XRD) Rietveld refinement method and thermo-gravimetric analysis (TG-DTA). Additionally, physical properties such as density and water content were measured. Subsequently, the effect of adding CCU materials on the fresh properties and compressive strength of the concrete is evaluated. To understand the impact on durability, further investigations were conducted on the freeze and thaw resistance, as well as the measurement of drying shrinkage of the concrete specimens.

The XRD and TG-DTA analyses revealed that the concrete sludge powder (CSP) mainly consisted of cement hydration products, including calcium hydroxide and ettringite, along with a small amount of β-C_2S. Additionally, CSP contained a significant amount of calcium carbonate, which was formed during the natural air drying process. Upon carbonation of CSP to produce carbonated concrete sludge powder (C-CSP), most of the cement hydration products are transformed into calcium carbonate. Synthetic calcium carbonate (SCC) primarily consists of calcium carbonate, with certain amounts of calcium hydroxide and residual water content. The investigation of physical properties demonstrated that CSP had a high water content and exhibited a lower density. However, upon carbonation to produce C-CSP, the density increased due to the formation of calcium carbonate and a reduction in water content.

In addition to investigating the carbonation process, it is crucial to examine the effects of incorporating these CCU materials on concrete properties. The results of the study indicated that the inclusion of CCU materials led to an increase in compressive strength. However, to achieve the desired slump, a higher dosage of high-range water reducer was required when incorporating the CCU materials. Furthermore, from the freeze and thaw experiments, it was observed that concrete containing coarse CSP exhibited a significant reduction in dynamic modulus of elasticity and an increase in mass loss. However, concrete containing the remaining types of CCU materials showed a better or comparable performance compared to control concrete. In all cases, a greater degree of surface scaling was observed. Regarding drying shrinkage, an increase in shrinkage was observed in all cases compared to the control concrete.

In conclusion, the investigation of concrete properties demonstrated that the incorporation of carbon capture and utilization (CCU) materials had a significant impact on the properties of concrete. These findings emphasize the importance of thoughtful selection and treatment of CCU materials considering that the production and application of these materials in concrete are expected to increase in the future, it is crucial to carefully evaluate the exposure conditions at the actual application site before utilizing concrete containing these CCU materials.

Keywords

Concrete sludge powder; waste upcycling; carbonation; durability; re-use

Acknowledgment

This paper is based on results obtained from a project, JPNP16002, commissioned by the New Energy and Industrial Technology Development Organization (NEDO).

DAMAGE EVOLUTION AND FRACTURE MECHANISM OF CONCRETE WITH FULL CARBONATED RECYCLED AGGREGATES

Yuxiang Tang[1,2], Jianzhuang Xiao[2], and Junhui Zhang[1]

[1]School of Traffic and Transportation Engineering, Changsha University of Science & Technology, Changsha, 410114, China
[2]Department of Structural Engineering, College of Civil Engineering, Tongji University, Shanghai, 200092, China
Email: yxtang@csust.edu.cn

Abstract

Carbonation treatment of recycled concrete aggregates enables fast carbon sequestration while improving engineering properties of prepared concrete. This study reports a systematic experimental study on the fracture behavior of concrete with carbonated recycled coarse and fine aggregates (CRCA and CRFA), in terms of fracture parameters and fracture process zone (FPZ). By relating the macroscopic fracture responses to microstructural features. the influence and enhancement mechanism of different particle sizes of recycled aggregate and its carbonation modification on the damage evolution and fracture behavior of concrete. The safety and low carbon synergies of fully utilizing carbonated recycled aggregates to concrete were demonstrated.

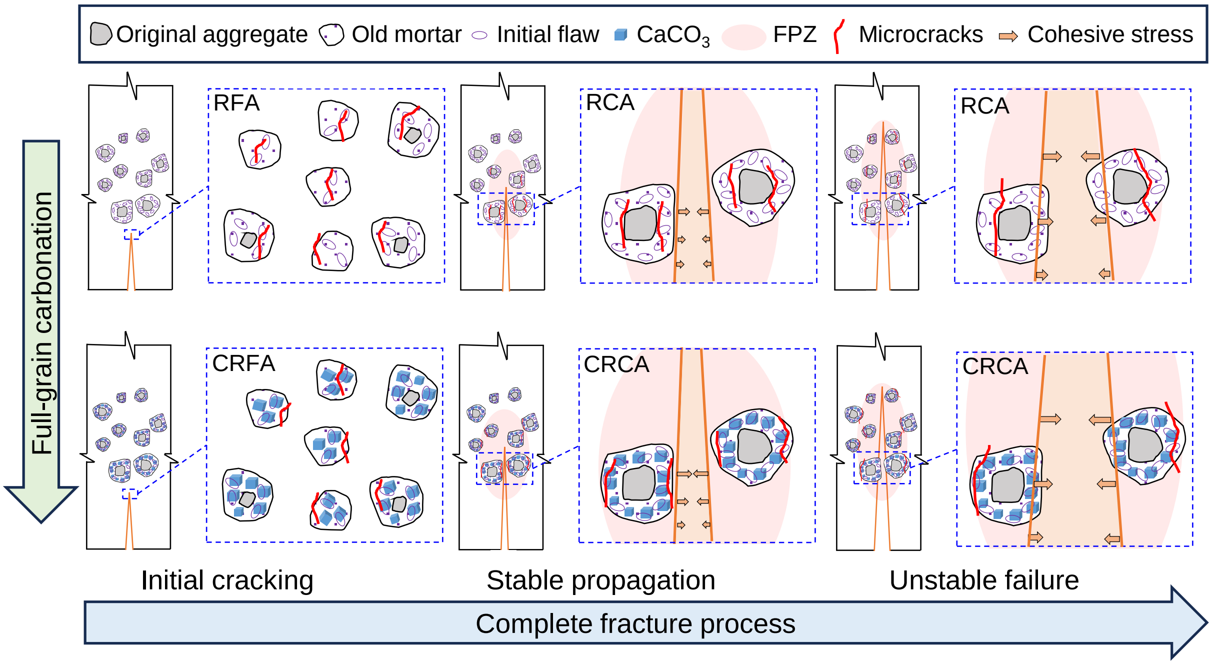


Figure 1. Fracture mechanism of concrete with full carbonated recycled aggregates.

