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High-Throughput Simulations of Phase Precipitation in Additively Manufactured Al Alloy

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Abstract. Additive manufacturing (AM) provides numerous advantages compared to conventional manufacturing methods, such as high design freedom and low material waste. Among the available materials, precipitation-hardenable aluminum alloys are highly attractive for AM due to their high specific strength and low density. Precise control of the processing conditions during AM and post heat treatment (HT) is required to tailor the final mechanical properties. Consequently, many variables, such as the chemical composition and process and HT parameters, must be considered to design suitable alloys for AM. Experimental investigations are, however, limited in variation of these variables. Therefore, computational alloy design approaches allowing for a faster evaluation of many possible variations must be developed. This work presents a high-throughput approach to determine the precipitation kinetics and thermodynamic properties based on the CALculation of PHAse Diagrams (CALPHAD) method. The developed approach is successfully validated for an Al-Mg-Si-Ti-Fe alloy and is applied to screen 243 combinations of chemical compositions and HT parameters. The results confirm the microstructural stability of the Al-Mg-Si-Ti-Fe system to small composition variations.

Introduction

AM offers high flexibility in both the component's design and locally tailored properties, leading to a shorter manufacturing time than traditional manufacturing methods. Precise control of the processing conditions and microstructure is required to design alloys with specific properties. For AM, it is particularly important to consider the high cooling rates and their impact on microstructure and final properties [1]. Amongst other alloy systems, recent literature studies have shown that the AM processing conditions, the chemical composition, and the HT parameters significantly affect the microstructure and mechanical properties of the well-known Al-Mg based alloys [2]. Precipitation-hardenable Al-Mg alloys with Sc, Zr, or Ti additions represent attractive AM alloy candidates showing excellent mechanical properties [3, 4]. Process-wise, the precipitation is especially affected by the high cooling rates, the complex intrinsic HT (i.e. the remelting/reheating of the already solidified material), and the post HT. More research is needed to control the precipitation during the AM process and the subsequent post HT, and thus the mechanical properties. This can be achieved by adjusting the thermal history of the AM process which includes the AM process itself with its rapid solidification and the cyclic reheating/recooling and as well as the subsequent post HT. This requires the consideration of numerous variables, e.g. the chemical composition and the process and HT parameters, that are mutually dependent on each other. To account for this high dimensionality, high-throughput computational approaches allow for investigating a significantly larger design space with fewer costs, time, and resources compared to traditional experimental approaches [5]. Therefore, using such high-throughput computational approaches reduces the risk of neglecting promising alternatives, allows testing more and even completely unknown variations, and leads to faster development times. The CALPHAD

method is a versatile, widespread, and well-known tool for rapid alloy design. In recent literature studies concerning rapid alloy development [6, 7], the CALPHAD method has been combined with computational high-throughput approaches allowing the screening of more than thousands of different variables. For instance, Tang et al. [6] developed a high-throughput CALPHAD-based framework to assess the cracking behavior of different nickel-based superalloys, including determining the main influencing factors. Wang and Xiong [7] developed a similar high-throughput CALPHAD-based approach to optimize the chemical composition of a specific steel while considering the composition variation in the feedstock material. This approach increased the rate of successful AM builds significantly. So far, the correlation between the entire thermal cycle (solidification and solid-state evolution during the intrinsic HT and the post HT) and the precipitation behavior has not been considered. The aim of this work is to include the entire thermal cycle into the high-throughput approach for predicting the precipitation behavior during AM and the post HT. The developed approach is applied to design Ti-modified Al-Mg alloys for laser-based powder bed fusion (PBF-LB) by using combined modeling of the solidification process during AM and the solid-state precipitation during post HT.

Materials and Methods

Computational alloy design approach

To perform the high-throughput CALPHAD-based calculations, a special framework CAROUSEL (ComputAtional fRamework for high thrOUpghput microStructurE simuLations) has been designed in this work and published on GitHub [8]. It is responsible for configuring the high-throughput calculations, distributing the configured calculations to multiple instances of the external CALPHAD software, and managing the data storage and visualization. For the CALPHAD computations, an external CALPHAD software (here, MatCalc [9]) is accessed by CAROUSEL via an implemented interface.

