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Increasing the energy efficiency of ultrasonic pre-treatment systems in wastewater treatment plants

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Abstract

The application of ultrasound-induced cavitation for the disintegration of sludge particles can lead to improved anaerobic digestion and an increase in methane yield. This effect was demonstrated at laboratory-scale by sonication of the substrate and subsequent incubation in batch tests multiple times.

While translating this concept to the full-scale, the very large volume to be sonicated poses a particular challenge. While the electrical energy demand and the size of the ultrasound system are insignificant at laboratory-scale, the energy consumption and the costs for the installation of an ultrasound system at full-scale are subject to aggravating economic circumstances. For the efficient scale-up of the systems and proper generation of cavitation bubbles in the medium, it is therefore of considerable importance to identify the limits for reducing the energy input and for optimizing the reactor design.

With this intention, this thesis first investigated the formation and propagation of cavitation bubbles in different substrates and different reactor configurations. For this purpose, specially constructed rectangular flow cells with different gap spacings were manufactured and equipped with piezoelectric surface oscillation systems.

The subsequent sonication tests demonstrated that the sonication of water differs considerably from the sonication of digested sludge containing solids or excess sludge, which is even more particle-enriched. The distinct attenuation of the ultrasonic field and the corresponding suppression of cavitation bubbles allow only a very small distance between the sound emitting surfaces of a flow reactor of a maximum of 60 mm.

In order to determine the influence of the cavitation intensity on the desired sonication result, the properties of the sludge before and after sonication were investigated in an extensive series of experiments. This included the change in viscosity, the development of the relative methane yield, the change in dissolved chemical oxygen demand (COD) as well as the degree of disintegration of the substrate.

In addition to the use of two different tubular surface oscillating systems, a reactor with a rod-shaped sonotrode was included in the test installation, and the possible advantages of one oscillating system design for increasing the energy efficiency of the sonication process were investigated.

For an optimized comparison of the different reactor designs, the experimental setup was arranged in such a way that a comparable specific energy input could be applied to the substrate. In order to investigate to what extent the energy input influences the sonication effect, the experiments were repeated with three different specific energy inputs. The results revealed that the sludge properties could be influenced by all the oscillation systems investigated. However, especially with regard to sonication with low energy input of 300 $kJ/kg_{TS,}$ which could be realized on a large-scale with comparatively low material expenditure, there are differences in the sonication efficiency of the respective substrates. While the sonication of digested sludge with tube reactors and low energy input showed the highest effect of up to 22% increase in methane yield, the use of a sonotrode for the sonication of excess sludge proved to be advantageous with an increase of 25%. In addition, sonication of digested sludge showed a much greater increase of 30% in gas production in the short term (4 days), which, however, decreased over the course of the batch test over 30 days compared to the unsonicated sample. With the sonication of excess sludge, only a small increase in yield of 18% on average was observed immediately after incubation in the batch test, which, however, proved to be largely stable over the entire test period.

When considering different surface oscillation systems, the tubular reactor with a small diameter of 53 mm proved to be superior to a larger tubular reactor with a diameter of 82 mm and a reduced power density with regard to the achievable increase in methane yield in digested sludge. These observations, therefore, favor a development strategy that uses a large number of small tubular reactors for scaling rather than increasing the size of the reactors.

This approach, however, leads to new questions in the sonication of large amounts of digested sludge in a limited time. The cost-effective connection of a large number of reactors in series causes high average flow velocities within the reactor and leads to the risk of suppressing cavitation development due to the unstable flow behavior. In order to investigate the influence of increased tube flow on the cavitation intensity in the reactor, a series of experiments was performed, which allowed the variation of the mean flow velocity in a high bandwidth from 0 up to 2 m/s.

The results obtained show an intensive suppression of cavitation formation in turbulent flows, both for the sonication of tap water and in the use of digested sludge. Deviating from the continuous decrease of the cavitation intensity with the increasing flow in water, an increase of the cavitation intensity at medium flow velocities of 0.8 to 1.3 m/s could be observed in the sonication of digested sludge. This can be explained by the shear-thinning properties of the digested sludge and the correspondingly favored formation of cavitation bubbles in the medium.

The results obtained in the course of the experiments were used to develop several large-scale reactor concepts, which were subsequently tested under field-scale conditions. The ultrasonic systems were installed in a bypass of the excess sludge line of the Starnberg wastewater treatment plant and of the digested sludge circulation line of the Traunstein wastewater treatment plant.

Although the tests carried out confirmed the feasibility of large-scale ultrasound systems for the disintegration of sewage sludge, they were only able to achieve a small additional yield in the already optimized and under-utilized digesters. The application of the investigated ultrasonic technology, therefore, offers the potential for optimal utilization of the existing digestion capacities, especially in plants with high load factors, and could reduce the necessity of a much more cost-intensive new construction of additional digestion volume at these plants.

Zusammenfassung

Der Einsatz von ultraschallinduzierter Kavitation zur Desintegration von Schlammpartikeln kann zu einem verbesserten anaeroben Abbau und einem Anstieg der Methanausbeute führen. Dieser Effekt wurde im Labormaßstab durch die Beschallung des Substrats und anschließende Inkubation in Batchtest bereits mehrfach nachgewiesen.

Zur Übertragung der Mechanismen auf den großtechnischen Maßstab, stellt insbesondere das sehr große Volumen der zu beschallenden Masse eine große Herausforderung dar. Während der Bedarf an elektrischer Energie und die Größe des Ultraschallsystems im Labormaßstab unerheblich sind, unterliegen der Energiebedarf und der Aufwand für die Errichtung des Ultraschallsystems im großtechnischen Maßstab erschwerenden wirtschaftlichen Rahmenbedingungen. Zur effizienten Skalierung der Anlagen und Mechanismen zur Erzeugung von Kavitationsblasen im zu beschallenden Medium ist es daher von erheblicher Bedeutung, die Grenzen zur Verringerung des Energieeinsatzes und zur Optimierung der Reaktorbauformen zu identifizieren.

Mit dieser Absicht wurde in dieser Arbeit zunächst die Entstehung und Ausbreitung von Kavitationsblasen in verschiedenen Substraten sowie unterschiedlichen Bauformen untersucht. Dabei wurden eigens konstruierte, rechteckige Durchflusszellen mit unterschiedlichen Spaltabständen angefertigt und mit piezoelektrischen Flächenschwingsystemen bestückt.

Die anschließenden Versuche machten deutlich, dass sich die Beschallung von Wasser erheblich von der Beschallung von feststoffhaltigem Faulschlamm oder dem noch stärker mit Partikeln angereichertem Überschussschlamm unterscheiden. Die deutliche Dämpfung des Ultraschallfeldes und entsprechende Unterdrückung von Kavitationsblasen erlauben nur einen sehr geringen Abstand der schallabgebenden Flächen eines Durchflussreaktors von maximal 60 mm.

Zur Ermittlung des Einflusses der Kavitationsstärke auf das gewünschte Beschallungsergebnis wurden die Eigenschaften des Schlamms vor und nach der Beschallung in einer umfangreichen Versuchsreihe untersucht. Dazu gehörte die Veränderung der Viskosität, die Entwicklung der relativen Methanausbeute, die Veränderung des gelösten chemischen Sauerstoffbedarfs als auch der Aufschlussgrad des Substrates.

Ergänzend zum Einsatz von zwei rohrförmigen Flachschwingsystemen wurde ein Reaktor mit einer stabförmigen Sonotrode in den Versuchseinbau einbezogen und die möglichen Vorteile einer Schwingsystembauform zur Steigerung der Energieeffizienz des Beschallungsvorgangs untersucht. Zum optimalen Vergleich der unterschiedlichen Reaktorbauformen wurde der Versuchsaufbau so gestaltet, dass ein vergleichbarer spezifischer Energieeintrag in das Substrat eingebracht werden konnte.

Zur Untersuchung inwiefern der Energieeintrag einen Einfluss auf das Beschallungsergebnis aufweist, wurden die Experimente mit drei unterschiedlichen Beschallungsdauern wiederholt. Die Ergebnisse zeigten, dass sich mit allen untersuchten Schwingsystemen eine Beeinflussung der Schlammeigenschaften erzielen lässt. Insbesondere im Hinblick auf Beschallungen mit geringen Energieeinträgen von 300 kJ/kg_{TS}, welche sich im großtechnischen Maßstab mit einem vergleichsweise geringen materiellen Aufwand realisieren ließen, gibt es jedoch Unterschiede in Bezug auf die Beschallungseffizienz der jeweiligen Substrate. Während die Beschallung von Faulschlamm mit Rohrreaktoren und geringen Energieeinträgen die höchste Steigerung des Methanertrags um 22% zeigte, so erwies sich der Einsatz einer Sonotrode bei der Beschallung von Überschussschlamm mit einer Steigerung um 25% als vorteilhaft. Darüberhinaus zeigte die Beschallung von Faulschlamm kurzfristig (4 Tage) einen deutlich größeren relativen Anstieg der Gasproduktion um 30%, welche sich jedoch über den Verlauf des Batchversuchs über 30 Tage gegenüber der unbeschallten Probe verringerte. Während der Beschallung von Überschussschlamm konnten unmittelbar nach der Inkubation im Batchversuch nur ein geringerer Mehrertrag von durchschnittlich 18% beobachtet werden, welcher sich jedoch über die gesamte Versuchsdauer als weitgehend stabil erwies.

Bei der Betrachtung unterschiedlicher Flachschwingsysteme zeigte sich der Rohrreaktor mit einem geringen Durchmesser von 53 mm gegenüber eines größeren Rohrreaktors mit einem Durchmesser von 82 mm und einer verringerten Leistungsdichte in Bezug auf die erzielbare Steigerung des Methanertrags in Faulschlamm, überlegen. Diese Beobachtungen begünstigen daher eine Entwicklungsstrategie, welche zur Skalierung eine Vielzahl kleiner Rohrreaktoren nutzt, anstatt die Größe der Reaktoren zu steigern.

Dieser Ansatz führt jedoch zu neuen Fragestellungen bei der Beschallung großer Mengen Faulschlamms in einer begrenzten Zeit. Die kostengünstige Reihenschaltung einer Vielzahl von Reaktoren verursacht hohe mittlere Fließgeschwindigkeiten innerhalb des Reaktors und führt zu dem Risiko einer Unterdrückung der Kavitationsentwicklung aufgrund des instabilen Strömungsverhaltens. Zur Untersuchung des Einflusses einer erhöhten Rohrströmung auf die Kavitationsstärke im Reaktor wurde eine Versuchsreihe durchgeführt, welche die Variation der mittleren Strömungsgeschwindigkeit in einer hohen Bandbreite von 0 bis zu 2 m/s ermöglichte.

Die erzielten Ergebnisse zeigen eine intensive Unterdrückung der Kavitationsentstehung bei turbulenten Strömungen, sowohl bei der Beschallung von Leitungswasser als auch bei dem Einsatz von Faulschlamm. Abweichend von dem kontinuierlichem Absinken der Kavitationsintensität mit ansteigender Strömung in Wasser, konnte bei der Beschallung von Faulschlamm ein Anstieg der Kavitationsintensität bei mittleren Strömungsgeschwindigkeiten von 0,8 bis 1,3 m/s beobachtet werden. Dies kann durch das scherverdünnende Verhalten des Faulschlamms und der dementsprechend begünstigten Entstehung von Kavitationsblasen im Medium erklärt werden.

Die im Rahmen der durchgeführten Experimente erzielten Ergebnisse dienten zur Entwicklung mehrerer großtechnischer Reaktorkonzepte, welche anschließend unter realen Bedingungen erprobt wurden. Die Installation der Ultraschallsysteme erfolgte dabei unmittelbar an der Überschussschlammleitung der Kläranlage Starnberg, sowie an der Faulschlammumwälzleitung der Kläranlage Traunstein.

Die durchgeführten Erprobungen zeigten zwar die grundsätzliche Realisierbarkeit großtechnischer Ultraschallsysteme zur Desintegration von Klärschlamm, konnten jedoch nur einen geringen Mehrertrag in den bereits optimierten und wenig ausgelasteten Faultürmen erzielen. Der Einsatz der untersuchten Ultraschalltechnologie bietet daher insbesondere in stark ausgelasteten Anlagen das Potential zur optimalen Ausnutzung der vorhandenen Faulturmkapazitäten und könnte in diesen Anlagen die Notwendigkeit eines wesentlich kostenintensiveren Neubaus weiteren Faulraums reduzieren.

Research papers, author contributions and topic-related publications

This cumulative doctoral thesis is based on the following peer-reviewed research papers, which are presented in Chapter 3.

Paper #1 (Chapter 3.1):

Bandelin, J.; Lippert, T.; Drewes, J. E.; Koch, K.: Cavitation field analysis for an increased efficiency of ultrasonic sludge pre-treatment using a novel hydrophone system. Ultrasonics Sonochemistry 42, 2018, 672-678

- Author Contributions: Jochen Bandelin conducted the literature research and designed the test reactors and peripheral setup. Experiments were performed by Jochen Bandelin and Thomas Lippert together. Jochen Bandelin was responsible for the data analysis and preparation of the manuscript. Thomas Lippert complemented the manuscript with initial proofreading. Jörg E. Drewes and Konrad Koch supervised this study and reviewed the manuscript.

Paper #2 (Chapter 3.2):

Bandelin, J.; Lippert, T.; Drewes, J. E.; Koch, K.: Assessment of sonotrode and tube reactors for ultrasonic pre-treatment of two different sewage sludge types. Ultrasonics Sonochemistry 64, 2020, 105001

- Author Contributions: Jochen Bandelin was responsible for the design and setup of the test and development of the experiment procedure. Experiments were performed by Jochen Bandelin and Thomas Lippert together. Jochen Bandelin conducted the data analysis and prepared the manuscript. Thomas Lippert complemented the manuscript with initial proofreading. Jörg E. Drewes and Konrad Koch supervised this study and reviewed the manuscript.

<u>Paper #3 (Chapter 3.3):</u>

Bandelin, J.; Lippert, T.; Drewes, J. E.; Koch, K.: Impact of high flow rates and increased viscosity of digested sewage sludge on the cavitation intensity in ultrasonic tube reactors. Chemical Engineering & Processing: Process Intensification 152, 2020, 107925

- Author Contributions: Jochen Bandelin designed the research plan and developed the experimental setup of the test. The assembly of the test system and the performance of the experiments were carried out by Jochen Bandelin and Thomas Lippert together. Jochen Bandelin was responsible for data analysis and prepared the manuscript. Thomas Lippert complemented the manuscript with initial proofreading. Jörg E. Drewes and Konrad Koch supervised this study and reviewed the manuscript.

<u>Topic-related international publications:</u>

Lippert, T.; **Bandelin, J.**; Musch, A.; Drewes, J. E.; Koch, K.: Energy-positive sewage sludge pre-treatment with a novel ultrasonic flatbed reactor at low energy input. Bioresource Technology 264, 2018, 298-305

Lippert, T.; **Bandelin, J.**; Schlederer, F.; Drewes, J. E.; Koch, K.: Impact of ultrasound-induced cavitation on the fluid dynamics of water and sewage sludge in ultrasonic flatbed reactors. Ultrasonics Sonochemistry 55, 2019, 217-222

Lippert, T.; **Bandelin, J.**; Schlederer, F.; Drewes, J. E.; Koch, K.: Effects of ultrasonic reactor design on sewage sludge disintegration. Ultrasonics Sonochemistry 68, 2020, 105223

Lippert, T.; **Bandelin, J.**; Vogl D.; Alipour Tesieh, Z.; Wild, T.; Drewes, J.E.; Koch, K.: Full-Scale Assessment of Ultrasonic Sewage Sludge Pretreatment Using a Novel Double-Tube Reactor, ACS ES&T Engineering 1,2, 2021, 298-309

Lippert, T.; **Bandelin**, J.; Xu, J.; Liu, Y.C.; Hernandenz Robles, G.; Drewes, J.E.; Koch, K.: From pre-treatment to co-treatment - How successful is ultrasonication of digested sewage sludge in continuously operated anaerobic digesters? Renewable Energy 166, 2020, 56-65

<u>Conference Oral Presentations:</u>

Koch, K.; **Bandelin, J.**: Effect of ultrasonic pre-treatment on anaerobic digestibility and sludge characteristics of two different effluents. 21st European Biomass Conference and Exhibition, 3-7 June 2013, Copenhagen, Denmark.

Bandelin, J.; Koch, K.: Effect of ultrasonic pre-treatment with tube-reactors on the biogas yield. BiogasScience 2014: International Conference on Anaerobic Digestion, 26-30 Oktober 2014, Vienna, Austria.

Bandelin, J.; Koch, K.: Effect of ultrasonic pre-treatment with tube-reactors on the biogas yield. 5th International Symposium on Energy from Biomass and Waste, 17-20 November 2014, Venice, Italy.

Bandelin, J.; Drewes, J.; Koch, K.: Cavitation field analysis for improving the substrate pre-treatment by ultrasound. 15th IWA World Conference on Anaerobic Digestion, 17-20 Oktober 2017, Beijing, China

Lippert, T.; **Bandelin, J.;** Musch, A.; Drewes, J.E.; Koch, K.: Performance assessment of a novel ultrasonic flatbed reactor for sludge pre-treatment. 15th IWA World Conference on Anaerobic Digestion, 17-20 Oktober 2017, Beijing, China

Lippert, T.; **Bandelin, J.;** Schlederer, F.; Drewes, J. E.; Koch, K.: "Power-to-cavitation (PTC)": Proposing a new parameter to assess the energy efficiency of ultrasonic reactors for sewage sludge pre-treatment. International Conference on Anaerobic Digestion "Biogas Science 2018", 17-19 September 2018, Turin, Italy

Lippert, T.; **Bandelin, J.;** Drewes, J. E.; Koch, K.: Energy-efficient pre-treatment of waste activated sludge using a novel ultrasonic flatbed reactor. SMICE 2018 - Sludge Management in Circular Economy, 23-25 May 2018, Rome, Italy.

Lippert, T.; **Bandelin, J.**; Koch, K.; Drewes, J. E.: Ökonomie der Klärschlammdesintegration mittels Ultraschall. 46. Abwassertechnischen Seminar „Innovative Strategien zum Umgang mit Klärschlamm", Lehrstuhl für Siedlungswasserwirtschaft, Technische Universität München, 4 July 2018, Ismaning Germany.

Bandelin, J.; Lippert, T.; Koch, K.; Drewes, J. E.: Energieeffiziente Klärschlammvorbehandlung durch innovative Ultraschalldesintegration. 46. Abwassertechnisches Seminar „Innovative Strategien zum Umgang mit Klärschlamm", Lehrstuhl für Sieldungswasserwirtschaft, Technische Universität München, 4 July 2018, Ismaning, Germany.

Conference Poster Presentations:

Bandelin, J.; Lippert, T.; Drewes, J. E.; Koch, K.: Cavitation field analysis for higher energy efficiency in substrate pre-treatment by ultrasound. SMICE 2018 - Sludge Management in Circular Economy, 23-25 May 2018, Rome, Italy.