Keywords
Recycled concrete; carbonation treatment; damage evolution; fracture behavior

STUDY ON DYNAMIC MECHANICAL BEHAVIOR AND DAMAGE EVOLUTION MECHANISM OF FIBER REINFORCED CEMENTED TAILINGS BACKFILL

Shizhuo Zou[1], **Zhou Yu**[1]

[1]Key Laboratory of Ministry for Efficient Mining and Safety of Metal Mines, University of Science and Technology Beijing, Beijing 100083, China
Email: zousz_ustb@foxmail.com

Abstract

The backfilling method stands as a widely employed extraction technique in contemporary mining practices, with the performance of cemented tailings backfill (CTB) being pivotal in determining the overall quality of backfilling. Nevertheless, traditional CTB exhibits suboptimal mechanical properties, particularly crack resistance, under complex stress environments. Presently, one of the extensively explored backfill types is the fiber-reinforced cemented tailings backfill (FRCTB), with a particular emphasis on those reinforced with polypropylene fibers. In order to scrutinize the mechanical characteristics of FRCTB under intricate stress states, this study, employing a triaxial Hopkinson pressure bar experimental apparatus, investigates the dynamic mechanical behaviors, fracture damage patterns, and energy dissipation features of FRCTB under five different confining pressures (0, 1, 2, 3, 4 MPa) and various strain rates.Key findings include:

(1) Under dynamic loading, FRCTB exhibits a pronounced strain rate strengthening effect along with a notable confining pressure strengthening effect. The presence of confining pressure significantly alters the stress-strain curve of FRCTB. The peak stress and dynamic increase factor (*DIF*) of FRCTB linearly increase with the augmentation of both confining pressure and strain rate. The peak strain linearly increases with the strain rate, with confining pressure exerting minimal influence on the peak strain. Confining pressure substantially enhances the elastic modulus of FRCTB, while the impact of strain rate is comparatively marginal.

(2) In the absence of confining pressure, FRCTB specimens, with increasing strain rates, exhibit an outward-expanding conical failure shape. The crack volume and surface area increase in a stepwise fashion, and the hollow cylindrical polypropylene fibers undergo flattened failure. At this juncture, the polypropylene fibers endure a limit strain rate ranging from 206.5 s^{-1} to 232.3 s^{-1}. As confining pressure gradually increases, the outward expansion tendency is progressively restrained until no discernible internal damage occurs. At this point, the polypropylene fibers manifest phenomena such as splitting, bending, and extraction. Under low (no) confining pressure conditions, the fractal dimension and porosity of FRCTB increase with the rising strain rate. In high confining pressure conditions, the fractal dimension and porosity of FRCTB are relatively similar across different strain rates.

(3) At lower confining pressures, the strain rate strengthening effect is evident in both fracture morphology and energy dissipation but diminishes as confining pressure increases. Dissipated energy density exhibits an increasing trend with the rise in confining pressure, while the energy dissipation rate shows a quadratic function decrease with increasing strain rate. The stress-strain curves of FRCTB under no confining pressure and with confining pressure can be categorized into four and five segments, respectively: elastic growth, plastic yield, post-peak energy

accumulation, and post-peak failure for the former, and elastic growth, plastic damage incubation, plastic damage development, plastic damage accumulation, and post-peak failure for the latter. Under low confining pressure conditions, the fractal dimension linearly increases with the growth of dissipated energy. This trend gradually transforms into a cubic function change as confining pressure increases. There exists a notable similarity between fractal dimension and energy dissipation rate, as well as between strain rate and confining pressure.

Keywords

Fiber reinforced cemented tailings backfill; triaxial Hopkinson pressure bar device; mechanical property; energy dissipation; damage mechanism

PREDICTION AND OPTIMIZATION DESIGN OF CONCRETE CHLORIDE DIFFUSION COEFFICIENT BASED ON MACHINE LEARNING APPROACH

Yiwei Zhang[1], Pan Feng[1]

[1]School of Materials Science and Engineering, Southeast University, Jiangsu, 211100, China

Email: pan.feng@seu.edu.cn

Abstract

1. Introduction

Concrete is the most widely used construction material among the world, but concrete industry is also one of the major sources of global carbon dioxide emissions. Therefore, concrete should meet the durability performance, environment requirement, and economic cost for engineering applications. However, there is a conflict between these three main characters, and it's important to discover a solution to achieve the balance between these requirements. Chloride diffusion coefficient reflects the chloride corrosion resistance of concrete. Therefore, it's important to estimate the chloride diffusion coefficient in order to increase the durability and service life of buildings. Machine learning has been successfully used in civil engineering performance prediction and the optimization design of concrete through its ability to extract patterns from complex data, and provides new approaches for cementitious material study. This work aims to establish a standard concrete chloride diffusion coefficient database, train and build machine learning model to predict durability performance, and finally achieve optimization design of concrete mix proportion.

2. Methodology

To achieve the research objectives, there are three procedures:

1) Establish a high-dimensional multi-source standard concrete chloride diffusion coefficient database. Collect chloride diffusion data under engineering environments. The original database underwent missing value handling and outlier detection to ensure data accuracy. Standardization was applied to adjust durability parameters from different sources to the same scale, enhancing model consistency. Correlation analysis was conducted to explore potential relationships between parameters, improving model training accuracy and efficiency.

2) Based on the established standard database, various machine learning models, including MLP, XGBoost, RF, and KRR, were constructed and trained. The models focused on concrete durability as the primary criterion, with cross-validation and hyperparameter optimization ensuring model robustness and accuracy. After the training process, partial dependence plots and SHAP analysis were employed to thoroughly analyze the contribution of key features, identifying important factors influencing chloride diffusion coefficient and providing data-driven guidance.