The respective workflow of CAROUSEL is shown in Fig. 1. CAROUSEL is divided into three different layers. In the data layer, the desired screening design space, consisting of the chemical compositions and the HT parameters of interest, the corresponding material databases for the CALPHAD-based and kinetic calculations, and the configurations for the calculations are defined. To model the AM process (point 1 to 1a in the input box of Fig. 1), the Scheil-Gulliver equation is chosen. Literature has demonstrated the feasibility of using the Scheil-Gulliver equation to approximate rapid solidification processes [10]. With the Scheil-Gulliver equation, the phase evolution can only be predicted until the material is fully solidified (point 1a in Fig. 1). After the rapid solidification, a rapid cooling until room temperature (point 1a to 2) takes place and is followed by a user-defined post HT (point 2 to 3). Each step in the HT can be defined separately. The solid-state precipitation during the post HT is determined by suitable kinetic models implemented in the selected CALPHAD software (here, MatCalc software [9]). The calculations configured in the data layer are then executed in the application and calculation layers, utilizing a distributed and parallel approach. For executing the CALPHAD-based and kinetic calculations, communication between the calculation layer and the external CALPHAD software MatCalc is established. After the calculations are performed successfully, all data is saved in a database. Typical outputs of the framework are information about the precipitation behavior: type of phases, phase fraction, number density, and size of the precipitates. Further outputs, such as strength contribution from precipitates, can be implemented on demand. Several visualization and data analysis tools are available to assess the output.

Validation and application

Ti-modified Al-Mg alloys were used for testing the developed simulation framework. First, a validation for one single composition of the Al-Mg-Si-Ti-Fe system in the as-built state (point 1a in 1) and for different heat treated conditions (at 250 °C and 350 °C for 1, 3, and 6 h) was performed. Second, after validation, the respective alloy system was screened with varying elemental compositions (0.5 - 1.0 Mg, 0.5 - 1.0 Si, 0.1 - 0.4 Fe, 0.9 - 2.0 Ti; in wt.%) and with varying HT conditions (at 250 °C, 350 °C, and 450 °C for 1, 3, and 6 h) to investigate their impact on the precipitation. 243 combinations

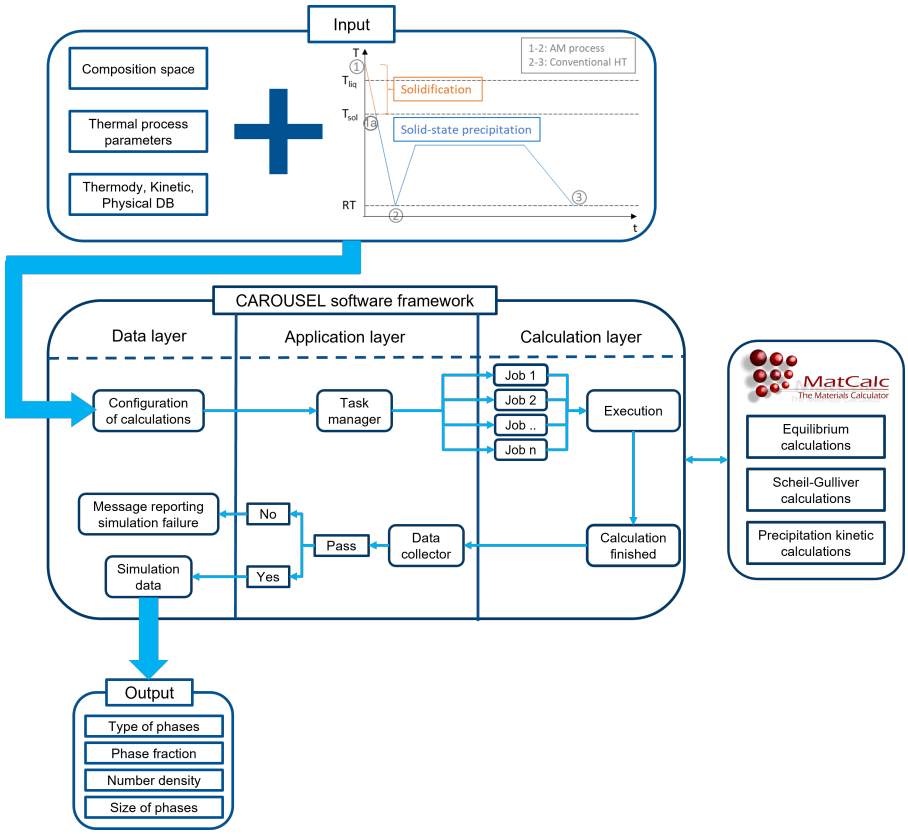

Fig. 1: Flowchart showing the workflow of CAROUSEL.

of different chemical compositions and HT parameters were assessed using the ME-Al database of MatCalc [9].