Lippert, T.; **Bandelin, J.**; Drewes, J. E.; Koch, K.: Impact of pressure on the efficiency of ultrasonic sewage sludge pre-treatment. 16th IWA World Conference on Anaerobic Digestion, 23-27 June 2019, Delft, The Netherlands

Bandelin, J.; Lippert, T.; Drewes, J. E.; Koch, K.: Practical experience with full-scale ultrasonic pre-treatment using a novel reactor design. 16th IWA World Conference on Anaerobic Digestion, 23-27 June 2019, Delft, The Netherlands

Abbreviations

AMPTS	Automatic Methane Potential Test System
ANAMMOX	Anaerobic ammonium oxidation
AOB	Ammonia-oxidizing bacteria
AOP	Advanced oxidation process
BMP	Biochemical methane potential
CAS	Conventional activated sludge
COD	Chemical oxygen demand
sCOD	Soluble chemical oxygen demand
$sCOD_0$	Soluble chemical oxygen demand before treatment
sCODus	Soluble chemical oxygen demand after ultrasonic treatment
$sCOD_{NaOH}$	Soluble chemical oxygen demand after NaOH treatment
DS	Digested sludge
dB	Decibel
DD_{COD}	Degree of disintegration
EBPR	Enhanced biological phosphorus removal
EPS	Extracellular polymeric substances
f_0	Ultrasonic frequency
HRT	Hydraulic retention time
NOB	Nitrite-oxidizing bacteria
PE	Population equivalent
PFAS	Per- and polyfluoralkyl substances
PTC	Power to cavitation
Re	Reynolds number
TS	Total solids
US	Ultrasound
VS	Volatile solids
WAS	Waste activated sludge
WWTP	Wastewater treatment plant

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1 Introduction

1.1 Use of anaerobic digestion in wastewater treatment

In wastewater treatment, the separation of contaminants takes place in various process steps. While coarse solids can be retained by sieves at the inlet of the treatment plant and disposed of as dry material, the dissolved solids must be removed from the water by multi-stage processes (Figure 1)[1]. The water first enters a sedimentation tank where heavy particles such as sand and gravel sink to the bottom and floating fat particles are separated from the water surface. In the following primary clarifier, the primary sludge is separated by sedimentation with a longer retention time compared to the sand trap [2].

Carbon and nitrogen removal takes place in the subsequent conventional activated sludge (CAS) system, which is partly or temporarily aerated for carbon removal and nitrification and subsequent denitrification, which requires anoxic conditions [3]. The formed particulate biomass resulting from the bioconversion of carbon and ammonium accumulates in the CAS tank and is separated from the water in a subsequent secondary settling tank by gravity. From here, most sludge is recycled back to the activated sludge tank to maintain the desired biomass concentration, while the excess sludge is discharged at regular intervals and further thickened by adding flocculants and subsequent centrifugation or screening. The thickening process increases the total solids (TS) content from about 2.5% to a level of 4% to 7%, depending on the amount of flocculant added. This sludge, called waste activated sludge (WAS), is fed into the digester together with the primary sludge harvested in the primary clarifier [1].

When the sludge is digested under anaerobic conditions, a consortium of bacteria and archaea will degrade the sludge components by fermentative metabolic processes. The resulting end products of the anaerobic digestion are mainly methane and carbon dioxide, as well as digested sludge, which is automatically withdrawn from the digester once fed. During anaerobic digestion, multiple process steps are conducted simultaneously in a symbiotic community and need to be well balanced for an efficient degradation.

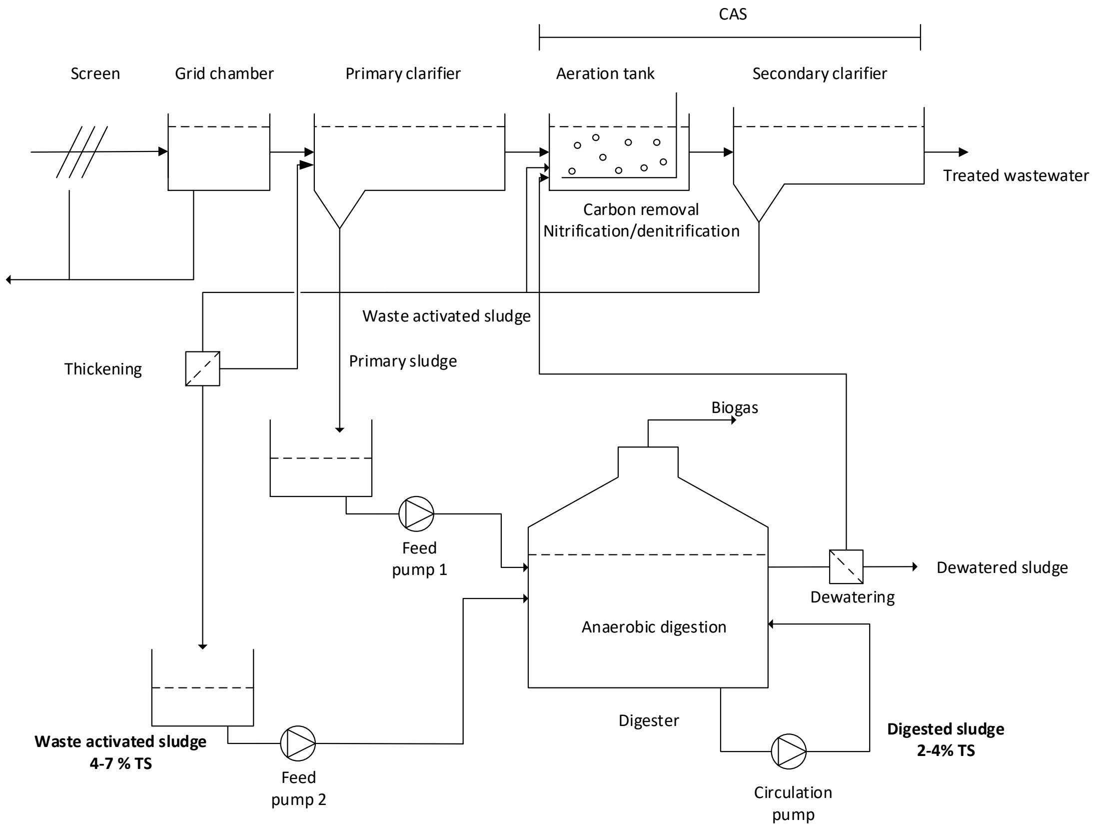

Figure 1: Flow scheme of a wastewater treatment plant with conventional activated sludge (CAS) system and anaerobic stabilization of sewage sludge.

The anaerobic digestion can be divided into four steps, as shown in Figure 2, including hydrolysis, acidogenesis, acetogenesis, and methanogenesis [2].

In the hydrolysis step, the cellular mass of the excess sludge consisting of macromolecules like proteins, polysaccharides, and fats are converted into molecules with a smaller molecular mass, including peptides, saccharides, and fatty acids. This mechanism is induced by enzymes excreted by fermentative bacteria and is often the rate-limiting step in anaerobic digestion of particulate material as a result of the limitations of mass transfer in the substrate [4].

The second step of anaerobic digestion is the acidogenesis, which includes the conversion of the products from hydrolysis to simple molecules with even smaller mass, such as volatile fatty acids (e.g., acetic, propionic and butyric acid), alcohols, aldehydes, and gases like CO_2, NH_3 and H_2 [2].

In the subsequent third step of anaerobic digestion, acetogenic bacteria transfer the products from acidogenesis into acetate, hydrogen, and, carbon dioxide.

The first three steps of anaerobic digestion, which can be described as acid fermentation, provide the fundamental conditions for the fourth step, called methanogenesis. In this stage, the products of acid fermentation, mainly consisting of acetic acid, hydrogen and CO_2, are converted to CH_4 and CO_2. The generated gases auto-diffuse from the liquid and can be collected in the digester headspace. To maintain a high conversion rate of methanogenesis, environmental conditions, such as temperature and pH level as well as the availability of nutrients should be kept constant in a certain range [1].

The methanogens are very sensitive to inhibiting components, which can be present in the inflow of the wastewater or originate from the anaerobic process itself, such as overly high concentration of NH_3.

During stable operation, the intermediate products resulting from hydrolysis are rapidly converted to methane. The concentration of acids is, therefore, relatively low. Disruption of the equilibrium of the anaerobic digestion processes, e.g., caused by shock loading or toxic compounds, can inhibite the highly sensitive methanogens and therefore lead to a higher concentration of intermediate products mostly consisting of volatile fatty acids. The consequent decrease in pH further reduces the activity of methanogens, which will lead to even higher levels of acids and inhibition of methane production, which can lead to a complete collapse of the whole process [5]. Restoration of an optimal pH for methanogenesis in the range of 6.5 to 7.5 can be obtained by drastically reducing the inflow or by the addition of alkalinity and requires a considerable adjustment time. To prevent unstable operating conditions, the composition and amount of the biomass supplied should therefore be as constant and homogeneous as possible [6].

Mechanical pre-treatment also was shown to cause an improvement in the operational stability of the process as the homogeneity of the substrate could be improved [7,8].

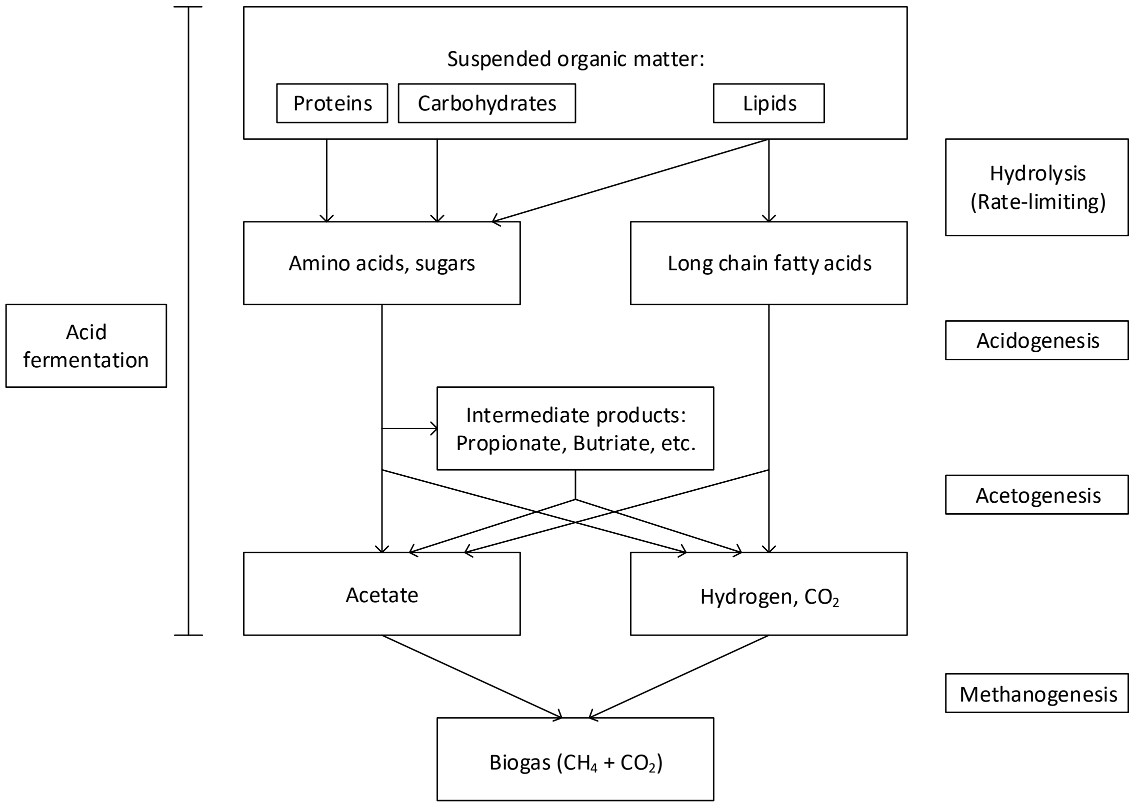

Figure 2: Schematic process of anaerobic digestion for utilization of waste activated sludge and primary sludge in a WWTP digester. Adapted from Gujer and Zehnder (1983).

The main purposes of anaerobic digestion are the improvement of the dewaterability for mass reduction of the sludge, the reduction of its putrefactive capacity, reduction of pathogens when intended for agricultural use, and recovery of resources.

A positive side effect of anaerobic digestion is the generation of methane, which can be used, among others, for power and heat generation by gas engines. With this energy recovery, it becomes possible to cover at least a part of the thermal and electrical energy consumption of the wastewater treatment.

On the other hand, the use of anaerobic digestion requires large-volume digester systems that must be reliably sealed from the atmosphere. In addition, an extensive plant system consisting of pumps, pipelines, and control systems is required for the precise supply of substrates, heating, and mixing of the system.

Finally, safe storage and utilization of the produced gas by combined heat and power units have to be implemented.

Due to the high construction costs of the digestion plant, anaerobic digestion has not yet been installed in much smaller WWTP, especially with a capacity of less than 50,000 population equivalent (PE), and the nutrient-rich activated sludge is used in agriculture as fertilizer on the fields after aerobic stabilization [9].

As a result of rising quality demands and efforts to reduce the concentration of heavy metals and organic compounds in the environment, agricultural use in Germany has been increasingly restricted. The percentage of agricultural use of sewage sludge, therefore, decreased from 45% to 28% between 2012 and 2017 [10]. Because the landfilling of untreated waste was banned simultaneously, the incineration of sewage sludge has become more important. With prior anaerobic treatment of the sludge, the dewaterability can be increased and incineration can be achieved with a heat surplus and without an additional and cost-intensive back-up firing with fossil fuels. For this reason, almost exclusively anaerobically pre-treated sewage sludge is incinerated, which further increases the relevance of the process and the need for appropriate capacities [9]. Due to the immense construction costs of a digester, smaller sewage treatment plants without anaerobic stabilization must now transport the resulting sludge quantities to plants with existing digesters.

The capacity of existing digesters to utilize the amount of sludge supplied depends on the volume of the tanks. The operation is usually carried out in a single stage tank with constant filling level, so that added sludge quantities always lead to a discharge of the digested sludge in the tank.

The hydraulic retention time (HRT) results from the usable tank volume, and the daily exchanged sludge quantity. Assuming an infinite residence time, the anaerobic digestion could be optimally conducted, and the full potential of residue reduction and methane gas production could be realized. Due to the high construction costs of the digestion capacities, additional excess sludge volume caused by reduced aerobic stabilization and agricultural use reduces the HRT in the existing digesters.

Another factor for a reduction in HRT can be a local increase in sludge generation due to ongoing urbanization and corresponding local population growth [11]. Owing to the limited conversion rate of the hydrolysis during anaerobic digestion, biomass that has not been completely converted is also discharged from the digesters, which means that the potential for methane gas production and residual sludge reduction cannot be fully utilized [12].

As an alternative to expanding digestion capacities, there is a considerable incentive to accelerate the digestion process, especially the rate-limiting hydrolysis, in the existing plants. This can be achieved by the disintegration of sludge particles, which are thus made immediately accessible for the fermentation process [13,14]. Hence, the methane yield can be increased, and the amount of biosolids in the digested sludge (DS) that need to be disposed of can be reduced due to both an improved digestibility and a better dewaterability [15–17].

1.2 Sludge disintegration methods

Disintegration can be carried out thermally, chemically, biologically, or mechanically, whereas each method provides specific advantages but also includes limitations regarding to the full-scale application [18,19]. The main challenge for the deployment of each pre-treatment method is the reduction of energy consumption and the search for pathways to reduce the need for process equipment and substances. Also, the combination of these mechanisms has been discussed to increase the disintegration efficiency and was investigated with the demonstration of synergetic effects [20–22].

1.2.3 Thermal disintegration

Thermal disintegration is conducted by the heating of the sludge to temperatures of up to 165 °C and revealed to be effective in increasing the gas yield and improve the dewaterability of the sludge [23]. Although this process allows for the reuse of excess heat emitted by combined heat and power units of the WWTP, the energy demand and the required process engineering equipment provide a challenge for an economically feasible operation [24,25].

Microwave irradiation allows for rapid and selective heating of sludge particles and has resulted in an increase in gas yield and dewaterability [26,27]. In addition, a higher increase in gas yield was observed from microwave irradiation compared to heating in a water bath with the same specific energy input [28].

1.2.4 Chemical disintegration

One approach to achieve chemical disintegration is the use of strong alkaline solutions such as NaOH, KOH, $Ca(OH)_2$ [29]. Depending on the concentration, a very effective disintegration, and an increase in soluble chemical oxygen demand (COD) can be achieved [20,30]. However, the benefits of increased gas yield are hindered by the high cost of chemicals, which could not be covered by the additional yield so far [31].

1.2.5 Biological disintegration

In the biological degradation process, enzymes such as glucosidases, lipases, and proteases are generated by microorganisms and result in the hydrolysis of polymeric substances and particles [32]. In order to accelerate the hydrolysis, as the rate-limiting step of anaerobic digestion, the presence of the enzymes can be increased by direct supplementation [33]. Although positive effects on biodegradation have already been observed by the addition of a mixture of industrial enzymes, there is still a substantial need for research and improvement in this field [34,35].

1.2.6 Mechanical disintegration

The disintegration by means of mechanical energy, however, represents a broad spectrum of processes in which mostly simple mechanical systems such as ball mills, high-speed cutter mills, or comparable devices have already been successfully applied [36,37]. With the use of a high level of energy, the gas yield could be increased by these processes, but the extensive mechanical wear and tear are a considerable cost factor in addition to the energy requirement [38,39].

Sludge disintegration with ultrasound, on the other hand, has been identified as a very effective way of mechanically pre-treating substrate [37,40–44]. Using ultrasound, very powerful cavitation bubbles, and thus high shear forces can be generated in the feed solution, which efficiently disaggregates sludge flocs and ruptures microbial cells

[15,45,46]. This results in a release of intracellular enzymes and organic matter and, consequently, an acceleration of the anaerobic digestion process [47–51].

Ultrasound-induced cavitation is always generated by an oscillating device, but the variety of transducer principles ranges from highly-focused horn-shaped sonotrodes to flat transducers with a broad cavitation field. Whereas horn-shaped sonotrodes with a small sound-emitting transducer surface allow high amplitudes to be generated with a remarkable ability to corrode solid materials in laboratory routine applications, they produce a very limited cavitation field and high thermal losses within the piezo-ceramic transducer plates [52,53].

Flat transducers, characterized by a sound-emitting reactor wall, produce a larger cavitation field, but due to the vertical transmission of the oscillation power, smaller amplitudes compared to horn-shaped sonotrodes are reached. Previous studies with tubular, flat transducer systems have shown an increase in methane yield of up to 63% during laboratory-scale experiments [51].

However, this remarkable increase at laboratory-scale was achieved with a comparatively high energy input of 5,443 kJ per kg of total solids (TS), which would lead to extensive power requirements in full-scale applications. A less intensive sonication with only 327 kJ/kg_{TS} showed a lower increase in methane yield of only 20% [13]. However, 50% more energy was obtained than previously invested for ultrasound generation in this case. Thus, the use of flat transducer systems seems to be a promising approach to the sonication of large quantities of sludge, but the efficiency of the ultrasonic system requires further improvement.

1.3 Generation and use of cavitation

Ultrasound with a power density of at least 0.1 W/cm^2 leads to the formation of cavitation bubbles in water, which have the ability to disintegrate solid materials such as sludge particles. In this process, the water is subjected to a pressure-tension change by a resonant oscillation system, which leads to the formation of negative local pressures (Figure 2). This results in the formation of very fine cavities in the water, which can grow up to cavitation bubbles over several pressure-tension cycles.