3) Based on the optimal machine learning model, a multi-objective optimization design for concrete durability was carried out. Using the NSGA method, multiple factors including chloride diffusion coefficient, carbon dioxide emissions, and economic cost were comprehensively considered to select the Pareto optimal solution.

3. Result

1) Build a standard durability database consisting 846 data sets. The environment factors are strongly connected with chloride diffusion coefficient according to the correlation analyses, as shown in figure 1(a). And the data distribution is presented in figure 1(b).

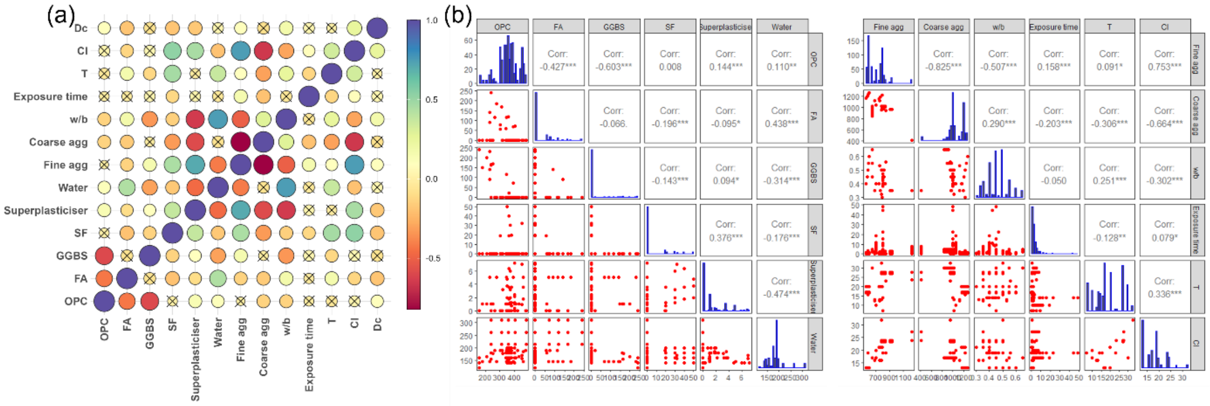


Figure 1. (a) concrete durability database correlation analyses heat map, (b) database structure matrix

2) The model performance is shown in figure 2(a), and MLP model presents the highest R2(0.945) and the lowest MAE(0.015). The results of SHAP analyses and partial dependence plots are shown in figure 2(b)(c), which indicates that environment factors also highly corresponded with chloride diffusion coefficient.

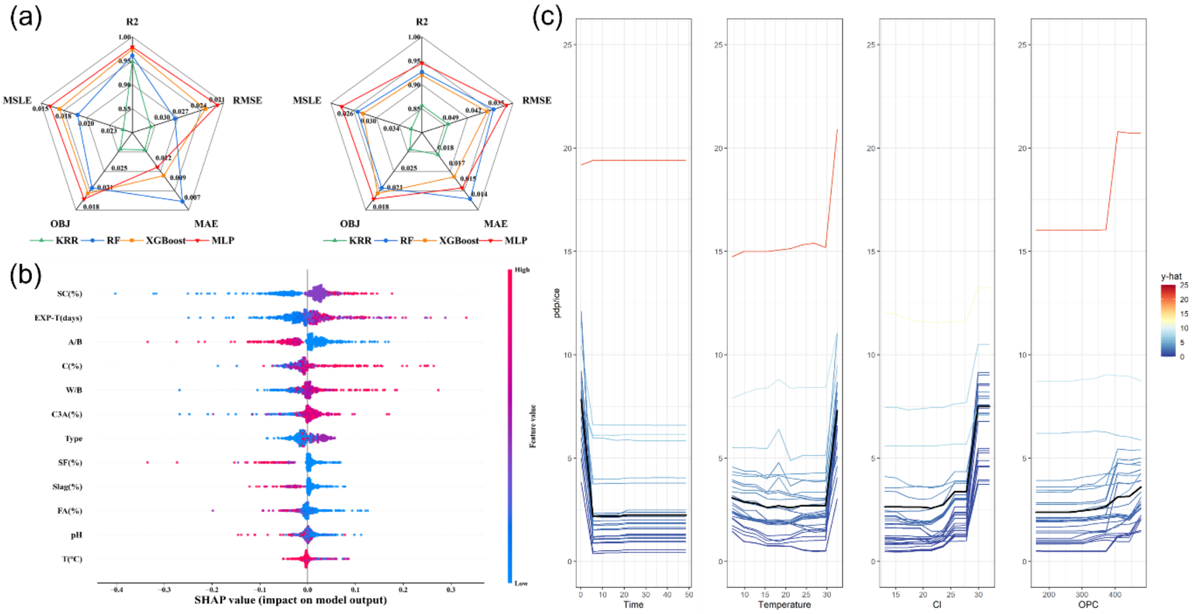


Figure 2. (a) model performance comparison, (b) SHAP analysis, (c) partial dependence plots

3) Use best machine learning model and NSGA strategy to optimize concrete mix proportion, increase the chloride diffusion resistance over 20% and reduce the emission of carbon dioxide over 10%.