Experimental procedure

Bulk specimens of the single composition were produced on an EOS M290 PBF-LB system (1050 nm laser wavelength, 300 W power, 60 µm layer thickness, 100 µm hatch spacing, 1200 mm/s scanning speed, meander scan strategy). The HTs were conducted in a Carbolite Gero CWF 13/13 furnace under atmospheric pressure at different temperatures, 250 and 350 °C, and for various durations of 1, 3, and 6 h at each temperature. The microstructure in the as-built and heat treated conditions was studied via scanning electron microscopy (SEM) using a Zeiss Merlin Gemini Field Emission SEM. The specimens were prepared through mechanical grinding down to 1000 grit SiC paper, followed by grinding using 9 µm diamond suspension, and finally being polished using a 0.25 µm silica suspension

Results and Discussion

Validation

The phase fraction and mean radius are important values that characterize the precipitates. In aluminum alloys, generally, a higher phase fraction and a smaller radius lead to a higher material strength. Fig. 2a and b show the computational calculation results for the single Al-0.9Mg-0.7Si-1.3Ti-0.1Fe (in wt.%) composition, i.e. the phase fraction and the mean radius of the precipitates over the HT time for exemplary HTs (250 °C and 350 °C up to 6 h). The Scheil-Gulliver calculations predict that the face-centered cubic (fcc) Al matrix, primary Mg_2Si, and Al_3Ti precipitates form during the solidification (time t = 0 h) which is in accordance with literature observations [11]. During all HTs, the formation of secondary Mg_2Si precipitates smaller than the primary ones is predicted. The phase fraction of the primary Mg_2Si increases slightly, especially with increasing temperature and duration, whereas the radius remains stable. At 250 °C, the phase fraction and the radius of secondary Mg_2Si precipitates increase linearly with time. At 350°C, the phase fraction and the radius increase for shorter

HTs (up to 2 h) and remain constant afterward (durations longer than 2 h). Compared to 250 °C, higher phase fractions and sizes are reached, as higher diffusion rates lead to coarser precipitates. Negligible changes in phase fraction and size of the primary Al_3Ti precipitates are predicted, which complies with the literature reporting on slow Ti diffusivity in Al [4]. No secondary Al_3Ti precipitates are computationally predicted to form under the selected HT conditions. According to the simulation results of the selected composition, the Ti is already solved in the primary Al_3Ti precipitats after solidification. Consequently, no or negligible Ti is available during the HT for secondary precipitation. Roscher et al. [4] revealed that secondary Al_3Ti precipitates can be formed under certain process conditions. However, the slow diffusivity of Ti usually leads to negligible Al_3Ti secondary precipitation during HT.