Above a critical size, the cavitation bubbles implode intensively with high pressure beyond 1.3 GPa [54] and temperature peaks of more than 10,000 K [55,56]. The microjets resulting from the implosion of a cavitation bubble lead to high shear forces at the boundary layer, which lead in turn to the desired mechanical effect of the sonication.

The formation of cavitation bubbles is enhanced at cavitation nuclei, which can consist of dirt particles or air bubbles. The properties of cavitation bubbles generated in a volume depend on a number of parameters. Firstly, the cavitation field is influenced by the design of the oscillating system, which is characterized by (i) the frequency, (ii) the power density, and (iii) the geometry of the sound emitting surface. In addition, the composition of the medium and the volume of the reactor to be sonicated have a significant influence on the result of the sonication.

While sound waves can propagate unhindered in water, reflection or absorption occurs on solids depending on the density of the respective materials. In particular hollow bodies, like reactor housing, lead to reflection of the sound waves and, therefore, the concentration of the mechanical energy applied. This effect can be utilized to increase the cavitation field by limiting the reactor volume to a defined size when the energy input is relatively high. The reflection at the reactor wall also favors the development of a homogeneous cavitation field. In contrast to the increase of cavitation intensity through an optimized reactor design, the sonication of sewage sludge is subjected to a reduction in cavitation intensity due to the damping effect of the very fine sludge particles.

The mechanical energy supplied in the medium is converted into heat by friction and is no longer available to the process for the generation of cavitation bubbles [57]. To achieve an intensive cavitation field, therefore, the solids content must be as low as possible and the power density as high as possible.

This combination, however, leads to high relative energy consumption as well as wear and tear of the oscillation systems. Therefore the minimum required power density to achieve the desired disintegration result should always be the objective of the optimization of the sonication process.

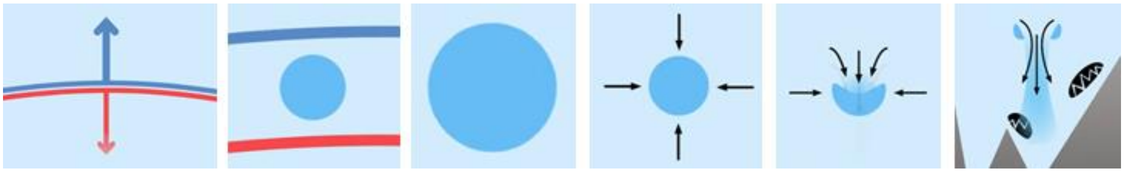

Figure 3: Generation of cavitation bubbles.

1.4 Generation of ultrasound

In order to generate high power ultrasound, electrical energy is converted into mechanical oscillation. In the past, a variety of physical conversion methods such as magnetostrictives, flat oscillators, and piezoelectric systems have been used in commercial ultrasound systems [58,59]. However, in the field of technical ultrasound, the use of pre-tensioned piezoceramic oscillation systems has been favored due to their highest efficiency in terms of converting electrical energy into mechanical energy [60].

To create a piezoceramic element, mostly a ceramic disc consisting of lead zirconate titanate is subjected to polymerization. This material belongs to the class of synthetic materials called ferroelectrics, with much higher piezoelectric potential than natural materials as tourmaline or quartz [61]. The synthetic material composites crystallize in the perovskite crystal structure.

As soon as the temperature falls below the piezoelectric Curie temperature, distortions of the ideal perovskite structure result in a dipole moment. Due to the sintering process used in the production of industrial ceramics, the dipoles are still disordered in their initial state. Therefore, areas with elementary cells of uniform dipole direction compensate each other. To generate the piezoceramic potential, the arbitrary orientation of the domains is subjected to an attached very high direct electric field (e.g. 2 kV/mm), and the dipoles are consequently aligned in the field (Figure 3) [27].

This process is performed with simultaneous heating of the material to just below the piezoelectric Curie temperature. After the removal of the external electric field and with the decrease in temperature, the dipoles mostly remain in their aligned position.

The polymerization process is thus completed and the piezoelectric ceramics can be further processed for the manufacturing of oscillation systems.

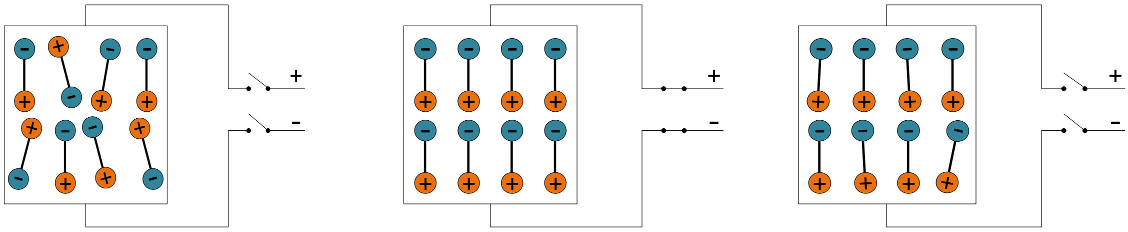

Figure 4: Polarization of ceramic disks with electric field.

Common oscillating systems for process engineering applications consist of piezoceramic hole discs with a metallic oscillating body made of aluminum or titanium that are firmly tightened with a bolt (Figure 4). This assembly enables the generation of an electric field to influence the dipoles within the piezo element, which can be improved by means of metal contact discs mounted directly on the surface of the piezoceramic element. When an external electric current is applied, the polarized material is deformed. Applying the polarity according to the previously conducted polarization causes the expansion of the piezoelectric element (Figure 4). Changing the polarity of the applied voltage causes the piezoelectric element to contract. This effect can be utilized for the transformation of electrical power to a mechanical oscillation.

For this purpose, an alternating electrical current is applied to the ceramic discs, which leads to an alternating expansion and contraction of the piezoceramic elements. The mechanical agitation can be transferred to other constructional elements, and the entire structure can be arranged as a resonant oscillation system. Due to the firm connection of the individual components and a suitable pretension of the piezoceramic discs, high forces can be emitted by the oscillation system.

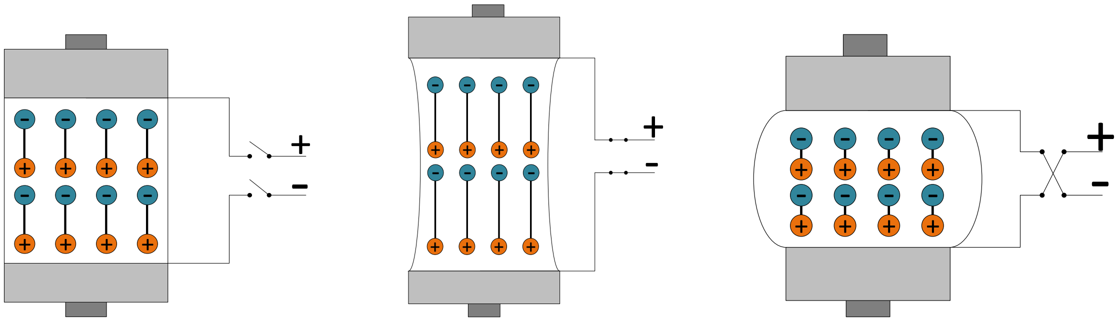

Figure 5: Piezoelectric effect of expanding disks with applied current.

1.5 Ultrasonic frequency

The alternating current employed to excite the oscillation system essentially determines the properties and the oscillation behavior of the entire setup. To generate ultrasonically induced cavitation, oscillation systems have to be supplied with a frequency in the range from 20 kHz to 1 MHz. The needed alternating current is converted by means of an ultrasonic generator from the mains frequency of 50 Hz as used in Europe. The predefined resonance frequency or ultrasonic frequency is given by the dimensions of the oscillation system and can be calculated in principle, as shown in the formula [1] and [2].

$$\boldsymbol{\lambda} = \frac{v}{f} \qquad [1]$$

$$\frac{\boldsymbol{\lambda}}{\mathbf{2}} = \frac{6{,}250\ \mathrm{m/s}}{2 * 25000\ Hz} = 12.5\ cm \qquad [2]$$

To achieve a resonant frequency of f = 25 kHz, the oscillation system with an exemplary sound velocity v of 6,250 m/s in aluminum, the length of the resonator must provide a multiple of the half wavelength λ. Depending on the bandwidth of the vibration system, a fine adjustment of the frequency may be necessary by measuring the resonance point in addition to the initial design. This need for adjustment is mostly caused by production-related material tolerances and the combination of different materials.

Owing to the design dependence of the resonance point, the frequency of the sonication is not a variable after manufacturing. Nevertheless, it is a parameter of the design and optimization of the treatment process because of its decisive influence on the cavitation field that is being generated. Due to the declining wavelength with increasing frequencies, the cavitation bubbles also decrease in size with increasing frequency. With the generation of ultrasound in a low frequency range of 20-25 kHz, larger cavitation bubbles with high shear forces can be generated [62]. Therefore this frequency range has proven to be advantageous for the mechanical disruption of sample material in laboratory applications and, in particular, for the disintegration of sewage sludge to release the organic matter and increase the gas yield [25].

On the other hand, higher frequencies lead to the formation of a large number of very fine cavitation bubbles. These more uniform cavitation fields are particularly suitable for ultrasonic cleaning in a range of 35 to 40 kHz, as they can reach the finest roughness on surfaces and effectively remove contaminants. Even higher ultrasonic frequencies can be applied to perform or accelerate advanced oxidation processes (AOP). In these sonochemistry applications, only low shear forces are required, and a high number of fine cavitation bubbles ensures a high conversion rate [63].

Nevertheless, even at low frequencies in the range of 25-28 kHz, always smaller cavitation bubbles are generated. These are caused by the implosion in an early growth stage as well as by the implosion and fragmenting of large cavitation bubbles, which in turn lead to smaller imploding cavitation bubbles [62].

1.6 Geometry of the sound emitting surface

In order to transfer the resonant oscillation into a liquid medium, the sound emitting surface must be placed in direct contact with the medium. For this purpose, the resonant system can be designed in such a way that the vessel wall and the circular transducer cone form a resonant oscillating system (Figure 6). The ultrasonic oscillation is transmitted directly and evenly from the sound emitting surface into the medium. By combining several oscillation systems at uniform intervals, a scaling of the ultrasonic power and nearly unlimited size of the sonicated chamber can be achieved.

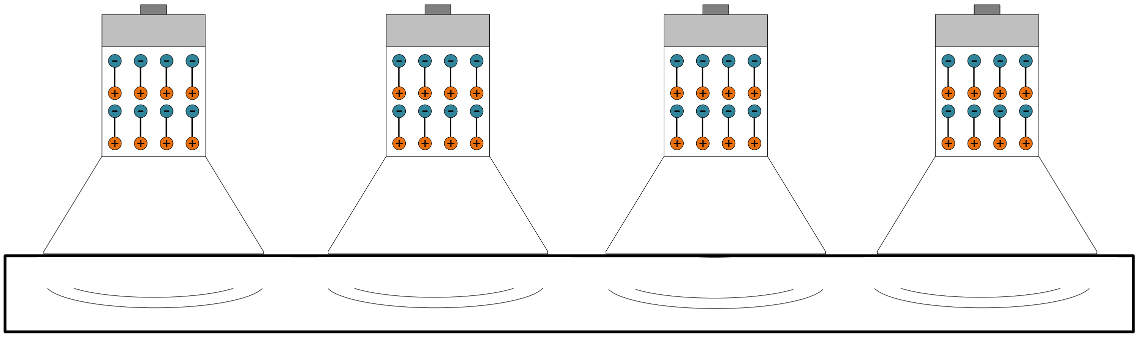

Figure 6: Schematic array of flat transducers.

Due to the broad distribution of the applied energy over a large number of oscillation systems, the mechanical stress on the individual oscillation systems is relatively low. This enables the use of cost-effective materials such as aluminum and steel for the manufacturing of the multiple oscillation systems.

The broadband characteristics of the resonance frequency resulting from the material composition and design enable a large number of oscillation systems to be connected in parallel and supplied with electrical energy from a common ultrasonic generator. With this design, the generated heat of the piezoceramics can also dissipate over a comparatively large surface area.

A further possibility for introducing the resonant oscillation into the medium to be sonicated is the transmission by means of a rod-shaped sonotrode, which is immersed in the medium (Figure 7). With this setup, the ultrasonic power is transmitted into the liquid at all surfaces perpendicular to the sound propagation. The length of the rod is defined by a multiple of half the wavelength λ.

In order to increase the power output without further increasing the material consumption for the implementation of additional probes, numerous piezoceramic discs are connected in series in the transducer head. This design requires the use of cost-intensive titanium to reduce mechanical losses at the generated high power densities. As a result of this setup, the frequency range for resonant excitation of the oscillating system is comparatively narrow, which requires the use of a highly adjusted ultrasonic generator module for each transducer system. In addition, it is necessary to ensure a defined dissipation of the heat due to the higher thermal losses [64].

This can be realized by a controlled airflow or by liquid cooling systems. Overall, the above-mentioned characteristics of rod-shaped transducer systems lead to an increase in material and production costs, which is reflected in investment costs that are up to twice as high as for flat transducer systems with the same power input.

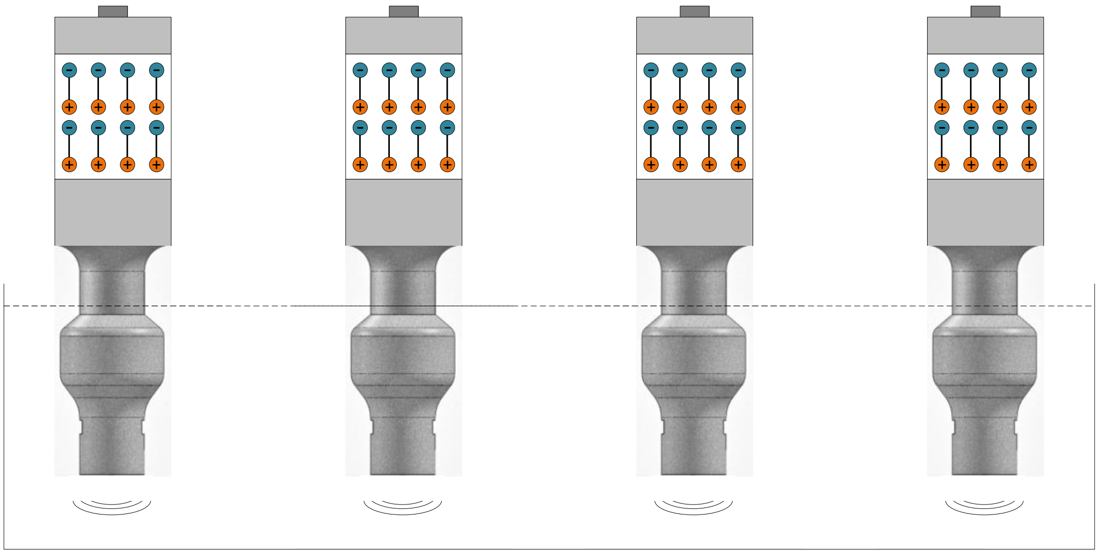

Figure 7: Schematic array of sonotrode transducers.

For both processes presented, a wide variety of possible geometries can be applied. These include rod-shaped oscillation systems with different lengths and diameters depending on the sonicated reactor volume and intended degree of disintegration. These systems can be integrated into any reactor design by means of a flange. Thus, the power output is scalable to the intended sonication process by increasing the number of oscillation systems and the reactor dimensions.

Also, the shape of surface oscillation systems, which involve the reactor wall in the oscillation, can be produced with a flat sound emitting surface as well as in a tubular design. The advantage of a tubular design is the insensitivity to disturbing substances and possible clogging. This property offers particular advantages in the design of reliable sonication systems for the sonication of sewage sludge in full-scale applications, but there are also further aspects that should be considered when designing suitable sonication systems for a specific task.

1.7 Power density and amplitude of the sound emitting surface

The cavitation field generated in a liquid by means of ultrasound can only be partially described by specifying the electrical energy applied for excitation of the oscillation systems and the reactor volume. Another relevant parameter for the magnitude and intensity of the cavitation field generated is the power density with regard to the sound emitting surface. The power density P_D is calculated from the dimensions of the sound-emitting area A in W/cm² and the electrical power P_{US} received by the ultrasonic system on the mains side. The power density is specified in W/cm² as follows:

$$P_D = \frac{P_{US}}{A} \qquad [3]$$

With an increase of power density, the amplitude of the oscillation at the sound emitting surface increases. For example, the transmission of the ultrasonic power of 400 W from an original transducer diameter of 50 mm to a tip diameter of 4 mm leads to amplitudes of 300 µm. Due to the strong focusing of the ultrasonic power, an intensive cavitation field with high shear forces is generated.

However, the disadvantage of a high transformation is the very small spatial propagation of the cavitation field and thus the low capacity of the system to sonicate larger volumes. With a lower focusing of the power on a larger area and a resulting amplitude of 6-10 µm, a larger cavitation field can be generated in a reactor volume, and more particles in the solution can be sonicated simultaneously.

A further parameter influencing the intensity of the cavitation field generated is the design of the reaction chamber. Spatial limitation of the propagation leads to the reflection of the ultrasonic waves and an increase in the ultrasonic intensity in the sonicated volume. The design of an optimized sonication system depends, therefore, on the composition of the medium to be sonicated and the desired degree of disintegration.

The power density should be reduced as much as possible in order to (i) keep the energy requirement per dissolved particle as low as possible and (ii) to keep the erosion on the sound emitting surface resulting from cavitation as low as possible for economic reasons. In the present work, the lowest possible intensity of the cavitation field that needs to be generated to disintegrate the given sewage sludge was investigated. This involved several ultrasonic devices consisting of flat surface transducer systems with different reactor sizes and a sonotrode. Furthermore, different sludge compositions were included in the investigation to examine to what extent the power density affects the sonication result.

1.8 Specific energy input per total solids

Due to the seasonally variable operating conditions of the WWTP, caused by seasons, dry or wet weather, and plant operating settings, the composition of the WAS and DS varies in terms of the contained solids content. To compare the effectiveness of different sonication methods with sludge from different sample times and even plant origins, the energy input should be related to the dry substance content of the medium. For this purpose, the amount of energy introduced E_{input} is divided by the mass of dry matter $M_{total\ solids}$ contained in the sonicated substrate volume and expressed in kJ/kg_{TS}:

$$E_{spez} = \frac{E_{input}}{M_{total\ solids}} \qquad [4]$$

The specific energy input can be enhanced by increasing the power density, the sound-emitting area as well as the retention time of the sludge in the cavitation field. However, the value does not provide any information about the intensity of the cavitation field used, since even low-intensity cavitation fields can achieve a high specific energy input with a relatively high retention time.

For the consideration of an ultrasonic system for application in full-scale plants, however, the specific energy input is the most important parameter, because it includes information on the energy efficiency of the respective sonication configuration.

For this reason, this parameter is used in the present study as the characteristic indicator of the energy input generated by the sonication system.

1.9 Application of ultrasonic cavitation for sludge disintegration

In order to achieve the disintegration of sewage sludge, a wide variety of ultrasonic reactor designs have been considered in previous studies (Figure 8) [65,66]. These include reactor configurations in which the substrate to be sonicated is passed through a meandering reactor [67]. The aim of this design is to ensure that as much of the sludge as possible is conveyed along the sound-emitting surface of the sonotrodes installed in the opposite direction to the flow. By limiting the sonication volume, a reflection of the sound and thus an intensive cavitation field is achieved [68]. The disadvantage of this design is the non-uniform flow through the reactor chamber, which can lead to deposits and blockages during continuous operation [69]. For this reason, regular flushing of the sonotrodes must be carried out with this design.