Keywords

Durability; machine learning; concrete mix proportion optimization; carbon dioxide

IMPREGNATE CARBONATION: CO_2-GUIDED IN SITU GROWTH OF ROBUST SUPERHYDROPHOBIC STRUCTURES ON CONCRETE SURFACES

Long Jiang[1], Zihan Ma[1], Zhenjiang Gu[1], Peiliang Shen[1], Yong Tao[1], Weihua Li[2], Chi-sun Poon[1]

[1]Department of Civil and Environmental Engineering& Research Centre for Resources Engineering towards Carbon Neutrality, The Hong Kong Polytechnic University, Hong Kong 999077
[2]North China University of Water Resources and Electric Power, Zhengzhou 450046
Email: peiliang.shen@polyu.edu.hk; cecspoon@polyu.edu.hk

Abstract

Superhydrophobic surfaces applying on concrete could greatly improve the durability of concrete by preventing the damage from water. However, traditional design of superhydrophobic concrete surfaces by external coating encounters to problems of flaking and poor surface robustness, while that by adding hydrophobic agents or particles faces the challenges of strength damage of concrete. Drawing inspiration from the carbonation phenomenon of concrete, here a new design of in-situ growing superhydrophobic structures on concrete is proposed: The concrete sample is impregnated into Mg^{2+}-containing silane-water system with continuous CO_2 injection. The contact angle of the concrete surface achieves 171.9° without obvious strength decrease after 120 minutes, which are mainly attributed to the formation of CaxMg1-xCO3 crystals with micro-nano-structures and the reduction of carbonates surface energy by silane. This superhydrophobic concrete structure can be divided into a superhydrophobic-hydrophobic-hydrophilic three layers structure, providing the stable water-proof protection under mechanical fatigue, capillary water absorption, UV aging, sulfate attack, and impurity water impact tests due to the in-situ growing robust superhydrophobic structures. Furthermore, it captures 29.80 g/m^2 CO_2 during the reaction process, providing new insights for the design and preparation of eco-friendly superhydrophobic concrete.

Keywords
Superhydrophobic concrete; in-situ growth; micro-rough structures; CO_2 sequestration; morphology segmentation

Session D4

Life Cycle Analyses and Quantification of CO_2 Uptake

CARBON SEQUESTRATION THROUGH CONCRETE CARBONATION: ENHANCING LOW-CARBON AND HIGH-PERFORMANCE CONCRETE FOR A SUSTAINABLE BUILT ENVIRONMENT

Sze-dai Pang

Department of Civil and Environmental Engineering, National University of Singapore, Singapore
Email: ceepsd@nus.edu.sg

Abstract

Carbonation of concrete has gained traction as a viable method for carbon sequestration, offering significant potential to decarbonize the built environment. This study investigates the carbonation process in both low-carbon concrete (LCC) and high-performance concrete (HPC), with a focus on early-age carbonation and its impact on carbon uptake and material performance. Specifically, pastes and mortars were prepared using cement blends with supplementary cementitious materials (SCMs) such as fly ash, silica fume, calcined clay, and ground granulated blast furnace slag (GGBS), replacing 20% by weight of the cement. Carbon curing was performed at two stages: fresh state (0–24 h) and hardened state (24–48 h) in a chamber with 99% CO2 at 1.5 bars for 24 hours.

Thermo-gravimetric analysis (TGA) revealed that hardened-state carbonation stored 1.5 times more CO2 than fresh-state carbonation, with calcium hydroxide (CH) carbonation proving to be more effective than AFt/C-S-H phase carbonation. Microstructural analysis using mercury intrusion porosimetry (MIP) showed densification in hardened samples, with up to 18% reduction in pore volume. Hardened-state curing did not significantly alter early-age strength, while fresh-state curing demonstrated a strength improvement of 7–37%.

Furthermore, the study explored carbon capture and storage (CCS) in ultra-high performance concrete (UHPC) by replacing cement with GGBS (30%, 50%, 70% by weight) and capturing CO2 during fresh-state curing. While carbonation led to a slight decrease in compressive strength, it significantly improved the degree of carbonation (DOC), with the highest CO2 uptake (80 kg/m^3) observed at 30% GGBS replacement.

This research demonstrates the feasibility of integrating carbonation into low-carbon and high-performance concrete production as a carbon capture and utilization (CCU) strategy. The findings suggest a path toward developing eco-friendly concretes that support decarbonization efforts in the construction industry, aligning with global sustainability goals.

CARBON DIOXIDE EMISSION REDUCTION BENEFITS AND COST SAVINGS CALCULATION FOR THE UTILIZATION OF RECYCLED AGGREGATE FROM HIGHWAY DEMOLITION

Yang, Ming[1,2], Chen, Chulong[1], Zhuang, Jiafeng[1], and Zhang, Jingbo[1]

[1]CCCC Second Highway Consults Consultant Co., Ltd
[2]China University of Geosciences (Wuhan)
Email: webd@vip.qq.com

Abstract

To assess CO_2 (carbon dioxide) emission reduction and economic benefits of bridge demolished RA(recycled aggregate) in highway engineering, a carbon reduction calcuation method is proposed and applied in the Guangzhou - Shenzhen reconstruction section of Beijing - Hong Kong and Macau Expressway. The results show that the CO_2 emission reduction benefits of RA is 4.26~22.53 kg/m^3, and the unit cost savings reach 80.84~123.27 CNY/m^3, compared with natural materials. The optimal application scenario of RA for carbon reduction and economic comprehensive benefits is special roadbed treatment, followed by cement stabilized crushed stone base and cement pavement, and finally the cast-in-place integrated layer of the bridge deck and graded crushed stone sub base. Through process optimization, 20 0000 m^3 demolished RA can be utilized with reducing carbon emissions by 2.07 million kilogram, and saving costs by 25.18 million CNY. The average carbon reduction benefit of RA is about 10.34 kg, plus saving cost by 125.78 CNY per cubic meter. It is concluded that the utilization of RA in highway can contribute to achieving a low-carbon bridge demolition and reuse.

Keywords

Highway engineering; demolition; recycled aggregate; carbon reduction; cost savings

Acknowledgement

Supported by Guangdong S&T programme 2021B1111610002 & CCCC major research and development project 2022-ZJKJ-14.