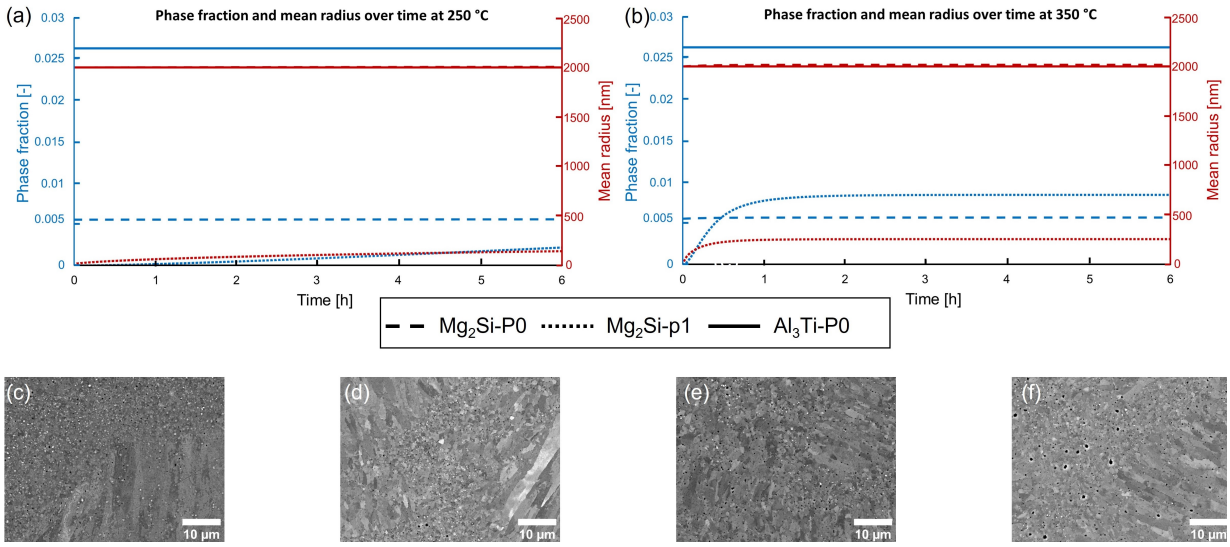

Fig. 2: Simulation results of Al-0.9Mg-0.7Si-1.3Ti-0.1Fe heat treated at (a) 250 °C and (b) 350 °C showing the mean radius and phase fraction over time (up to 6 h). Comparison of the simulation results with exemplary SEM micrographs of the specimens in the (c) as-built state and in the heat treated conditions (d) at 250 °C for1 h, (e) at 350 °C for 1 h, and (f) at 350 °C for 6 h. The suffix ”P0” refers to primary precipitates and ”P1” to secondary precipitates.

The results of the calculations are as well confirmed by the SEM micrographs of the selected composition (Al-0.9Mg-0.7Si-0.1Fe-1.3Zr), as shown in Fig. 2c-f. In the SEM micrographs, the small white spots are Al_3Ti precipitates and the black spots are Mg_2Si precipitates, both embedded in the Al fcc matrix. The comparison of the SEM micrographs between the HT at 250°C-1h (d) and 350 °C-1h (e) and between the HT at 350°C-1h (e) and 350 °C-6h (f) confirms that the phase fraction and mean radius of the Mg_2Si precipitates increase with increasing temperature for a fixed duration (1 h) or with increasing duration at a fixed temperature (350 °C). Further characterization is needed to determine the exact fractions of primary and secondary precipitates such as electron backscatter diffraction investigations. The SEM micrographs do not allow a detailed investigation of the Al_3Ti precipitates including an assessment of the sizes and the type of precipitates (primary and/or secondary). Higher resolution characterization, such as transmission electron microscopy, would be needed. To conclude, successful validation for one single composition in the as-built state and for various HTs is shown.

Application

Recent studies in literature highlighted the variations in the chemical composition of AM parts compared to the targeted composition (originating from e.g. composition variation in the feedstock material or process-induced evaporation) and its impact on the microstructure and final properties [7]. In this context, the performed screening aims to investigate the alloy design space around the validated composition, detect underlying correlations between chemical composition, HT, and precipitation behavior, identify potential new promising compositional alternatives, and assess the microstructural

stability of the corresponding composition regarding minor variations. Fig. 3 displays the phase fraction and the mean radius for each single variable combination of chemical composition and HT parameters (each circle represents several combinations with similar values). It reveals that for all chemical compositions and HTs the same phases are formed, only the phase fraction and mean radius changes slightly. For instance, the phase fraction of the primary Al_3Ti precipitates increases with higher Ti contents. The screening confirms the high microstructural stability of this alloy system against small composition changes. A deeper analysis is needed to identify the underlying correlations between chemical composition, HT parameters, phase fractions, and mean radius, allowing a more profound understanding of the alloy system and a sophisticated selection of promising combinations of chemical compositions and heat treatment parameters in the future.

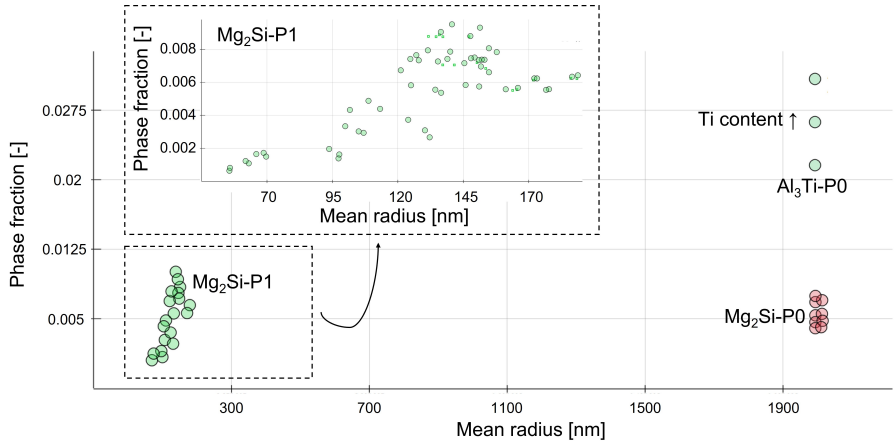

Fig. 3: Phase fraction versus mean radius for all 243 screened variables combinations of chemical composition and HT conditions (each circle represents several combinations with similar values). The suffix ”P0” refers to primary precipitates and ”P1” to secondary precipitates.