Another system for the sonication of WAS was realized by means of a large batch reactor [69]. Here, the sludge is pumped into a tank with a capacity of about 1,000 liters. The rod-shaped ultrasonic transducers emit the ultrasonic power laterally to the medium. For improved mixing and uniform exposure of the sludge in the ultrasonic field, continuous stirring is performed by means of a central agitator. The sonicated excess sludge is finally returned to the main transport line.

A relatively new approach to the sonication of WAS and DS is the use of a tubular reactor in a flow-through process [51]. Deviating from conventional oscillation systems, this involves the use of surface oscillation systems attached to the outside of the tube. This offers the great advantage that the risk of clogging is comparable to that of a simple pipe. Although laboratory tests have shown a good disintegration effect of surface oscillation systems, further information was required on the effectiveness of this sonication method compared to other reactor designs [70]. In order to achieve the objective of the highest possible disintegration effect at the lowest possible cost, further possibilities for improving the efficiency of the systems were to be sought within the scope of this study.

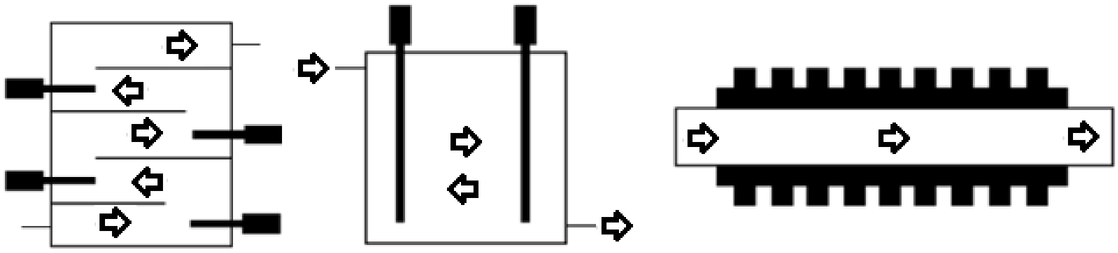

Figure 8: Reactor concepts consisting of sonotrode reactor (left), batch reactor (middle), and tube reactor (right).

The first approach for the optimization of existing ultrasonic systems for the ultrasonic treatment of aqueous media was the analysis of the damping of the cavitation intensity by the solids-containing substrate. The findings of the penetration depth of the sound field into the cross-section of the flow provided information about an improved reactor shape and supported the development of efficient reactor designs.

These investigations also revealed that the dry matter content has a significant influence on the formation of cavitation fields and that a reduced dry matter content has a positive effect on the propagation of the cavitation bubbles generated by the ultrasound. For this reason, DS was also subjected to sonication in addition to the pre-treatment of WAS. The advantage of sonication of DS is the reduced TS, which necessitates a significantly lower energy input for the generation of an intensive cavitation field. An additional advantage of sonication of DS is that only substances that are less available for the anaerobic process receive a cavitation treatment. Hence, two potential application scenarios for the sonication of anaerobic sludge at an industrial scale result from the sonication configurations can be considered (Figure 9).

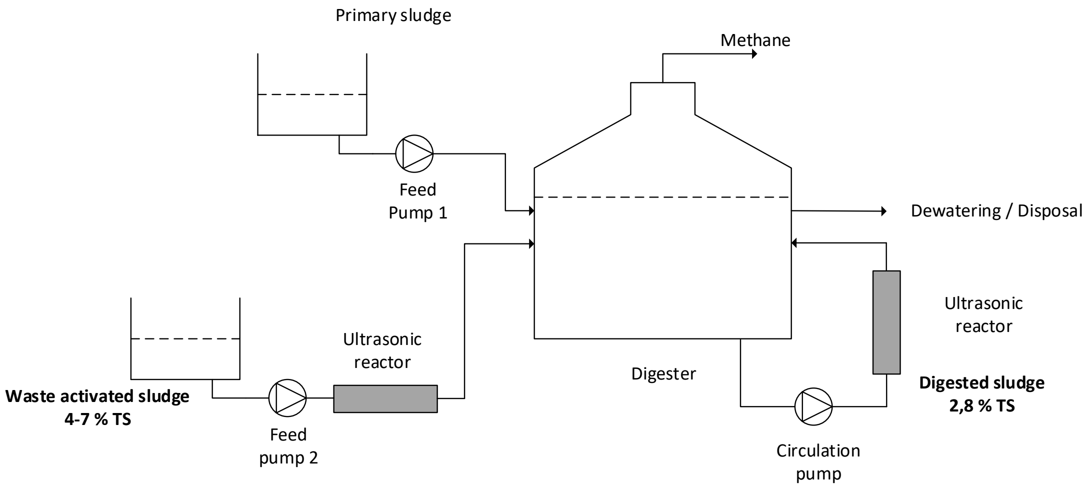

Figure 9: Potential installation locations of ultrasonic reactors.

The influence of static pressure on the cavitation intensity showed an increased effect in experiments with water [71] but has not been investigated in WAS or DS yet. In the second step, various reactor concepts were designed, simulated, and tested on the basis of the framework conditions determined.

When full-scale ultrasonic reactors were used to treat large quantities of sewage sludge, the volume flow was limited by the size of the reactors. Experiments with horn-shaped sonotrodes showed a weakening of the cavitation field during the admission of a perpendicular flow into the sound field [72]. In the context of the investigation of different reactor designs, the influence of different flow velocities was therefore considered.

The lifetime of the ultrasonic system to be designed plays an essential role in a holistic evaluation of an ultrasonic system with regard to economic and ecological effects. The lifetime of an ultrasonic system is limited by the wear of the sound emitting surface in the medium. Intensive cavitation bubbles lead to an erosion of the stainless steel surface. Over a long period of operation, this results in the formation of very fine pores and the leakage of the entire reactor. The extent of wear and tear is, therefore, also an aspect to be investigated in the design of ultrasonic systems.

2 Thesis objectives

2.1 Research significance

With the intention of improving anaerobic digestion in WWTPs, a number of investigations on the utilization of ultrasonic disintegration has been carried out in the past decades [13,15,48,68]. A large number of studies could reveal an increase in gas production after sonication using laboratory equipment, both of added excess sludge and digested sludge [25,51,73].

The positive effects motivated the transfer of the experimental approaches to full-scale installations, but the effects shown in the laboratory could not be fully demonstrated on an industrial-scale so far. Especially the large volumes of digesters in the range of 2,500-4,000 m^3 hampered the technical transferability of the effects observed in the laboratory to full-scale [74].

In the development of suitable ultrasound systems, the (i) costs of the ultrasound systems, (ii) a sufficient intensity, and (iii) a sufficient spatial extension of the cavitation fields used have so far been partially opposed to each other. In order to develop a qualified process sheme, it is therefore of great interest to examine both the fundamental influences of the substrate to be sonicated on the formation of cavitation fields and the effectiveness of cavitation fields of varying intensity on the sonication result. In order to investigate this question, a series of experiments were performed in this study, focusing on the respective aspects.

The observations obtained served to adjust existing assumptions for the design of ultrasonic reactors and were directly incorporated into the design of two full-scale test facilities. The questions considered relate to basic mechanisms for the generation of acoustic cavitation in media containing solids and provide the potential to transfer the results to further process engineering applications.

2.2 Research questions and hypotheses

Paper #1: "Cavitation field analysis for an increased efficiency of ultrasonic sludge pre-treatment using a novel hydrophone system."

Research Question #1: In which way does the solid content influence the cavitation intensity in a treated substrate? Can an optimized reactor shape and optimized operation conditions increase the cavitation intensity in the treatment of sludge?

Hypothesis #1: 'The cavitation intensity in an ultrasonic reactor is reduced by the soundwave attenuation of particles but can be increased by an optimized reactor shape and by the increase in static pressure.'

Paper #2: "Assessment of sonotrode and tube reactors for ultrasonic pre-treatment of two different sewage sludge types."

Research Question #2: In which way does the sonication technology influence the pre-treatment efficiency of WWTP sludge?

Hypothesis #2: 'The influence of the ultrasonic pre-treatment method on the degree of disintegration, gas increase, decrease in viscosity and cavitation intensity during pre-treatment of WWTP sludge with the same energy input is higher than 10%.'

Paper #3: "Impact of high flow rates and increased viscosity of digested sewage sludge on the cavitation intensity in ultrasonic tube reactors."

Research Question #3: Is there a limit concerning the flow rate in tube reactors during ultrasonic pre-treatment of WWTP sludge?

Hypothesis #3: 'An average flow velocity of more than 0.3 m/s suppresses the cavitation field during sludge treatment with ultrasonic tube reactors.'

2.3 Thesis structure

Table 1: Thesis structure

Chapter	Research questions	Hypothesis	Publication
3.1	In which way does the solid content influence the cavitation intensity in a treated substrate? Can an optimized reactor shape and optimized operation conditions increase the cavitation intensity in the treatment of sludge?	**Hypothesis #1:** 'The cavitation intensity in an ultrasonic reactor is reduced by the soundwave attenuation of particles but can be increased by an optimized reactor shape and by the increase in static pressure.'	**Paper #1:** Bandelin et al.: Ultrasonics Sonochemistry 42, 2018, 672-678
3.2	In which way does the sonication technology influence the pre-treatment efficiency of WTTP sludge?	**Hypothesis #2:** 'The influence of the ultrasonic pre-treatment method on the degree of disintegration, gas increase, decrease in viscosity and cavitation intensity during pre-treatment of WTTP sludge with the same energy input is higher than 10%.'	**Paper #2:** Bandelin et al.: Ultrasonics Sonochemistry 64, 2020, 105001
3.3	Is there a limit concerning the flow rate in tube reactors during ultrasonic pre-treatment of WTTP sludge?	**Hypothesis #3:** 'An average flow velocity of more than 0.3 m/s suppresses the cavitation field during sludge treatment with ultrasonic tube reactors.'	**Paper #3:** Bandelin et al.: Chemical Engineering & Processing: Process Intensification 152, 2020, 107925

3 Publications

3.1 Paper #1 Cavitation field analysis for an increased efficiency of ultrasonic sludge pre-treatment using a novel hydrophone system.

With the aim of improving the disintegration performance of flatbed reactors, in particular with the sonication of particles containing substrates such as sewage sludge, several variations in the reactor shape and operation conditions were utilized to test **Hypothesis #1:**

The cavitation intensity in an ultrasonic reactor is reduced by the soundwave attenuation of particles but can be increased by an optimized reactor shape and by the increase in static pressure.

Three different flatbed reactors with different gap sizes have been set up in order to measure the cavitation intensity inside the reactor during operation with three different substrates. Water was used as a particle-free reference, while digested sludge, as well as waste activated sludge, represented the particle-containing suspensions with different solids contents. A screw pump and a pressure sensor coupled with a ball valve allowed for the adjustment of the static pressure inside the reactor chamber. The results presented in **Paper I** demonstrate a nearly complete suppression in cavitation intensity with an increasing solids content in large reactors of 80 mm but indicate a reduction of this effect in a smaller reactor of 40 mm, reaching at least 30% of the intensity observed in water.

With the adjustment of the static pressure to 1 bar, the cavitation intensity was enhanced, but a further increase to 2 bar resulted in a reduction of cavitation intensity. Thus the **hypothesis #1 can be accepted with the limitation** of an optimum pressure increase below 2 bars.

The results indicate a relevant impact of the gap size on the cavitation intensity in the ultrasonic reactors. This limitation hedges the increase in the volume of possible reactor shapes in the full-scale application. A parallel or series connection of small size reactor increments can be a suitable response to this observation.

The positive influence of an increased pressure suggests the design of a pressurized reactor setup without any sonication vessels or gutters at ambient pressure.

The following text was published with editorial changes as:

Bandelin, J.; Lippert, T.; Drewes, J. E.; Koch, K.: Cavitation field analysis for an increased efficiency of ultrasonic sludge pre-treatment using a novel hydrophone system. Ultrasonics Sonochemistry 42, 2018, 672-678

Author contribution: **Bandelin, J. (80%)**; Lippert, T. (10%); Drewes, J. E. (5%); Koch, K. (5%)

3.1.1 Abstract

The generation of cavitation fields for the pre-treatment of anaerobic sludge was studied by means of a novel acoustic measuring system. The influence of different reactor dimensions (i.e., choosing reaction chamber widths of 40, 60 and 80 mm) on the cavitation intensity was determined at various solid contents, flow rates and static pressures. Results suggest that the cavitation intensity is significantly reduced by the sonication of liquids with a high solid content. By increasing the pressure to 1 bar, the intensity of bubble implosions can be enhanced and the sound attenuation in the solid fraction is partly compensated compared to ambient pressure. However, a further increase in pressure to 2 bars has a detrimental effect due to the suppression of powerful bubbles. A reduction of the reactor gap permits an intensification of the treatment of waste activated sludge (WAS) by concentrating the ultrasound power from 6 to 18 dB. This effect is less relevant in digested sludge (DS) with its markedly lower total solids content (2.2% vs. 6.9 % of solids in WAS). Increasing the flow rate, resulting in a flow velocity of up to 7 m/min, has no influence on the cavitation intensity. By adapting the reactor design and the static pressure to the substrate characteristics, the intensity of the sonication can be notably improved. This allows the design of sonication devices suitable for the intensive treatment of wastewater sludge.

3.1.2 Keywords

Ultrasonic sludge pre-treatment, pressurized cavitation, flat-bed reactor, hydrophone tests, cavitation intensity

3.1.3 Introduction

Due to the current debate on how to increase the generation of renewable energy without any negative effects on the environment, recent studies have mainly focused on energy recovery from waste streams [75–77]. Apart from the construction of new biogas plants, the optimization of existing anaerobic systems offers a great potential to increase energy recovery, especially in wastewater treatment plants (WWTP). This can be achieved by the disintegration of sludge particles, which are thus made immediately accessible for the fermentation process [13,14]. Hence, the methane yield can be increased, and the amount of biosolids that need to be disposed of can be reduced [15–17]. Disintegration can be carried out thermally, chemically, or mechanically, whereas disintegration with ultrasound has been identified as a very effective way of mechanically pre-treating the substrate [37,40–43]. Using ultrasound, very powerful cavitation bubbles, and thus high shear forces can be generated in the substrate, which efficiently disaggregate sludge flocs and rupture microbial cells [15,45,46]. This results in a release of intracellular enzymes and organic matter and, consequently, an acceleration of the anaerobic digestion process [47–51].

Ultrasound-induced cavitation is always generated by an oscillating device, but the variety of transducer principles ranges from highly-focused; horn-shaped sonotrodes to flat transducers with a broad cavitation field. Whereas horn-shaped sonotrodes with a small sound-emitting transducer surface allow high amplitudes to be generated with a remarkable ability to corrode solid materials in laboratory routine, they produce a very limited cavitation field and high thermal losses within the piezo-ceramic transducer plates [52,53]. Flat transducers, characterized by a sound-emitting reactor wall, produce a larger cavitation field, but due to the vertical transmission of the oscillation power, smaller amplitudes compared to horn-shaped sonotrodes are reached.

Previous studies with tubular, flat transducer systems have shown an increase in methane yield of up to 63% during laboratory-scale experiments [51].

However, this remarkable increase in laboratory-scale was achieved with a comparatively high energy input of 5,443 kJ per kg of total solids (TS), which would require extensive power requirements in full-scale applications. A less intensive sonication with only 327 kJ/kg_{TS} showed a lower increase in methane yield of only 20%. However, 50% more energy was obtained than previously invested for ultrasound generation [13] in this case. Thus, the use of flat transducer systems seems to be a promising approach to the sonication of large quantities of sludge, but the efficiency of the ultrasonic system requires further improvement.

The treatment of liquids with a solid fraction results in an attenuation of the ultrasonic waves by the sludge particles and a reduction of the cavitation performance [78].

To compensate the negative effect of the absorption, the intensity of the ultrasonic field has to be increased. This can be achieved by reducing the reaction space, resulting in a reflection and concentration of the ultrasonic waves in the reactor [60].

The ultrasonic energy per volume can be increased by using a flat-bed reactor, which is comparable to a square tube. Unlike in a round tube (ratio between area and circumference is maximized in a circle), a large oscillating surface can affect a comparatively small sonicated volume with a high cavitation intensity. However, the gap between the sound-emitting surfaces cannot be reduced indefinitely, since otherwise the flow velocity would increase excessively at a given treatment throughput. A very large gap would in turn lead to an absorption of the ultrasound energy close to the pipe walls, and the substrate would not be uniformly sonicated.

In addition to the reactor design, the process parameters of static pressure and flow rate are of considerable importance for the occurrence of cavitation too [71,79]. Cavitation is generated by rupturing a liquid with an acoustic field. A cavitation bubble is thereby expanded in the phase with negative pressure. Due to the inertia of the liquid, the bubble is only slightly compressed in the positive pressure phase and continues to grow in the next phase with negative pressure. After around 4-12 acoustic cycles [71], the cavitation bubble implodes in the pressure phase under high pressure differentials, extreme temperatures, and sonoluminescence [56,80–82]. This creates a strong micro-jet that can destroy solid materials [81].

In addition, new cavitation nuclei are produced that start to grow and collapse, thus creating a cloud of cavitation bubbles [71]. Whereas an increased static pressure leads to an intense implosion of the cavitation bubbles, it also diminishes the size of the clusters of implied cavitation bubbles and reduces their ability to develop in the next acoustic cycle [71]. This means that a static pressure which is increased over ambient pressure only increases the measurable cavitation intensity to that pressure range where the ultrasonic power induced is too weak to generate powerful cavitation bubbles [83].

The increase in the static pressure, on the one hand, enhances the implosion power of the cavitation bubbles, but more ultrasound energy has to be available for the formation of the bubbles [71].

Le et al. [83] observed a greater increase in soluble COD and a decrease in particle size in the sludge by increasing the static pressure up to 10 bars after sonication with 7,000 kJ/kg_{TS}. The experiments were carried out in a batch system using a sonotrode with a significantly higher amplitude compared to flat transducers. Thus, these results cannot be transferred to the formation of cavitation fields in the novel approach using flat-bed reactors under pressure.

Previous studies using ultrasound flat transducers for the disintegration of sludge were only conducted at ambient pressure [13,51]. In order to examine the barely studied influence of static pressure on the cavitation intensity in a flat reaction space, the present study was also carried out at a static pressure of up to 2 bars, which is within the common range of WWTP operation since the infrastructure (pumps for feeding and recirculation) is commonly available for a typical digester.

Cavitation bubbles can be inhibited by turbulence at high flow rates when an ultrasound field is attached to a perpendicular liquid flow [79]. Therefore, a variety of flow rates were tested, resulting in a flow velocity of up to 7 m/min.

Using the novel hydrophone system enabled the innovative approach of an intensity-oriented reactor assessment in the present study. This was the first time that the cavitation intensity in a flat transducer system was investigated with different substrates and at different static pressures.