THE NEGATIVE EMISSION POTENTIAL OF INDUSTRIAL SOLID WASTES: CARBONATION EFFICIENCY EVALUATION AND CONSTRAINTS IN CO_2 REDUCTION

Yikai, L.[1], **Alejandro, F. M.**[1], **Jesús L. C.**[2], **Bin, M.**[3], **Gilberto, A.**[4], **Winnefeld, F.**[5]

[1] University of Grenoble Alpes, University of Savoie Mont Blanc, CNRS, IRD, IFSTTAR, ISTerre, 38000 Grenoble, France
[2] ArcelorMittal, Global Research and Development, 33402 Avilés, Spain
[3] Laboratory for Waste Management, Paul Scherrer Institut (PSI), Forschungsstrasse 111, 5232, Villigen, Switzerland
[4] Department of Geosciences and CIRCe Centre, University of Padua, Via G. Gradenigo 6, 35129, Padua, Italy
[5] Empa, Swiss Federal Laboratories for Materials Science and Technology, Dübendorf 8600, Switzerland
Email: yikai1995L@gmail.com

Abstract

Mineralization of industrial solid waste (converting atmospheric carbon dioxide into carbonate minerals such as calcite ($CaCO_3$) or magnesite ($MgCO_3$)) is a top priority in achieving our pursuit of the Paris Agreement goals [1]. This is one of the earliest carbon capture, utilization, and storage (CCUS) technologies to reach commercial scale [2-3]. It is attributable to its relatively low cost and energy consumption, which makes long-term sequestration of CO_2 applicable. Therefore, it is critical to find infrastructure capable of long-term sequestration of mineralized CO_2 and to identify suitable recycling applications to reuse carbonate products. The integration of waste-derived products has the potential to partially or entirely replace raw materials used in industrial production [4].

This work aims to determine the carbonation efficiency of industrial solid wastes from different sectors (e.g., steel slag and iron tailings from the steel-making process, phosphate tailings and phosphate slag from fertilizer production, coal fly ash from electricity generation, demolition wastes from construction sectors, etc.), as well as the quantification of CO_2 absorbed. Different carbonation experiments were applied in water-rich, semi-dry, and atmospheric air conditions. Further, we examined the potential of industrial solid waste to remove carbon dioxide from the atmosphere by projecting the amount of industrial solid waste generated in conjunction with experimentally observed reductions in CO_2. The results show that a large proportion of the future negative emission requirements may be met through weathering or carbonation of these materials. However, it is worth noting that there are uncertainties in this negative emission technology, especially the presence of harmful trace elements, which may constrain the rate and extent of its scale-up.

Keywords

Carbon capture utilization and storage; solid waste management; carbon mineralization; CO_2 emission reduction

Acknowledgment

This project has received funding from the European Union’s Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie Postdoctoral Fellowship grant agreement No 101154963.

References

1. Agreement, P. (2015). Adoption of the Paris Agreement. In United Nations Climate Change Secretariat (UNFCCC). FCCC/CP/2015/L. 9/Rev. 1.
2. Snæbjörnsdóttir, S. Ó., Sigfússon, B., Marieni, C., Goldberg, D., Gislason, S. R., & Oelkers, E. H. (2020). Carbon dioxide storage through mineral carbonation. Nature Reviews Earth & Environment, 1(2), 90-102.
3. Matter, J. M., Stute, M., Snæbjörnsdottir, S. Ó., Oelkers, E. H., Gislason, S. R., Aradottir, E. S., ... & Broecker, W. S. (2016). Rapid carbon mineralization for permanent disposal of anthropogenic carbon dioxide emissions. Science, 352(6291), 1312-1314.
4. Liu, W., Teng, L., Rohani, S., Qin, Z., Zhao, B., Xu, C. C., ... & Liang, B. (2021). CO2 mineral carbonation using industrial solid wastes: A review of recent developments. Chemical Engineering Journal, 416, 129093.

CARBON NEUTRAL POTENTIAL OF THE CONCRETE RECYCLING PROCESS THROUGH CO_2 SEQUESTRATION FROM FLUE GASES

Besklubova, S.[1], **Kravchenko, E.**[2], **Zhong, R.Y.**[1], **and Brilakis I.**[3]

[1] University of Hong Kong
[2] Southern Federal University, Russia
[3]University of Cambridge,
Email: ekakra@sfedu.ru