The obtained results highlight the capability of the CAROUSEL framework to rapidly investigate many different chemical compositions and HT parameters without needing time- and cost-intensive experiments. Compared to equilibrium or Scheil-Gulliver calculations, combined Scheil-Gulliver and precipitation kinetic calculations require longer calculation times per combination. Although only 243 combinations were selected for the first application, with the developed framework a screening with more than thousand of different combinations of chemical composition and heat treatment parameters can be performed. This will require high-performance computing and is the goal of further development.

Conclusion and Outlook

In the present work, a combined high-throughput thermodynamic and kinetic simulation approach has been developed for the rapid design of alloys for AM. A special framework called CAROUSEL has been designed to implement this approach. Successful validation was conducted for a specific Al-0.9Mg-0.7Si-1.3Ti-0.1Fe alloy heat treated at various conditions. The comparison between the calculation results and the SEM micrographs revealed the same phases and phase evolution behavior during the HT. CAROUSEL was then applied to investigate the precipitation behavior of additively manufactured Al-Mg-Si-Ti-Fe alloys by screening 243 combinations of different chemical compositions and HT parameters. The obtained results have confirmed the microstructural stability of the Al-Mg-Si-Ti-Fe alloy system to small composition variations concerning the occurring phases. Nevertheless, the phase fraction and size are affected by the chemical composition and the HT parameters. Following conclusions regarding the phase evolution in the Al-Mg-Si-Ti-Fe alloy system can be drawn. First, the phase fraction and size of the Mg_2Si precipitates increase with higher aging temperatures and longer durations. Second, for the selected chemical composition and heat treatment conditions, negligible

changes of the Al_3Ti precipitates during the HT are observed due to the low diffusivity of Ti in Al. The results of the performed screening highlight the benefits of such high-throughput thermodynamic calculations: many different (not yet studied) chemical compositions and HT parameters can be easily simulated without performing any time-, cost-, and resource-intensive experiments. Compared to existing solutions, the approach proposed in the present work considers both solidification during AM processing and solid-state precipitation after the post HT. Further development is planned in the future, which considers the intrinsic cyclic HT during AM and the modeling of mechanical properties.

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References

[1] T. DebRoy et al., Additive manufacturing of metallic components - Process, structure and properties, Progress in Materials Science 92 (2018) 112-224.

[2] A.I. Mertens, J. Delahaye and J. Lecomte-Beckers, Fusion-Based Additive Manufacturing for Processing Aluminum Alloys: State-of-the-Art and Challenges, Advanced Engineering Materials 19 (2017) 1700003.

[3] A.B. Spierings et al., Microstructural features of Sc- and Zr-modified Al-Mg alloys processed by selective laser melting, Materials & Design 115 (2017) 52-63.

[4] M. Roscher et al., Development of Al-Ti-based alloys for laser powder bed fusion, Additive Manufacturing 47 (2021) 102315.

[5] G.B. Olson, Genomic materials design: The ferrous frontier, Acta Materialia 61 (2013) 771-781.

[6] Y.T. Tang et al., Alloys-by-design: Application to new superalloys for additive manufacturing, Acta Materialia 202 (2021) 417-436.

[7] X. Wang and W. Xiong, Uncertainty quantification and composition optimization for alloy additive manufacturing through a CALPHAD-based ICME framework, npj Computational Materials 6 (2020) 188.

[8] S. Carrion Ständer, N. Barschkett, E.Kabliman, CAROUSEL (Version v2.2-beta.6) [Computer Software], https://github.com/cmatdesign/CAROUSEL.

[9] E. Kozeschnik, Mean-Field Microstructure Kinetics Modeling, in: F.G. Caballero (ed.), Encyclopedia of Materials: Metals and Alloys, Elsevier, Oxford, 2022, pp. 521-526.

[10] B. Bocklund et al., Experimental validation of Scheil–Gulliver simulations for gradient path planning in additively manufactured functionally graded materials, Materialia 11 (2020) 100689.

[11] R. Li et al., Developing a high-strength Al-Mg-Si-Sc-Zr alloy for selective laser melting: Crack-inhibiting and multiple strengthening mechanisms, Acta Materialia 193 (2020) 83-98.