3.1.4 Materials and methods

3.1.4.1 Cavitation measurement instrument

The intensity of the cavitation bubbles was measured using a novel acoustic method that records the secondary sound frequencies of imploding cavitation bubbles. Conventional measuring devices only detect the sound pressure level of the entire acoustic spectrum, so that the applied ultrasonic power is predominantly recorded, while no statement can be made about the formation of cavitation bubbles [84–86]. This can lead to inaccurate conclusions about the cavitation intensity in the medium, especially in liquids with a solid fraction that attenuates the ultrasound.

The use of a RESON TC4034-1 hydrophone (RESON A/S, Slangerup, Denmark) in the present study allowed the separate detection of the sound pressure of the ultrasonic frequency (f_0) and the implosion noise of imploding cavitation bubbles. The applied software detected the sound peak pressure in a frequency range of $2.15 \cdot f_0$ to $2.35 \cdot f_0$ and determined the cavitation noise level in decibels (dB). The radial-sensitive hydrophone, with a diameter of 11 mm, was centered in the reactor chamber, and the medium was circulated during the measurement (Figure 10). For this purpose, a stainless steel stick was passed through the entire reactor and fixed to the tube connectors.

The hydrophone was mounted to the side of the stick, thus ensuring a defined distance from the sound-emitting surfaces and an unrestricted detection of the cavitation noise.

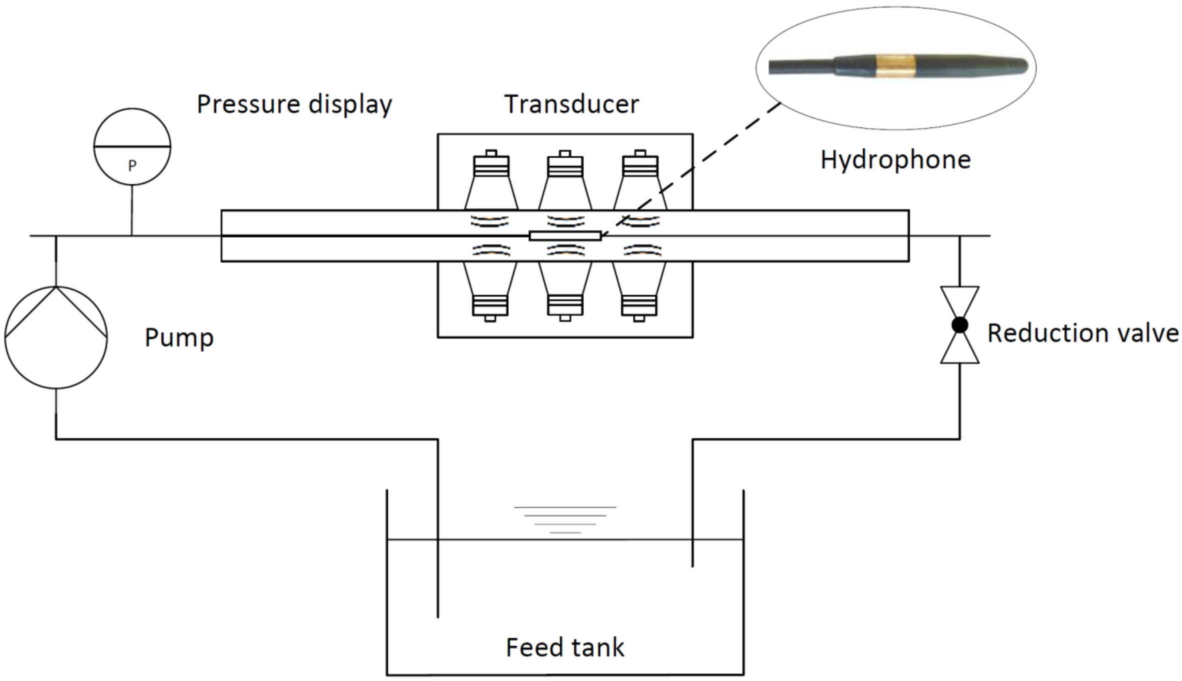

Figure 10: Flow chart of the test system.

3.1.4.2 Test configuration

The experiment was carried out by means of a circulation system, consisting of a feed tank from which the substrate was pumped through the test reactor by an eccentric screw pump (NETZSCH GmbH & Co. KG, Selb, Germany). A ball valve at the outlet and a pressure gauge at the inlet of the reactor allowed the pressure to be adjusted in the range of 0 to 2 bars at a constant volume flow. The flow rate was gradually increased by varying the speed of the pump between 0 and 1,400 L/h and was calibrated with a measuring beaker and a stopwatch.

3.1.4.3 Ultrasound source

The experiments were carried out in a custom-built ultrasound test system (BANDELIN electronic GmbH & Co. KG, Berlin, Germany) consisting of a rectangular flow cell and 6 diametrically arranged piezo-ceramic oscillators with a diameter of 50 mm, positioned at a distance of 20 mm from each other.

This design allowed the generation of a focussed sound field in the sonicated area with a specific intensity of 10 W/cm² and a total power of 300 W. The oscillators were covered to avoid any contact between the liquid and the electrical system. To guarantee a homogeneous flow, the overall length of the flow cell was increased to 1,000 mm and equipped with tube nozzles. A high-frequency generator of the type LG 1003 (BANDELIN electronic GmbH & Co. KG, Berlin, Germany) was responsible for the conversion to an ultrasound frequency of 25 kHz.

Three versions of the test system with different gap distances of 40, 60, and 80 mm were investigated to determine the influence of the gap dimensions on cavitation intensity (Figure 11). The mean value of the noise level was recorded over a period of 30 s. In order to consider a possible inhomogeneity in the sludge composition, the measurement was carried out in triplicate. To avoid any influence of the disintegration effect on the test results, the treated substrate was replaced after each cycle.

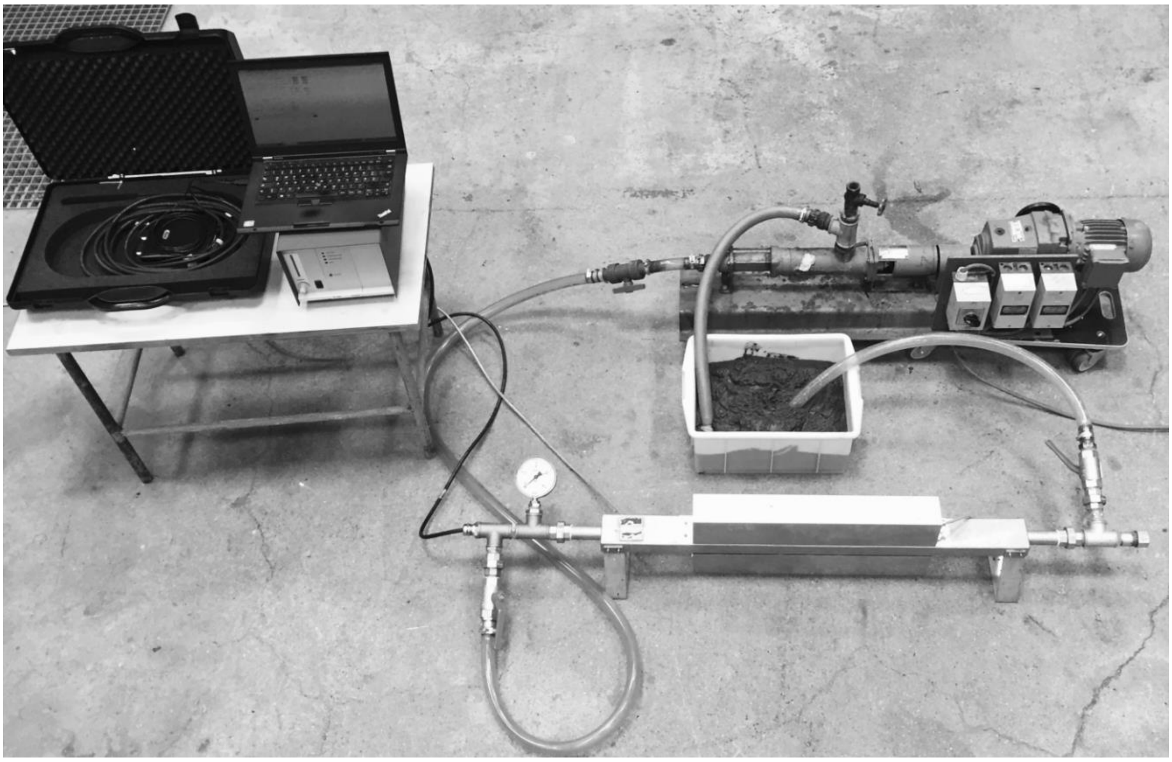

Figure 11: Test setup with flat-bed reactor and 40 mm reaction space.

3.1.4.4 Substrates

The key objective of this study was to determine the cavitation intensity in WWTP sludge. The cavitation noise level in water was taken as reference. To study the effect of sonication on the methane yield, either WAS (as the input medium of the digester with a comparatively high TS content) can be taken into consideration, or the already degraded DS from the circulation loop of the digester could be subjected to ultrasound treatment. In order to determine the influence of the sludge properties on the cavitation intensity, WAS with a TS of 6.9% as well as DS with a TS of 2.2% was investigated. The two sludge samples originated from the municipal WWTP Starnberg, Germany.

3.1.5 Results and discussion

The cavitation intensity was measured in 27 different test configurations, using three gap distances (20, 40, 80 mm) for the sonication of WAS, DS, and water at three different pressure levels (0, 1, 2 bar). Figure 11 summarizes the corresponding cavitation noise levels in the three different substrates, pressures, and reactor dimensions. The results indicate that the cavitation intensity decreases significantly with an increasing TS content. For example, the cavitation noise level is halved at ambient pressure with a gap of 40 mm from 24 dB in water to 12 dB in DS. An even greater drop occurs in the highly viscous WAS, reaching a cavitation intensity of only 7 dB under the same conditions. In WAS at ambient pressure and a gap of 60 and 80 mm, the hydrophone could not detect a valid cavitation noise level. This can be explained by the different rheological characteristics of the substrates. A higher TS of the sonicated medium has a greater attenuating effect on the transmission of sound waves, and the absorption of ultrasonic power by the solids in the substrate also reduces the intensity of cavitation [87]. Varying the flow rates in the range from 0 to 1,400 L/h, which was considered reasonable for a possible scale-up, resulted in a flow velocity of up to 7 m/min in the present experimental set-up. The flow velocity had no significant influence on the cavitation intensity at ambient pressure, except for WAS with a reaction space of 40 mm..

In this case, the flow resistance in the downstream reactor chamber led to a pressure increase in the sonicated area without the active reduction by the reduction valve. As a result, the WAS was sonicated with an increased pressure resulting in a higher cavitation intensity (Figure 12). The flow rate for further tests was kept constant at 600 L/h.

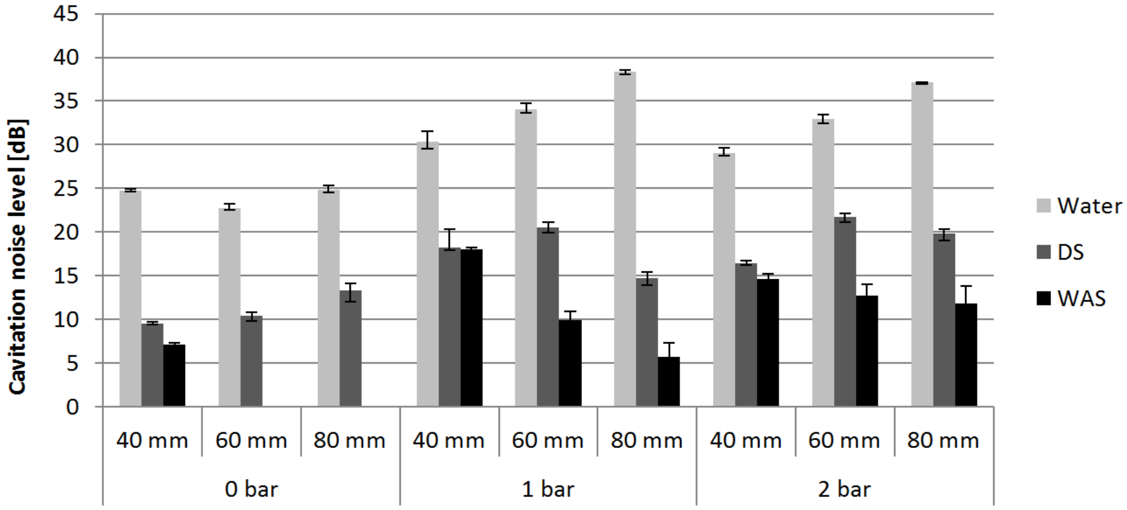

Figure 12: Comparison of cavitation noise levels in differing substrates, pressures and reactor dimensions.

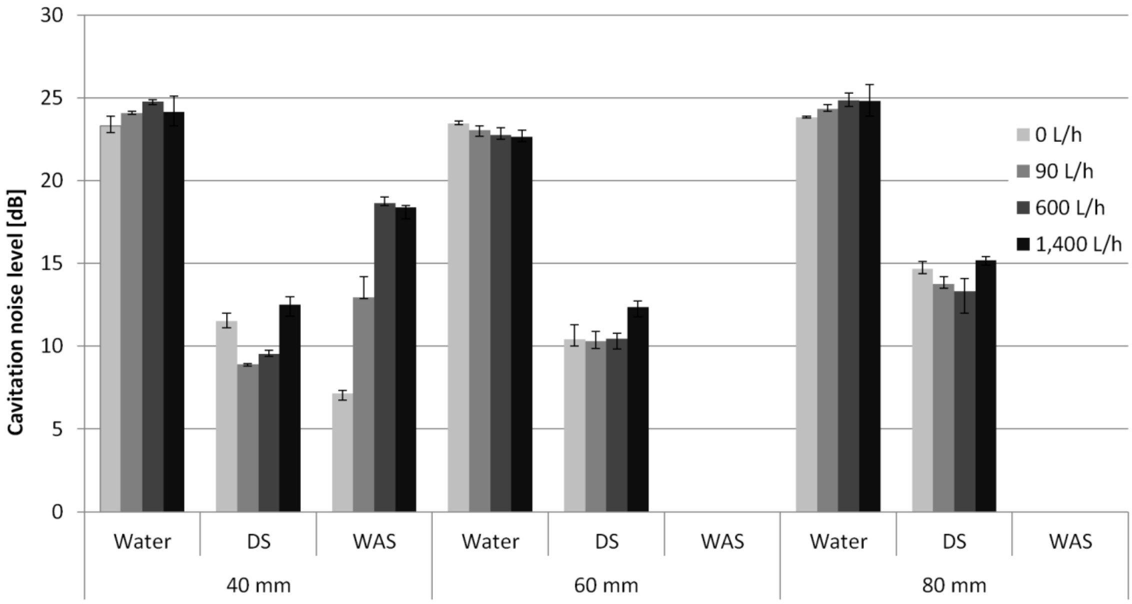

Figure 13: Cavitation noise intensity with differing flow rates at ambient pressure.

Table 2: Overview of cavitation noise level

Pressure	bar	0			1			2		
Gap size	mm	40	60	80	40	60	80	40	60	80
Cavitation noise level in water		24.8 ± 0.1	22.8 ± 0.3	24.9 ± 0.9	30.3 ± 0.5	34.1 ± 0.2	38.4± 0.4	29.1 ± 0.4	33.0 ± 0.4	37.1 ± 0.1
Cavitation noise level in DS	db	9.5 ± 0.2	10.4 ± 0.4	13.3 ± 0.9	18.2 ± 1.5	20.6 ± 0.4	14.7 ± 0.5	16.5 ± 0.2	21.8 ± 0.5	19.8 ± 0.6
Cavitation noise level in WAS		7.1 ± 0.3	0	0	18.0 ± 0.2	9.9 ± 0.8	5.8 ± 1.2	14.6 ± 0.5	12.7 ± 1.0	11.8 ± 1.4

In the present study, increasing the static pressure to 1 bar led to an increase in the cavitation noise level in all of the substrates under investigation. In addition to the measurements, an increase in the typical audible sound emission of the ultrasound device could be observed during the experiment. The increase of the cavitation intensity at a higher static pressure was particularly noticeable in the case of WAS with a comparatively high TS. Whereas a low cavitation level could only be measured in a narrow cavitation field of 40 mm at ambient pressure, the intensity could be more than tripled at a higher pressure of 1 bar (Table 2).

Apart from the more intense implosion of the bubbles, the significant influence of a pressure increase on the cavitation intensity in WAS can also be explained by the compression of gas bubbles enclosed in the sludge, and consequently a better transport of the ultrasonic waves into the centre of the substrate stream, or in this case, the measurement point of the hydrophone. A further increase in the static pressure to 2 bars led to a slight reduction of the cavitation intensity in the 40 mm reactor by 20% for WAS. This can be explained by the markedly reduced number of cavitation bubbles due to the high pressure and absorption by the solids. A considerable influence of the gap dimension on the cavitation intensity could also be observed in WAS. The cavitation intensity dropped from 18 dB (1 bar, 40 mm) to 5 dB (1 bar, 80 mm) with a broader reaction space. A further increase of the pressure to 2 bars reduced the impact of a broader reaction space.

The highest cavitation intensity (37 dB) was measured for the sonication of water at 1 bar and a reactor gap of 80 mm (Figure 13, Table 2). In contrast to the sonication of WAS, an increase in the cavitation intensity with broader reaction space at higher pressure was observed in water.

Thus, the cavitation noise level increased from 30 dB (40 mm, 1 bar) to 37 dB (80 mm, 1 bar). This contradicts the observations in water at ambient pressure and is remarkable inasmuch as the same ultrasound power was introduced into a larger sonicated volume. This can be explained by the previously observed reduction of the cavitation intensity in pressurized bubble clusters. Pishchalnikov et al. [25] and Zhang et al. [36] discovered that the bubbles deplete the energy of the standing waves, and bubble clusters also lead to a local reduction of density and compressibility in the liquid, which prevents the ultrasonic waves from propagating in the reactor space. This effect was also partly observed in DS with a TS of 2.2%. In DS, the highest cavitation intensity was measured with a gap of 60 mm and a pressure of 2 bars (Table 2). Unlike in water, no further increase was observed in a broader reaction space of 80 mm. In the case of WAS, the highest cavitation intensity was measured with a gap of 40 mm and a pressure of 1 bar. Broader reaction spaces lead to a strong attenuation of the cavitation bubbles due to the high solids content of the substrate (Figure 14, Figure 15).

On account of the notable influence of the static pressure on the cavitation intensity in the sludge, this aspect should be investigated in further studies. This could be achieved by realizing smaller pressure stages as well as a further increase in the static pressure. Biogas potential tests should be conducted with the treated substrates to transfer the test results to an optimised reactor design and operation conditions.