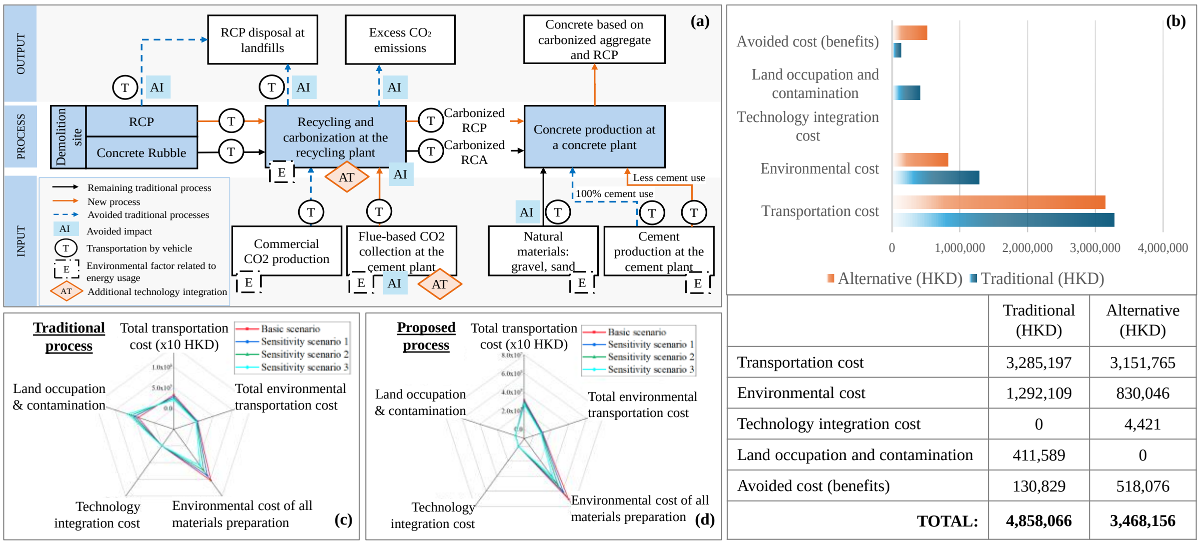
Abstract

The construction industry faces significant environmental challenges, largely due to the substantial volume of demolished concrete waste it generates, and the significant CO_2 emissions produced during cement manufacturing. Efforts to recycle concrete aggregates (RCA) produce a significant amount of recycled cement powder (RCP), estimated to comprise 30-70% of the total waste concrete mass (Oh et al. 2021). However, the effective integration of RCP into the recycling process remains limited, leading to landfill disposal and associated environmental risks (Besklubova et al. 2023). A potential solution involves merging carbon-neutral technology with large-scale concrete production, using RCP to replace cement and fine aggregates. Additionally, carbonization enhances the properties of both RCA and RCP while capturing significant amounts of CO_2. Although material scientists have developed this technology, its financial viability requires further evaluation. Addressing this gap is an important step in facilitating the widespread implementation of technology. Therefore, this study aims to evaluate the integration of carbon-neutral technology into recycling waste concrete for concrete mixes, focusing on active carbonation using flue CO_2 from cement plants. Considering cement clinker production contributes about 8% to global CO_2 emissions (Andrew 2019), carbonating concrete waste not only supports carbon neutrality but could also improve the strength of materials.

A comprehensive model that combines Cost-Benefit Analysis and Activity-Based Costing (ABC)—referred to as the ABC-based value assessment model—has been developed to conduct a detailed cost analysis comparing current concrete recycling practices with a proposed approach that integrates carbon-neutral technology. Figure 1(a) illustrates both the traditional and alternative scenarios to provide a clear visual representation of their difference and the specific parameters involved. The model quantifies all relevant design variables, calculating the net difference between costs and benefits. Costs include transportation of RCP, RCA, CO_2, and natural materials; environmental impacts from commercial and flue CO_2 generation, carbonization, natural material processing, cement production, and post-carbonization CO_2 release; and technology integration for flue CO_2 collection and carbonization. Costs related to land occupation and contamination were also considered. On the benefits side, the model captures savings from reduced CO_2 emissions by utilizing flue CO_2, decreased use of natural materials, and CO_2 absorption during carbonization (Kravchenko and Besklubova 2024).

Utilizing data from the Hong Kong construction industry, a cost analysis on the demolition of prefabricated facades in a Tuen Mun high-rise employed the ABC-based value assessment model, revealing a total of 5,144 tonnes of concrete materials (Kang et al. 2022). Of this, 30% were RCP and 70% consisted of RCA, highlighting opportunities for material recycling in such projects. The findings show the alternative concrete recycling method is about 29% more cost-effective than the conventional method (Figure 1b), with similar transportation costs. The alternative method also reduces environmental costs by over 51%, while technology integration

costs remain minor. The study performs sensitivity analyses for three scenarios, demonstrating RCP generation rates of 40%, 50%, and 60% during building demolition. In traditional concrete recycling (Figure 1c), land occupation and contamination costs increase by 21.4-64.3%, yet the environmental transportation costs decrease by 12.5-37.4%, resulting in a net cost reduction of 10-30%. Conversely, in alternative recycling methods (Figure 1d), the environmental costs of material preparation escalate by 13.6-40.7%, but the overall costs drop by 6.7-20.2%, with negligible changes in technology integration costs.



	Traditional (HKD)	Alternative (HKD)
Transportation cost	3,285,197	3,151,765
Environmental cost	1,292,109	830,046
Technology integration cost	0	4,421
Land occupation and contamination	411,589	0
Avoided cost (benefits)	130,829	518,076
TOTAL:	**4,858,066**	**3,468,156**

Figure 1. Study results: (a) Traditional and alternative concrete waste recycling processes; (b) Comparative analysis of traditional and alternative scenarios; Sensitivity analysis for the amounts of RCA and RCP in (c) traditional recycling and (d) alternative recycling.

Keywords

Recycled cement powder; construction waste management; concrete recycling; flue gas; CO_2 sequestration

Acknowledgment

This work was supported by the UGC Research Impact Fund (R7036-22), UGC Theme-based Research Scheme (T32-707-22-N), UGC GRF (17203620); Guangdong Special Support Talent Program—Innovation and Entrepreneurship Leading Team under Grant 2019BT02S593, and HKU Teaching Development Grant (TDG) Award (Project No. 951).

References

Andrew, R. M. (2019). Global CO 2 emissions from cement production, 1928–2018. *Earth System Science Data*, *11*(4), 1675-1710.

Besklubova, S., Kravchenko, E., Tan, B. Q., & Zhong, R. Y. (2023). A feasibility analysis of waste concrete powder recycling market establishment: Hong Kong case. *Environmental Impact Assessment Review*, *103*, 107225.

Kang, K., Besklubova, S., Dai, Y., & Zhong, R. Y. (2022). Building demolition waste management through smart BIM: A case study in Hong Kong. *Waste Management*, *143*, 69-83.

Kravchenko, E., & Besklubova, S. (2024). Value stream assessment of the sustainable concrete recycling process with sequestration of CO2 from flue gases. *Sustainable Production and Consumption*.

Oh, D., Noguchi, T., Kitagaki, R., & Choi, H. (2021). Proposal of demolished concrete recycling system based on performance evaluation of inorganic building materials manufactured from waste concrete powder. *Renewable and Sustainable Energy Reviews*, *135*, Article 110147.