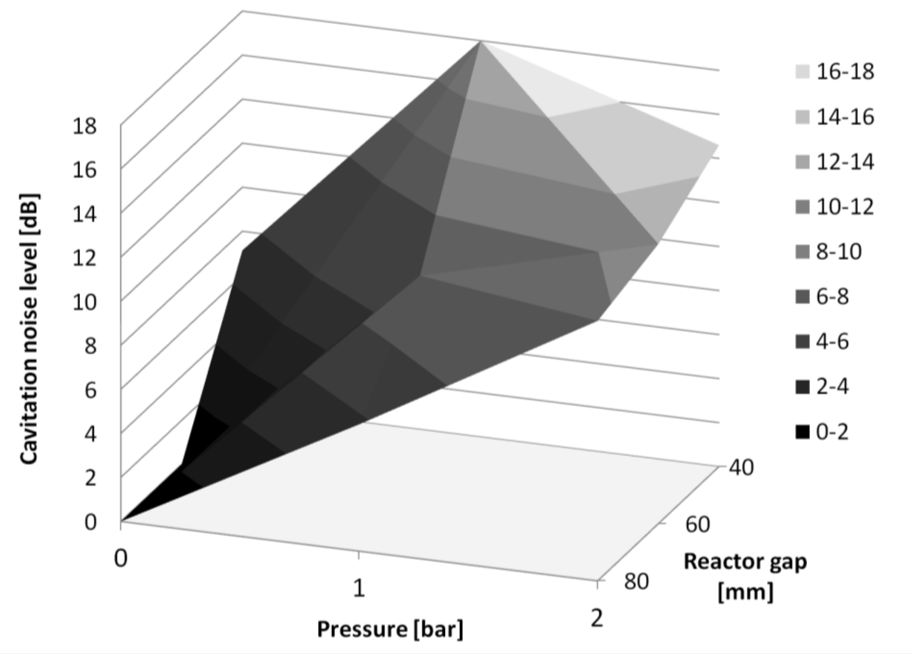

Figure 14: Cavitation intensity in waste activated sludge as a function of pressure and reactor gap distance.

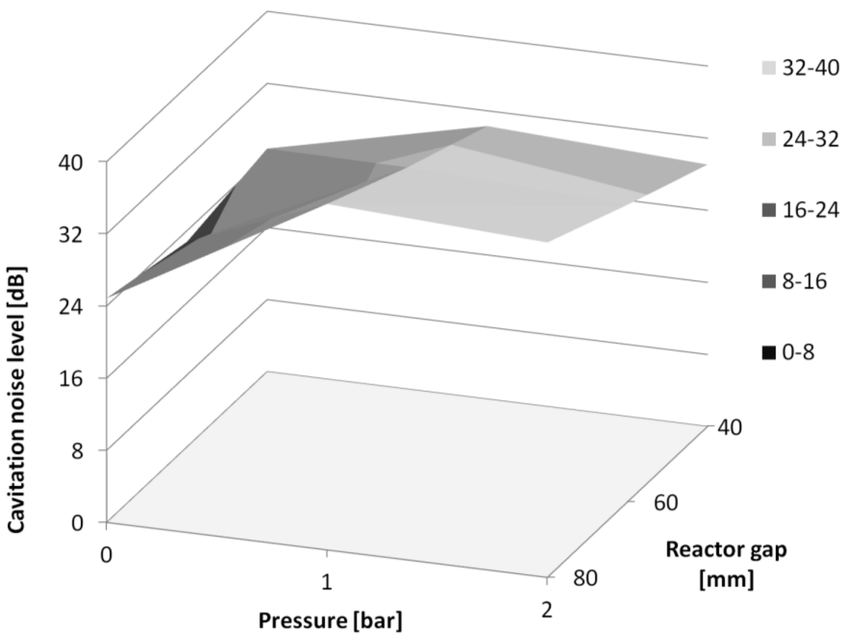

Figure 15: Cavitation intensity in water as a function of pressure and reactor gap distance.

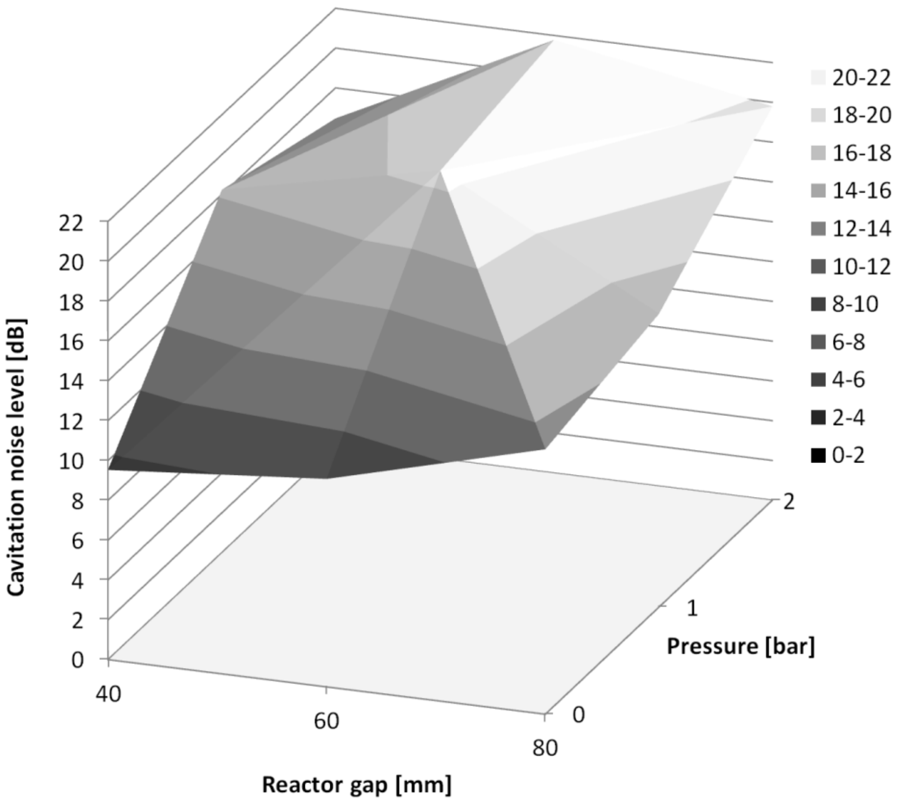

Figure 16: Cavitation intensity in digested sludge as a function of pressure and reactor gap distance.

3.1.6 Conclusions

A novel hydrophone system was used to evaluate the influence of the reaction space, substrate TS and static pressure on the intensity of the induced cavitation field. The results indicate that the cavitation intensity decreases significantly with an increasing TS in the substrates DS and WAS. When the pressure is increased to 1 bar, the intensity of bubble implosions can be enhanced and the attenuation in the solid fraction is partly compensated compared to ambient pressure. However, a further increase in pressure to 2 bars has a detrimental effect, in particular with the sonication of water at the present ultrasonic amplitude. Regarding bubble-suppressing effects, an increase in pressure is therefore only useful in a limited range to intensify the cavitation intensity. A reduction of the reactor space results in an increased intensity in WAS. This effect is less relevant in DS with its markedly lower TS content. Whereas a broad reaction space allows an intense cavitation in a pressurised water stream, it becomes clear that contradictory effects influence the development of cavitation bubbles in liquids with a solid fraction and the optimal choice of the appropriate sonication system configurations depends on the substrate to be treated. By adapting the reactor design and the process conditions to the substrate characteristics, the intensity-reducing effect of sludge that contains solids can be effectively counteracted. This allows the design of sonication devices suitable for the more intensive treatment of WWTP sludge.

3.1.7 Acknowledgements

The authors gratefully acknowledge the funding of this study by the Federal Ministry of Economics and Technology (BMWi) Grant 03ET1396A.

3.2 Paper #2 Assessment of sonotrode and tube reactors for ultrasonic pre-treatment of two different sewage sludge types

Ultrasonic treatment of sewage sludge has been investigated since many years [68,74]. Previous studies investigated the influence of the sonication on the properties of the treated sludge by the use of one certain sonication device selected mainly by chance. The influence of the design of the ultrasound generating principle was assumed to be negligible in these configurations and not in the focus of the investigation.

Concerning the aspect of identifying a sonication system that is as energy-efficient and cost-effective as possible for large-scale applications, the use of tube reactors and sonotrode reactors proved to be advantageous technical solutions [15,51]. In order to investigate the extent to which one of these systems could be particularly advantageous for sonication of a particular type of sludge, DS and WAS were sonicated using three different reactor concepts to test **Hypothesis #2:**

'The influence of the ultrasonic pre-treatment system on the degree of disintegration, gas increase, decrease in viscosity and cavitation intensity during pre-treatment of WTTP sludge with the same energy input is higher than 10%.'

To compare different ultrasonic reactors at similar conditions, a circular test setup was developed. The variation of the treatment time allowed for the sonication with comparable energy inputs, even at different reactor sizes and operation at the maximum rated power of each reactor. A broad spectrum of parameters was analyzed before and after the sonication to identify the impact and differences of each adjustment.

The results revealed significant differences concerning the tested reactor systems and were published in **Paper #2**. With the focus on low energy inputs of 300 kJ/kg_{TS}, the following statements could be made: Tube reactors revealed a 50% higher increase in gas yield with the sonication of DS than the sonotrode. On the other hand, the sonotrode reactor revealed a 77% higher increase in yield than tube reactors with the sonication of WAS. This advantage of the sonotrode was also observed at the decrease of viscosity after sonication but could not be drawn concerning the cavitation intensity and degree of disintegration. Thus, the teste**d hypothesis #2 can be partially accepted.**

The following text was published with editorial changes as:

Bandelin, J.; Lippert, T.; Drewes, J. E.; Koch, K.: Assessment of sonotrode and tube reactors for ultrasonic pre-treatment of two different sewage sludge types. Ultrasonics Sonochemistry 64, 2020, 105001

Author contribution: **Bandelin, J. (80%)**; Lippert, T. (10%); Drewes, J. E. (5%); Koch, K. (5%)

3.2.1 Highlights

- Sonotrode and tube reactors are both suitable for sewage sludge disintegration

- Sonotrode reactors provide the highest increase in gas yield of waste activated sludge

- Tube reactors provide the highest increase in gas yield of digested sludge

- Sonication of sewage sludge results in accelerated degradation kinetics

- Sonication decreases the viscosity of waste activated sludge

3.2.2 Graphical abstract

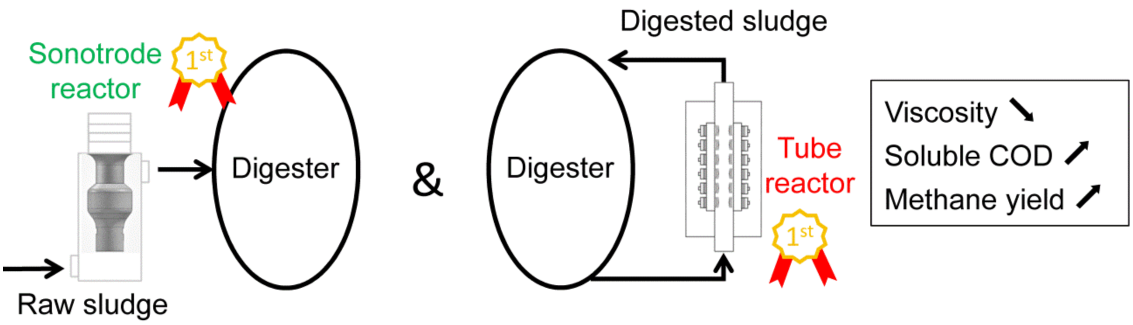

Figure 17: Graphical abstract of Paper II

3.2.3 Abstract

The effectiveness of tube and sonotrode reactors for the sonication of sewage sludge under identical conditions was compared for the first time. Despite the considerable structural differences, sonication with each ultrasonic reactor led to an accelerated degradation rate and an increased methane production within the first five days for the majority of the sewage sludge samples tested. On closer examination, however, it becomes clear that the investigated sonication systems are not equally suitable for the substrates considered. While the use of a sonotrode proved to be particularly advantageous for the treatment of waste activated sludge (+25% methane yield at 300 kJ/kg$_{TS}$), the use of a 2-inch tube reactor achieved the highest enhancement for low-intensity sonication in digested sludge (+22% methane yield at 300 kJ/kg$_{TS}$).

With increasing energy input, more chemical oxygen demand was solubilized, but this did not result in an increase in methane yield for all samples. Sonication of waste activated sludge led to a significant reduction in viscosity of up to 50%, and a reduction of up to 60% was observed after sonication of digested sludge with low energy inputs. The study, therefore, demonstrates that the choice of the most suitable sonication system essentially depends on the properties of the sludge to be sonicated.

3.2.4 Keywords

Ultrasonic tube reactor; sonotrode reactor; sludge disintegration; biogas production; sludge viscosity; hydrophone

3.2.5 Introduction

Increasing energy recovery by anaerobic digestion has become a major objective for many municipalities while transitioning to a more energy-efficient treatment of wastewater. Ultrasonic pre-treatment of sewage sludge has revealed a high potential for the increase in methane yield in laboratory studies. This can be explained by the mechanic disintegration of the particles and the release of organic matter, which is thus available for the digestion process. [68,70]. In addition, a reduction in viscosity, observed at laboratory-scale sonication [88], could reduce the energy demand for agitation and provide a more homogeneous distribution of the substrate in the digester.

In order to transfer these promising results to full-scale applications, the most suitable ultrasonic technology needs to be identified for the given conditions. For this purpose, different sonication technologies were tested under the same conditions and with different sludge types for the first time. Besides the intended increase in gas yield and reduction in viscosity, further parameters were considered for a broad comparison of the ultrasonic systems. These included the direct analysis of the cavitation intensity in the reactors as well as the increase in soluble chemical oxygen demand (sCOD) after sonication, which is seen as a useful surrogate parameter for the assessment of the extend of cell disintegration [89]. Two fundamentally different ultrasonic generating technologies are employed to provide intensive cavitation fields in the liquid media. These include rod-shaped sonotrodes, which are immersed in the medium to be sonicated, as well as surface oscillation systems, in which the reactor wall serves as the resonant system. While both technologies convert the electrical energy into mechanical oscillations by piezo-ceramic disks [90], the sonication effect, structural design, materials and production effort differ substantially [91]. The shape of the resonant oscillation system determines the size of the sound emitting surface and the amplitude of the oscillation system. While rod-shaped sonotrodes generate an intensive ultrasonic field with a high amplitude in a small area mostly at the tip [92], a large cavitation field with a smaller amplitude can be achieved with surface oscillation systems [93]. Tube reactors are a prominent example of surface oscillating systems [51]. They provide the advantage of no obstructing objects in the reaction chamber, which may cause clogging or blocking of the system [94]. These fundamentally different properties dictate the possible designs of sonication systems. So far it remains unclear whether one system is possibly superior for the treatment of a certain sewage sludge type.

Several laboratory-scale studies have already revealed the potential of ultrasound treatment to increase the methane yield with both technologies applied. Sonotrode induced sonication of waste activated sludge (WAS) with an energy input of 6,250 kilojoules per kilogram of total solids (TS) led to a 51% increase in methane yield and a degree of disintegration (DD_{COD}) of 15% based on the increase in sCOD [89].

A lower energy input of 620 kJ/kg_{TS} caused a 27% increase in methane yield when using a sonotrode for the sonication of WAS in a separate study [95].

In contrast, by the application of tube reactors an increase in methane yield of sonicated digested sludge (DS) of up to 63% was observed [51]. This remarkable result was achieved in a laboratory-scale study with a comparatively high energy input of 5,443 kJ/kg_{TS}, which would lead to extensive and not cost-efficient power requirements in full-scale applications. A less intensive sonication of WAS with only 327 kJ/kg_{TS} in another study with a smaller tube reactor still achieved an increase in methane yield of 20% [13]. In a further study investigating sonication of WAS with very low energy inputs of 200 kJ/kg_{TS} and the use of rectangular tube reactors, a DD_{COD} of 6% and an increased in methane yield of 12% were achieved on average. This treatment configuration was identified to possibly reach a positive energy balance [70].

Thus, the use of sonotrode systems as well as tube reactors revealed a positive influence on the methane yield and the increase of the sCOD at laboratory-scale for both substrates, DS and WAS. The wide range of results obtained so far indicates that the composition of the substrate has a considerable impact on the effects of ultrasonic treatment [70,88,93]. The efficiency of sonotrode and surface oscillation systems can therefore not be reliably evaluated based on previous, independently conducted experiments with different oscillation systems, sludge compositions, and energy inputs [25].

In the present study, a direct comparison between sonotrode systems and tube reactors under identical conditions was performed for the first time. In order to determine the influence of the ultrasonic technology and the reactor design on the effectiveness of the disintegration, three different ultrasonic systems consisting of a tube reactor with an inner diameter of 2 inch (2”-tube reactor), 3 inch (3”-tube reactor), and a flow-through sonotrode reactor were selected and the cavitation intensity in the respective ultrasonic system was measured with a hydrophone. In addition, samples of DS and WAS from the same source were exposed to the ultrasound treatment. The effectiveness of the process was evaluated with regard to the change in viscosity, the degree of disintegration (DD_{COD}), and the methane yield achieved in a biochemical methane potential (BMP) test.

3.2.6 Material and methods

3.2.6.1 Substrates

The examined substrates were collected from the wastewater treatment plant (WWTP) Freising, Germany. WAS samples were collected immediately after the belt filter press and provided a TS content in a range of 4.15% to 5.09% and a volatile solids (VS) content from 3.09% to 3.64% depending on the operational conditions. Due to the limited capacities of the BMP test system, different energy inputs were investigated in a series of sequential phases, which caused the slight variation in the substrate properties. The DS samples were taken from the recirculation loop of the digester. The sludge provided a TS content in a range from 2.78% to 2.94% and a VS content in a range from 1.64% to 1.75%.

As DS contained few coarse particles (such as plastic parts, stones/concrete, wood chips, etc.), the sample was sieved through a 1.5 mm mesh prior to the experiments to avoid a negative impact on the reproducibility among the replicates in the BMP tests. TS and VS were determined according to standard methods [96]. Regular tap water with a temperature of 20°C was used as reference medium for the cavitation field analysis.

3.2.6.2 Experimental setup

The experiments were carried out in a circulation system, consisting of an ultrasonic test reactor (2”-tube reactor, 3”-tube reactor, and sonotrode reactor, respectively), an eccentric screw pump, and tube connections (Figure 18). In a previous study, the increase in overpressure to 1 bar intensified cavitation intensity, which provides the potential for further optimization with close to none additional effort or energy input [93]. Therefore, a pressure gauge and a ball valve were installed at the outlet of the reactor to adjust the overpressure at a level of about 1 bar at a constant flow of 300 L/h. DS was treated with specific energy inputs of 300 kJ/kg_{TS}, 437 kJ/kg_{TS}, and 3,500 kJ/kg_{TS}. WAS was treated with energy inputs of 300 kJ/kg_{TS}, 500 kJ/kg_{TS}, and 3,500 kJ/kg_{TS}. The specific energy inputs were achieved by sonicating and circulating the substrate for a certain time based on the reactor volume and ultrasonic power.

The slightly lower specific energy input of 437 kJ/kg_{TS} in DS compared to 500 kJ/kg_{TS} in WAS was the consequence of the experimental procedure in which the duration of the sonication was defined based on the TS from a previous experiments. The real TS of the sample was measured subsequently.