A POTENTIAL CARBON SINK: RECYCLED AGGREGATE STOCKPILES

Yunlu Hou[1,2,3], **Li Qi**[2,3], **Pierre-Yves Mahieux**[1], **Philippe Turcry**[1], **Jérôme Lux**[1], **and Abdelkarim Aït-Mokhtar**[1]

[1] LaSIE, Laboratoire des Sciences de l'Ingénieur pour l'Environnement, UMR 7356 CNRS,
La Rochelle Université, La Rochelle
[2] State Key Laboratory of Geomechanics and Geotechnical Engineering, Institute of Rock and
Soil Mechanics, Chinese Academy of Sciences, Wuhan 430071, China
[3] University of Chinese Academy of Sciences, Beijing 100049, China
E-mail (corresponding author): luuhou6@outlook.com

Abstract

While concrete is a building material that consumes natural resources and emits large amounts of CO_2, it is also a material capable of sequestering CO_2 through atmospheric carbonation during the lifetime of the structures for which it was manufactured, and after demolition and processing in the form of recycled aggregates (RA). The aim of this case study is to estimate the amount of CO_2 (m_{CO_2} in kg; Eq.1 and Eq.2) that a stockpile of 2000 m^3 of RA can sequester between two crushing campaigns 4 months apart (Figure 1). The approach is based on two laboratory studies: firstly, the determination of the CO_2 trapping potential (Q_{CO_2} in kg/m^3) at the RA grain scale, and secondly, the determination of the CO_2 diffusivity (D_{CO_2} in m^2/s) in the granular medium formed by the stockpile of materials studied.

$$m_{CO_2} = S \times X_c \times Q_{CO_2} \qquad \text{Eq. 1}$$

$$X_c = \sqrt{\frac{2\, D_{CO_2}[CO_2]}{Q_{CO_2}} \times t} \qquad \text{Eq. 2}$$

Where, S is the surface (m^2) of the outer envelope of the recycled aggregates stockpile determined by photogrammetry, X_C is the carbonatation front (m) deduce by calculation from the CO_2 diffusivity test and the first Fick's law.

Figure 1: Manufacture (a) and storage (b) of RA on the Valosphère recycling platform in France.

According to the experiment results in the laboratory by the developed natural carbonation cell and oxygen diffusion cell, we found that for a recycled aggregate stockpile with 2000 m^3 in 4 months could be captured more than 630 kg CO_2, i.e., 0.2 kg CO_2/t recycled aggregates. The results provided that the recycled aggregates stockpile sequestered under atmospheric conditions the actual capacity of CO_2 is very dependent on the manufacturing process, storage on the platform and weather conditions. It is recommended that accelerated carbonation methods be considered to accelerate the recycled aggregate stockpile carbonation to achieve rapid and significant CO_2 sequestration.

Keywords

Recycled aggregates; CO_2 sequestration; natural carbonation; carbon neutrality; carbon sink

CARBONATED RECYCLED CONCRETE POWDER AS AN ALKALI-ACTIVATED BINDER: PERFORMANCE OPTIMIZATION AND ENVIRONMENTAL EVALUATION

Wenjing Zhao[1], **Zhenhua Duan**[1], **Jiaqian Ning**[1], **Jianzhuang Xiao**[1]

[1]Department of Structural Engineering, College of Civil Engineering, Tongji University, Shanghai 200092, China
Email: zhduan@tongji.edu.cn

Abstract

Reducing clinker content and carbon capture are important measures to promote decarbonization in the cement industry. In this study, carbonated recycled concrete powder (CRCP), an abundant in-situ available material, was developed as a precursor to produce a new alkali-activated binder. The effects of the CRCP substitution, alkali content (AC), and silicate modulus (SM) on the compressive strength of AABs were firstly investigated. The response surface method was further used to optimize the above parameters. The results show that when AC and SM are 14.28% and 0.92, the AAB containing 50 wt.% CRCP can gain the highest strength (46.8 MPa at 28 days). Moreover, chemical analyses reveal that AABs are mainly composed of polycrystalline $CaCO_3$ (such as calcite and vaterite), C-(A)-S-H and silica-alumina gel, both Si-Al gels and polycrystalline $CaCO_3$ are the primary reactants in CRCP. Lastly, the life cycle assessment indicates the global warming potential (GWP) of CRCP is -0.16 kg·CO_2·eq/kg. Comparing with the equal strength Portland cement, the optimally proportioned mortar with containing 50% CRCP only have a 54.4% of GWP, confirming the environmental advantages of the CRCP-contained AABs.

Keywords

Carbonated recycled concrete powder; alkali-activated binder; response surface method; performance optimization; environmental assessment

ESTIMATION OF CO_2 CAPTURE THROUGH CONCRETE WEATHERING CARBONATION IN MAINLAND CHINA IN PAST 30 YEARS

Xin Shao[1] and Tung-Chai Ling[1]

[1] College of Civil Engineering, Hunan University, 410082 Changsha, China
Email: tcling@hnu.edu.cn; tcling611@yahoo.com

Abstract

According to a study published in Nature Geoscience, a cumulative amount of 4.5 gigatons of carbon (GtC) can be sequestrated via concrete weathering carbonation from 1930 to 2013. Such a huge amount can offset 43% of the CO_2 emissions associated to cement production during the same period, excluding emissions from fossil fuel use in the production process. As concrete is exposed to significant climatic variations across different regions, its carbon absorption patterns vary significantly. Environmental factors such as temperature, relative humidity and CO_2 concentration significantly impact concrete carbonation, and these factors have not adequately accounted in calculating the global carbon absorption of cement concrete. To further refine the calculation, this study collects the latest survey datasets and establish a model to analyze the absorption of atmospheric CO_2 by various types of concrete structures under different climatic conditions. The calculation of concrete CO_2 sequestration in mainland China can cover the influence of different climate sessions on concrete weathering carbonation and resulting carbon capture.

Our results indicate that a significant portion of CO_2 emissions from the cement manufacturing process in mainland China has been reabsorbed through the carbonation of concrete, and as illustrated in fig. 1(a), a substantial increase occurred in absorption, especially from the beginning of the 21st century. As seen, the annual CO_2 uptake was 0.03 GtC per annual in 1992 rose to 1.22 GtC in 2021. This can be understood by the fact of rapid economic growth in mainland China and the use of concrete for infrastructures and building construction. The geographical distribution of CO_2 uptake reveals a distinct pattern (fig. 1(b)), with higher CO_2 uptake occur in the eastern regions compared to the western regions. This spatial trend aligns with the developmental landscape of mainland China, where southeastern coastal cities have experienced a rapid growth of economic. Additionally, the different climate distribution across the mainland China, particularly the relatively high average temperatures along the southeast coast could promote the carbonation process of concrete and higher CO_2 uptake across this region. We further analyze the quarterly climate impact on CO_2 uptake (fig. 1(c)), and found that there was an obvious high absorption rate observed in the 2nd and 3rd quarters compared to the 1st and 4th quarters. This seasonal trend aligns with climatic characteristics in China, where high summer temperatures in 2nd and 3rd quarters which can accelerate the carbonation rate of concrete. In addition, the 1st quarter coincides with the Chinese New Year holiday, resulting in substantially lower concrete production compared to other quarters.

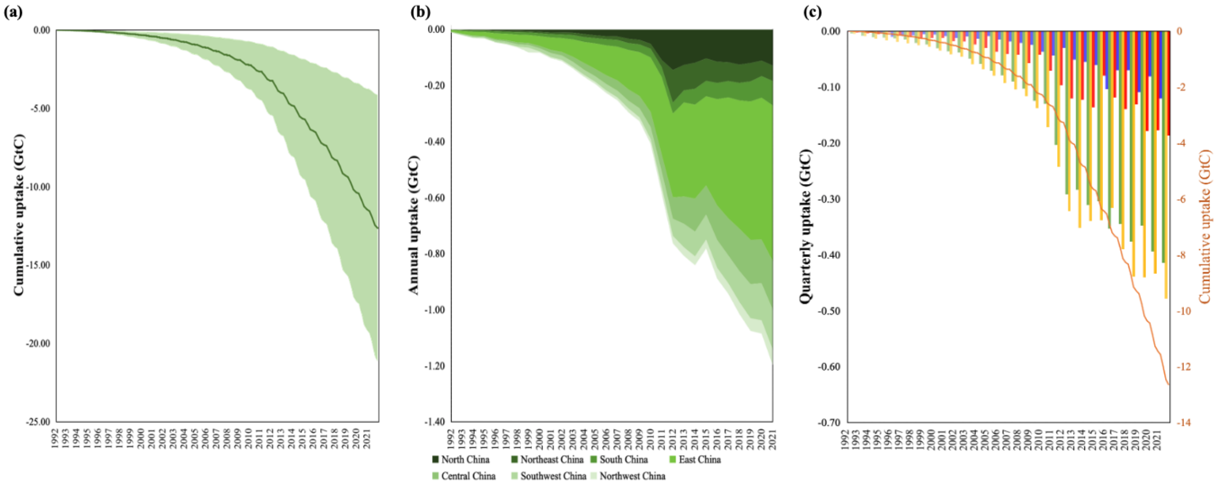


Fig.1. Carbon sequestration by concrete weathering carbonation from 1992 to 2021. (a) National cumulative uptake national (b) Annual uptake by area and (c) National quarterly uptake.

From our calculation, approximately 36.6% of the cumulative CO_2 emissions from cement production in mainland China between 1992 and 2021 can be offset through concrete weathering carbonation. An analysis of the carbon offset rates of each province during this period (fig. 2) reveals that carbon offset rate are significantly higher in developed regions, especially in the eastern coastal areas. For example, Beijing and Shanghai possess a carbon offset rate of over 100%. This reflects a carbon negative emission in both cities due to the fact that cement used to produce concrete in these regions is mainly sourced from the surrounding regions or elsewhere.

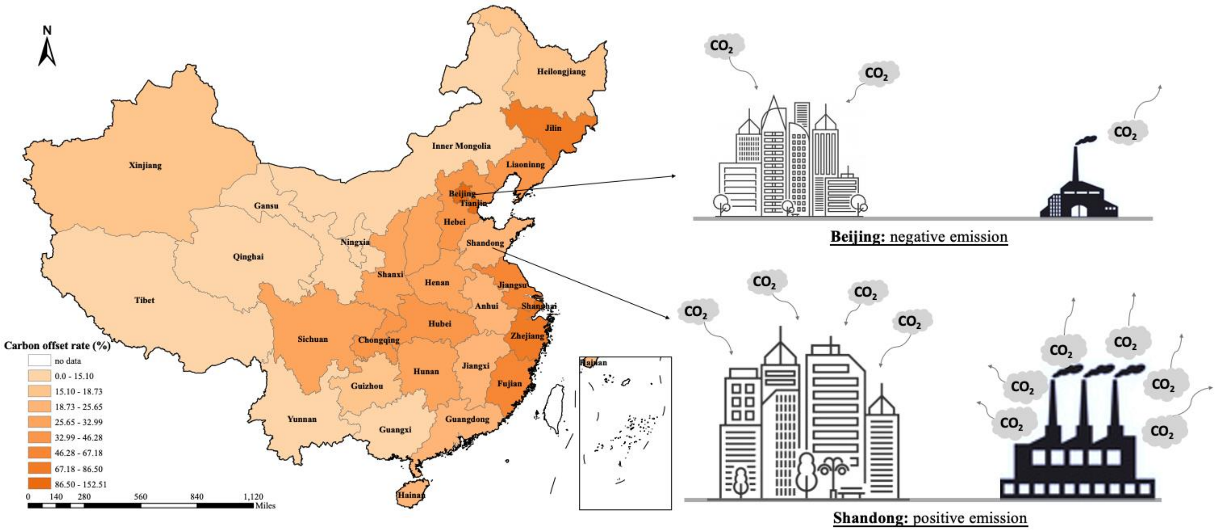


Fig.2. Average carbon offset rate across different provinces in mainland China from 1992 to 2021.

Keywords

Concrete; weathering carbonation; carbon sequestration; Mainland China; quarterly climate impact

Sponsor:

NELD
耐尔得智能科技