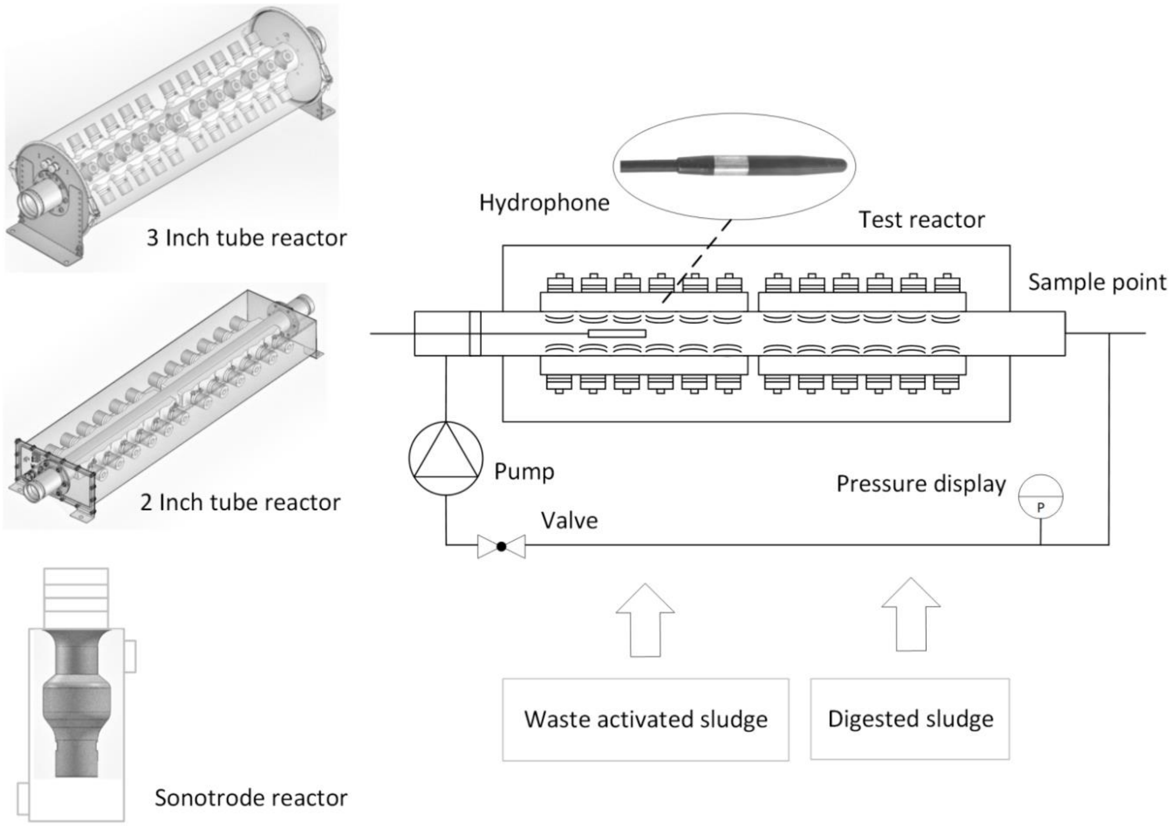

Figure 18: Schematic illustration of the tested ultrasonic reactors (left). Experimental setup with test reactor (2”-tube reactor as an example), eccentric screw pump, and tube connections (right).

3.2.6.3 Ultrasonic reactors

In order to determine the influence of the ultrasonic technology and the reactor design on the effectiveness of the disintegration, two different tube reactors and one sonotrode reactor were applied in the experiment. The frequency was selected in a range of 20-25 kHz, since a particularly high proportion of large cavitation bubbles with high shear forces was expected to occur in this range [67,97]. Previous experiments regarding the sonication of high solids content WAS revealed that the distance between the sound emitting surfaces in flat transducer systems is highly relevant for the cavitation intensity in the ultrasonic reactor [93], while a reactor limited in size would increase the average flow velocity and reduce the retention time [98].

Therefore, the three tested sonication systems were chosen to balance these requirements and to be possibly applicable at full-scale.

The comparatively small 2”-tube reactor SB 8 (BANDELIN electronic GmbH & Co. KG, Berlin, Germany) with an ultrasonic frequency of 25 kHz and a nominal power of 1,000 W served as example for a small reaction chamber. The reactor consists of a stainless steel tube with 24 piezo-ceramic ultrasonic transducers attached to the tube with a wall thickness of 3.65 mm and an inner diameter of 53 mm. The alignment of the transducers results in a sonication area of 150 cm^2 and a sonicated volume of about 2 liters with a power density of 500 W/L.

The larger 3”-tube reactor SB 101 (BANDELIN electronic GmbH & Co. KG, Berlin, Germany) with an inner diameter of 79.3 mm operated with an ultrasonic frequency of 25 kHz provided a nominal power of 2,000 W. However, the power density in the sonication area was slightly reduced to 440 W/L due to the significantly larger volume of about 4.5 L.

The sonotrode TS 550 (BANDELIN electronic GmbH & Co. KG, Berlin, Germany) was operated with an ultrasonic frequency of 20 kHz and achieved an output of 1,000 W with a probe diameter of 50 mm. The sonotrode was installed in a flow cell with a quadratic cross-section of 120 mm and a length of 400 mm resulting in a sonicated volume of about 5.8 L. Due to the reduced power density of 172 W/L the retention time was increased to ensure the same specific energy input for each reactor tested.

3.2.6.4 Cavitation measurement

The intensity of the cavitation bubble implosions in the reactor was measured by means of a hydrophone according to IEC TS 63001:2019 [99]. The use of a RESON TC4034-1 hydrophone (RESON A/S, Slangerup, Denmark) in the present study allowed the separate detection of the sound pressure of the ultrasonic frequency (f_0) and the emitted noise of imploding cavitation bubbles. The control unit detected the sound peak pressure in a frequency range of $2.15 \cdot f_0$ to $2.35 \cdot f_0$ and determined the cavitation noise level in decibel [dB]. The radial-sensitive hydrophone, with a diameter of 15 mm, was positioned in the reactor chamber, immersed into the medium that was circulated during the measurement.

3.2.6.5 Viscosity analysis

The dynamic viscosity of WAS and DS was determined by a rotational rheometer (Viscotester VT 500, HAAKE Messtechnik GmbH & Co., Karlsruhe, Germany) using a cylindrical spindle of the type MV-DIN for WAS and SV-DIN for DS, respectively. All investigations were conducted in triplicates at a constant temperature of 20°C.

3.2.6.6 BMP tests

The BMP tests were carried out with the Automatic Methane Potential Test System (AMPTS II; Bioprocess Control Sweden AB, Lund, Sweden). In order to achieve the highest possible comparability of the results, the experimental protocol strictly followed the recommendation proposed by Holliger et al. [15]. The ratio of inoculum (untreated DS) and substrate (WAS) was adjusted to be 2:1 based on VS content, which is compliant to the standardized test conditions described elsewhere [100]. In order to avoid a weakening of the effect of the ultrasonic treatment, the DS samples were not mixed with untreated DS assuming that the biocenosis can still be reactivated. The inoculum was tested concerning its endogenous methane production and its activity by the use of blanks and positive control samples with microcrystalline cellulose as substrate. The digestion temperature was set to 38 ± 0.5 °C, which corresponds to the operating temperature of the digester the substrate originated from. The homogenous heat distribution was ensured by the submersion of the samples in a covered water bath.

All samples were tested in triplicate and the tests were performed for at least 28 days and until the daily methane production during three consecutive days was less than 1% of the total methane production [100,101]. The test results were validated based on a relative standard deviation of less than 5% within each triplicate [101]. The significance of differences between the sonicated and blank samples was investigated by means of the Student's t test. Samples with a result of $p < 0.05$ were considered *significant* and samples with a result of $p < 0.01$ were considered *highly significant*.

3.2.6.7 Chemical oxygen demand (COD) analysis

sCOD was determined according to standard procedures [96]. The disintegration effect of the sonication was subsequently analysed by the comparison of the relative increase in the sCOD in the sonicated sample ($sCOD_{US}$) in comparison to the maximum increase after the alkaline hydrolysis in a NaOH solution ($sCOD_{NaOH}$). The 1 M NaOH solution was mixed with the substrate in a ratio of 2:1 and stirred at room temperature for 24 h in accordance to a previous study [89]. The initial concentration of sCOD was defined as $sCOD_0$. The resulting degree of disintegration (DD_{COD}) describes the ratio of the sCOD increase of the two methods and provides a reliable indicator of the release of organic matter in the sonicated substrate:

$$DD_{COD} = \frac{sCOD_{US} - sCOD_0}{sCOD_{NaOH} - sCOD_0} \quad [5]$$

3.2.7 Results and discussion

Three different reactor systems were tested on their performance for the treatment of different sludge types. The intensity of the ultrasonic treatment of the respective reactor was measured in water, WAS and DS. The disintegration performance of the applied sonication systems was also studied by the analysis of the rheological properties before and after sonication. sCOD was determined in order to detect the soluble organic matter release during the sonication process. The methane yield after sonication was measured in comparison to the untreated sample. In order to determine the most suitable energy input, the investigations were performed with three different energy inputs in the range from 300 kJ/kg_{TS} to 3,500 kJ/kg_{TS}.

3.2.7.1 Cavitation field analysis results

The cavitation field analysis allows for the quantification of the cavitation intensity in the ultrasonic reactor. There are several factors influencing the formation of cavitation bubbles, which is a required prerequisite for a successful sludge disintegration. Firstly, the design of the ultrasonic system including the type of sound-emitting surface, energy input, frequency, etc. [102,103]. Secondly, the solids content of the medium, as particles are known to attenuate the propagation of the ultrasound [93,104]. Thirdly, the flow conditions of the medium, as a turbulent flow pattern in the reactor might suppress the formation of cavitation bubbles [79].

The results in Figure 19 suggest a decrease of cavitation intensity with increasing TS content of the medium. This behaviour results from the sound field attenuation by the solid particles and compares well with the results of a previous experiment with other ultrasonic systems [93]. These results reveal that the cavitation intensity of the different ultrasonic systems varies by less than 6% when the reference medium water is sonicated. In contrast, a distinct difference in the cavitation intensity of up to 24% could be measured between the 3”-tube reactor and the sonotrode reactor in DS. However, this difference is largely negated in the sonication of WAS. High cavitation levels were measured in all configurations. Therefore, intensive cavitation fields can be assumed for all considered reactors.

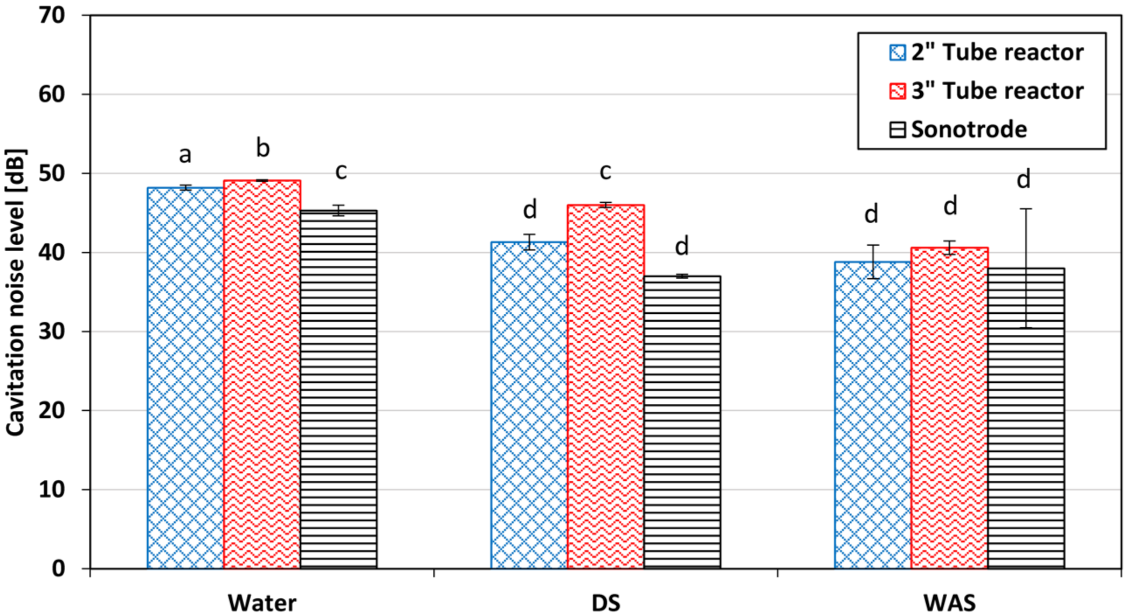

Figure 19: Cavitation noise level in different substrates and reactor types. Error bars denote standard deviation from the average of the five repetitions. Letters indicate groups of significant differences according to a Student’s *t* test at 5% significance level.

3.2.7.2 Rheological results

The sonication of DS in the 3”-tube reactor with an energy input of 300 kJ/kg_{TS} led to a reduction in viscosity of 35% at a shear rate of 58.44 s^{-1}, which is in the range of a fast running stirrer at large-scale. For better comparability and clear differentiation, this shear rate was taken into account in all viscosity considerations in this study.

By increasing the energy input to 437 kJ/kg_{TS}, the viscosity was further decreased by 60% (Figure 20). In this case, the sonication of DS with the 3”-tube reactor revealed a 32% stronger reduction compared to the 2”-tube reactor and a 7% enhanced reduction compared to the sonotrode. On the other hand, the sonication of DS with an energy input of 3,500 kJ/kg_{TS} resulted in an increase of viscosity of up to 29%. This finding could possibly be explained by the release of extracellular polymeric substances during highly intensive sonication [105].

The ultrasonic treatment of WAS with a low energy input of 300 kJ/kg_{TS} did not show a noticeable effect on the rheological properties. Further increase of the energy input to 500 kJ/kg_{TS} led to a 44% reduction in viscosity in the sonotrode system.

The increase in the energy input to 3,500 kJ/kg_{TS} resulted in an even higher reduction in apparent viscosity of 50% for WAS in the 2”-reactor. The results confirm the clear relationship between the energy input and viscosity reduction in WAS observed in previous studies with a higher specific energy input in the range of 4,000 kJ/kg_{TS} to 27,000 kJ/kg_{TS} [88,106]. Another study observed an abating decrease at a level of 73 % and a shear rate of 30 s^{-1} with an energy input of up to 33,000 kJ/kg_{TS} [16].

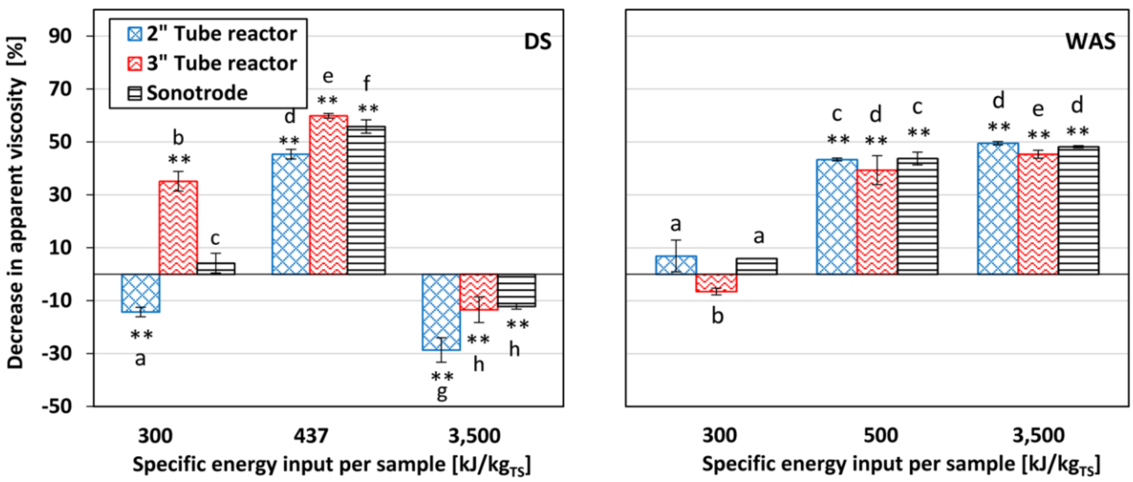

Figure 20: Change in apparent viscosity of DS (left) and WAS (right) at a shear rate of 58.44 s^{-1} after sonication with different reactor types and specific energy inputs. Error bars denote standard deviation from the average of the three replicates. Letters indicate groups of significant differences according to a Student’s *t* test at 5% significance level.

3.2.7.3 BMP test results

All specific methane production curves followed the typical pattern over time [107]. Sonication of the samples led to an acceleration of the degradation rate and an increase of the methane yield within the first five days for the majority of the samples (17 out of 18). This can be explained by the release of easily degradable substrates induced by the mechanical disruption effects of imploding cavitation bubbles [67]. However, the advantage achieved initially was mainly reduced over the course of the experiment. This restraint was most evident after the intensive sonication of DS with 3,500 kJ/kg_{TS}, resulting in a 2.5-times higher methane production on the first day compared to the untreated sample and the consequent reduction of the increase to 60% after an incubation time of 28 days (Figure 21). Subsequent to the sonication with an average energy input of 437 kJ/kg_{TS}, the initial additional production of 25% was even fully compensated after an incubation time of 28 days and even a lower methane yield was obtained compared to the untreated sample. A further reduction of the energy input for the sonication of DS showed a positive effect and resulted in a sustained increase of the methane yield by an average of 10%. These results confirm the observations of a previous study, which also reported a deterioration of the methane yield of DS at energy inputs in the range of 500 to 1,200 kJ/kg_{TS} [51]. A potential reason for this observation can be found in the partial disintegration of the biomass, which results in a proportionally high release of floc-forming substances and thus, an inhibition of degradation [70,108]. As a high-intensity sonication at full-scale aiming for an increased methane production might lead to a negative overall energy balance [70], focusing on the sonication with very low energy inputs of 300 kJ/kg_{TS} was considered as more appropriate. Especially for WWTPs operating digesters at their design capacity and a correspondingly short retention time of less than 20 days, the installation of an ultrasonic system could increase the methane yield and at the same retention time serve as a suitable alternative to the construction of an additional digester. Concerning the choice of the best-suited reactor design for the co-treatment of DS, all tested reactor designs revealed a positive effect on the methane yield. Especially with low energy inputs, the 2-inch-tube reactor was able to achieve the highest effect with a significant increase in methane yield of 22% after 28 days. With a very high energy input, this advantage was caught up, which can be explained by saturation effects once the highest possible particle disintegration is achieved. In this case, the sonotrode reactor revealed a slightly higher increase compared to the tube reactors.

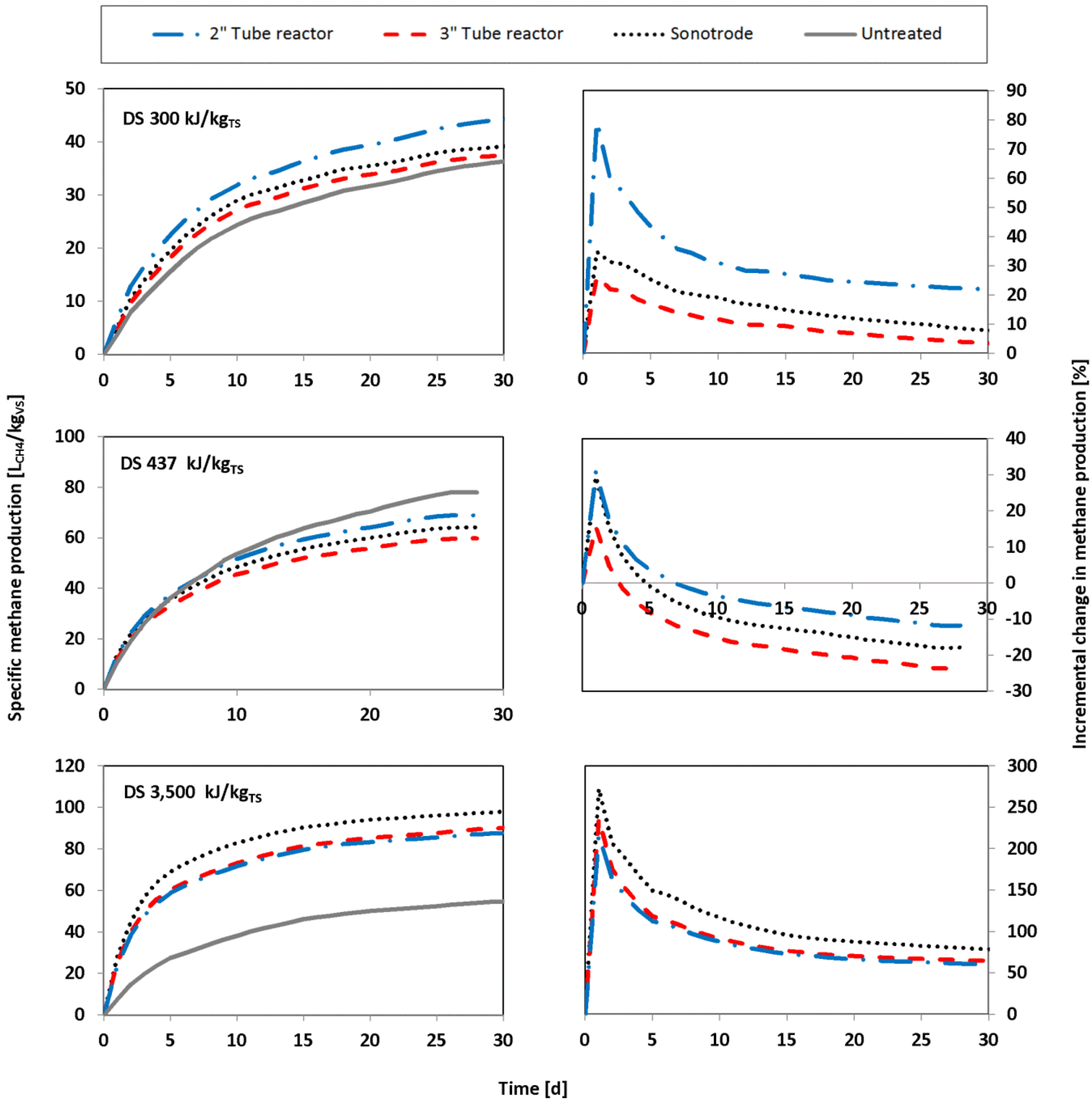

Figure 21: Average specific methane production (left) of DS and average incremental increase in specific methane production relative to the untreated sample (right) after sonication with different reactor types and different specific energy inputs. 4 out of 12 samples did not match the quality criterion of a relative standard deviation lower than 5%: 6.2% for sonotrode DS 300 kJ/kg_{TS}, 12.7% for 2-inch-tube reactor DS 300 kJ/kg_{TS},

6.6% for 2-inch-tube reactor DS 437 kJ/kg$_{TS,}$ and 7.0% for 3-inch-tube reactor DS 437 kJ/kg$_{TS}$.

When considering the potential for increasing the methane yield of WAS, a similar pattern was observed as for the sonication of DS (Figure 21).

Even with the lowest energy input tested, the methane yield could be significantly increased with all sonication technologies by an average of 18% after 28 days of incubation. In this constellation, the enhanced production established over the entire incubation time and even rose in particular samples. An increase in the energy input to 500 kJ/kgTS also led to an increase in the methane yield when sonotrode and 3”-tube reactor were used, but the overall surplus after 28 days with this energy input was only 6% and 4%, respectively. The absence of an increase in methane production in WAS after sonication with a specific energy input in a range of 500 kJ/kgTS to 1,500 kJ/kgTS confirms the observations of previous studies [1,10,22] and is independent of the sonication technology applied. A further distinct increase at the energy input to 3,500 kJ/kgTS during the sonication of WAS led to a short-term increase in production, but was reduced throughout the incubation time and a significant increase in gas yield of only 12% could be observed with the sample treated with the sonotrode reactor.

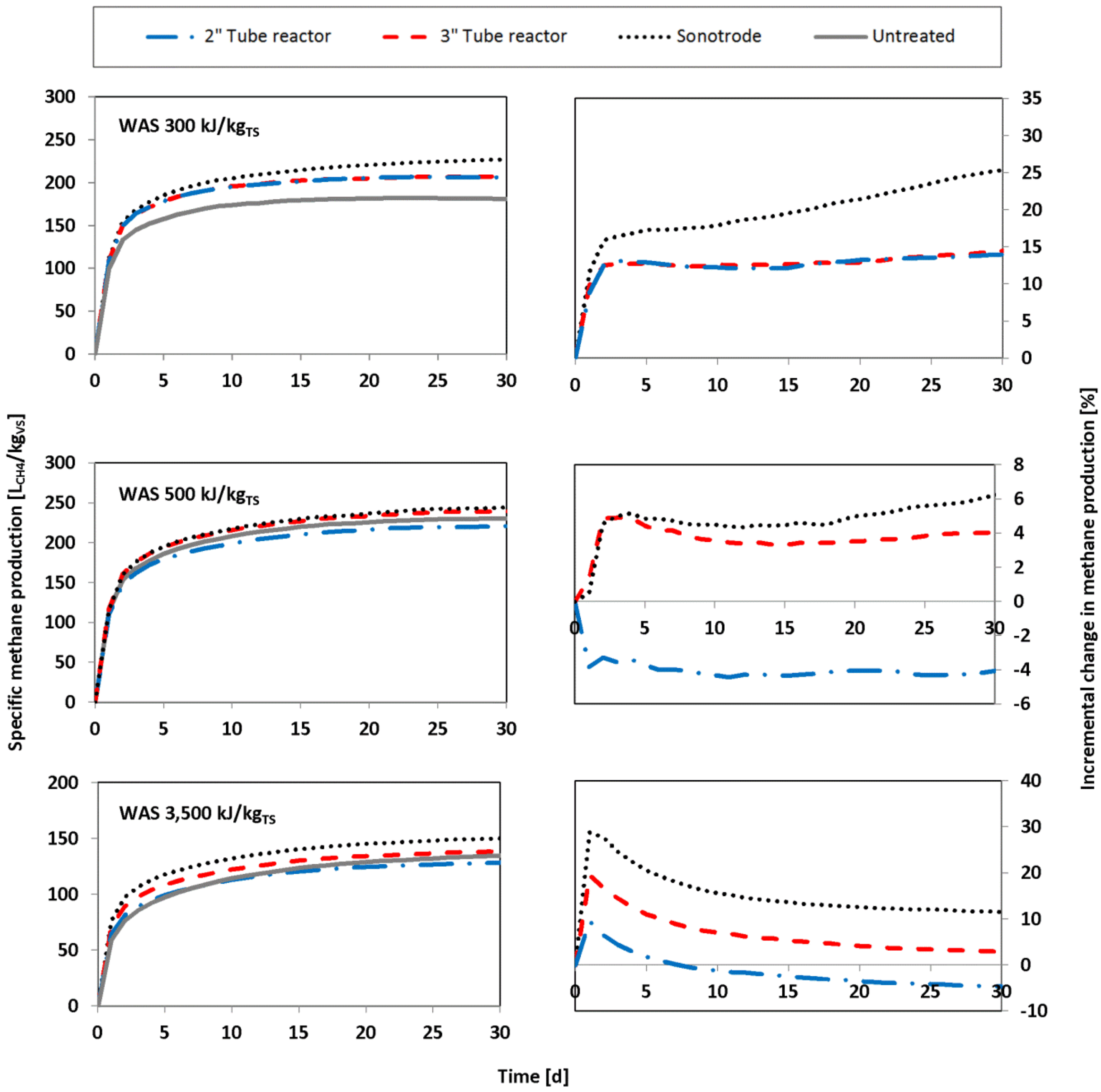

Figure 22: Average specific methane production (left) in WAS and average incremental change in specific methane production relative to the untreated sample (right) after sonication with different reactor types and different specific energy inputs.

Altogether, a considerable impact of the sonication, especially by accelerating the degradation in the first days, was observed. The most significant relative proliferation potential was observed during the sonication of DS due to its improved digestibility of recalcitrant substances that are otherwise poorly available for the degradation process (Figure 23).

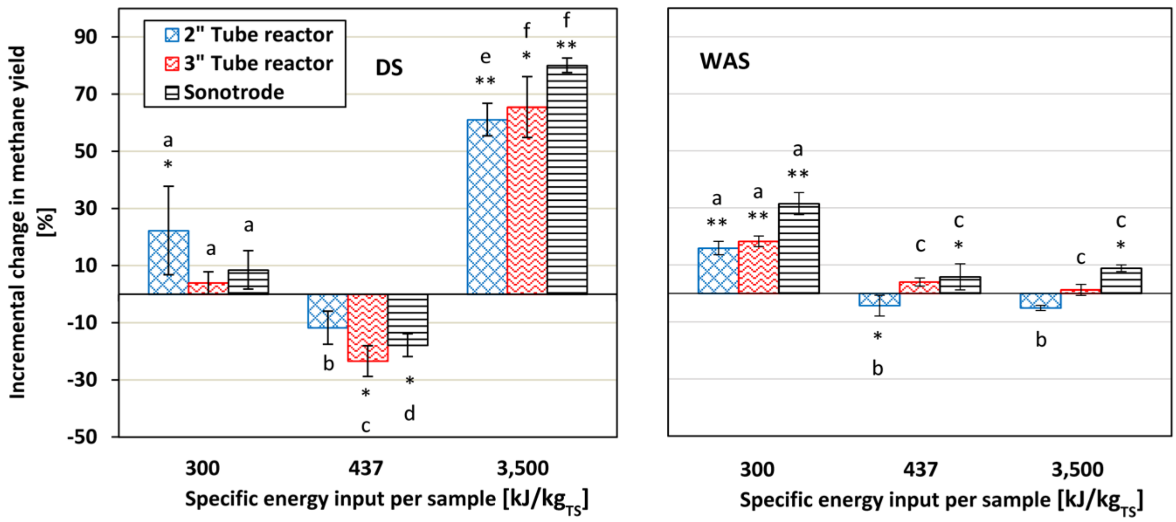

Figure 23: Incremental change of specific methane yield of DS (left) and WAS (right) relative to the untreated sample after 28 days incubation time. Error bars denote standard deviation from the average of the three replicates. Significant differences to the untreated samples according to the Student's *t* test are marked with one asterisk at $p < 0.05$ and with two asterisks at $p < 0.01$. Letters indicate significant differences according to a Student's *t* test at 5% significance level.

3.2.7.4 Impact on soluble COD release

There are conflicting information in the literature whether sCOD release is a suitable surrogate parameter for the chance of the methane yield of a sample due to the treatment [51,109]. While the sonication of DS with an energy input of 300 kJ/kg$_{TS}$ did not result in a significant increase in DD_{COD} (Figure 24), the sonicated samples revealed a significant increase in methane yield by 12% compared to the untreated sample (Figure 23). Increasing the specific energy input to 437 kJ/kg$_{TS}$ resulted in a DD_{COD} of 3% after treatment with the 2”-tube reactor, but the final methane yield was lower than the untreated sample. A substantially higher DD_{COD} of 27% could be achieved with a more intensive sonication of 3,500 kJ/kg$_{TS}$ resulting in an additional methane yield of 70%.

The sonication of WAS with a low energy input of 300 kJ/kg$_{TS}$ resulted in an average DD_{COD} of 14%. The increase in the energy input to 500 kJ/kg$_{TS}$ revealed an average DD_{COD} of 21%, which was only surpassed by an average DD_{COD} of 71% after the treatment with 3,500 kJ/kg$_{TS}$.

In both experiments, the sonication with the sonotrode resulted in the highest effect in comparison to the other sonication methods. However, concerning the sonication with low energy input, the difference between the 3”-tube reactor and the sonotrode was of small extent.

The increase in sCOD after sonication is reflected by an additional methane production in the BMP tests, but cannot be taken into account for a prediction of the absolute methane yield after the complete incubation time. Therefore, the degree of disintegration (DD_{COD}) can be utilized to quantify the release of readily degradable substances. However, as indicated by the BMP tests, the desired increase in the achievable methane yield appears to be additionally dependent on other factors, which confirms the observations of previous studies that the degree of disintegration can only give a first indication of the efficiency of a treatment method, while digestion tests have to follow to evaluate the impact on the methane yield [8,27].

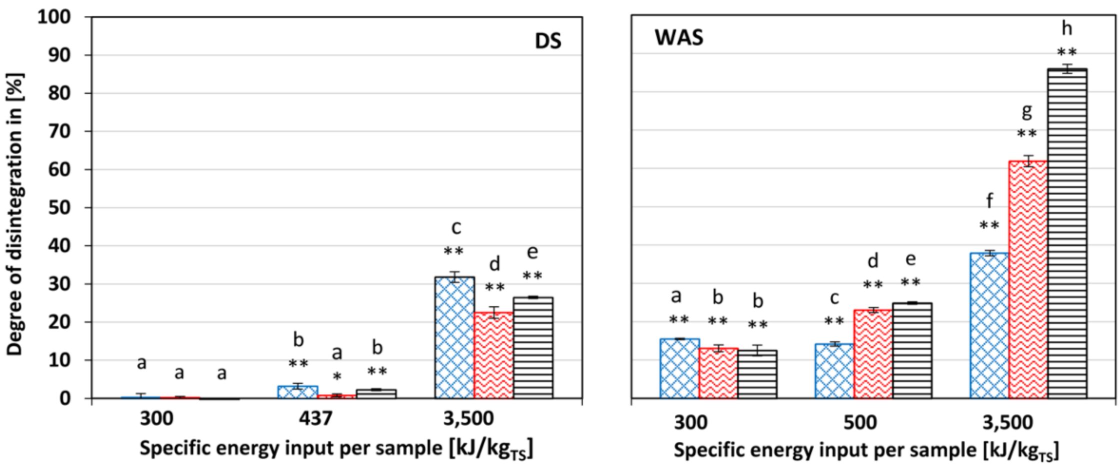

Figure 24: Degree of disintegration after sonication of DS (left) and WAS (right). Error bars denote standard deviation from the average of the three replicates. Significant differences to the untreated samples according to the Student’s *t* test are marked with one asterisk at $p < 0.05$ and with two asterisks at $p < 0.01$. Letters indicate significant differences according to a Student’s *t* test at 5% significance level.

3.2.7.5 Comparison of ultrasonic reactors

Both tubular and sonotrode reactors confirmed their suitability for the sonication of sewage sludge and the generation of intensive cavitation. While sonication of WAS led to a significant reduction in viscosity, a decrease was observed after sonication of DS with low energy inputs and an increase in viscosity after intensive sonication. With increasing energy input, more sCOD was released, but this did not automatically resulted in an increased methane yield for all samples. While the use of a sonotrode reactor proved to be particularly advantageous for the treatment of WAS, the use of a 2”-tube reactor achieved the highest effect for low-intensity sonication in DS. For the evaluation of the optimal sonication system for full-scale application, other factors such as lifetime, maintenance intervals (for instance, the exchange of sound emitting parts exposed to erosion), and resistance to interfering substances must be taken into account in addition to the differences determined in terms of efficiency to increase the methane yield. The resistance of tubular reactors for clogging and the positive effect on the methane yield make them particularly suitable for the sonication of DS, which is often contaminated with plait-forming materials originating from the primary sludge. Sonotrodes on the other hand revealed a more comprehensive release of organic matter in sonication of WAS with a high energy input.

3.2.8 Conclusions

The experiments in this study have shown that the sonication applying both tube and sonotrode reactors achieved a significant effect in sewage sludge. Different characteristics of the ultrasonic devices applied led to different results depending on the sonicated substrates. Sonotrodes proved to be advantageous for the sonication of the highly viscous WAS with a high TS content. Tube reactors, however, achieved a larger increase in methane yield with the sonication of the thinner and lower viscous DS with the same specific energy input. The investigation therefore demonstrated, that the choice of the most suitable sonication system essentially depends on the properties of the sludge to be sonicated.

3.2.9 Acknowledgments

The authors gratefully acknowledge the funding of this study by the German Federal Ministry for Economic Affairs and Energy (BMWi), Grant 03ET1396A. The company BANDELIN electronic GmbH & Co. KG is kindly acknowledged for providing the test equipment.

3.2.10 Declaration of interest

There are no conflicts of interest associated with this publication.

3.3 Paper #3 Impact of high flow rates and increased viscosity of digested sewage sludge on the cavitation intensity in ultrasonic tube reactors

Analysis of the results presented in **Paper #1** indicates that a small gap size and the lowest possible TS have a positive effect on the intensity of the cavitation field. This observation was confirmed in **Paper #2** by the analysis of the methane yield after sonication of two sludges with different reactor chamber dimensions.

In order to derive suitable design criteria for the application of the technology in full-scale, this implies that the use of a large number of smaller reactors would result in a more effective sonication than the use of fewer large sonication systems with a gap size of more than 60 mm. With regard to the most cost-effective plant scheme possible, the series connection of many reactors would be more advantageous than a parallel connection with complex flow distribution.

In terms of generating an intensive cavitation field, DS has an advantage over WAS. On the other hand, a flow rate of almost twice as high is required to achieve a specific energy input of 300 kJ/kg_{TS}, which has shown an increased potential in previous tests [110]. These two objectives result in a treatment setup in which many small tube reactors connected in series have to be operated with a large flow rate. This operation mode would inevitably lead to a high average flow speed that could limit the cavitation intensity [72]. To which extent this effect also occurs with flow properties differing from those of water as in DS was evaluated by testing hypothesis #3.

Hypothesis #3: *'An average flow velocity of more than 0.3 m/s suppresses the cavitation field during sludge treatment with ultrasonic tube reactors.'*

For the analysis of hypothesis #3, at conditions and reactor designs that may also be relevant for full-scale use, a technical-scale experimental setup was used, which allowed for the continuous adjustment of volume flows of more than 16 m^3/h in a tube reactor.

The results revealed the expected reduction of cavitation intensity at volume flows higher than batch operation. However, in contrast to the assumptions made in the hypothesis, the complete suppression of the cavitation intensity does not occur at a volume flow of 0.3 m/s, but only at significantly higher volume flows above 1.5 m/s**.** **Hypothesis #3 can, therefore, be partly accepted.**

When performing the experiment, also a slight increase in cavitation intensity at medium flow velocities in the range of 0.8-1.3 m/s was observed in DS (TS 2.52 to 2.78%) compared to clear water. In terms of plant design for the full-scale application, this implies that a flow rate of 7 m^3/h could be sonicated with an optimum intensity of 300 kJ/kg_{TS} by means of a series connection of 15 reactors with a diameter of 2 inches and an intput of 1 kW. With an average capacity of 2,500 m^3, the entire digester volume would be sonicated within 15 days on average, which exceeds the hydraulic retention time of most facilities.

The following text was published with editorial changes as:

Bandelin, J.; Lippert, T.; Drewes, J. E.; Koch, K.: Impact of high flow rates and increased viscosity of digested sewage sludge on the cavitation intensity in ultrasonic tube reactors. Chemical Engineering & Processing: Process Intensification 152 (2020) 107925

Author contribution: **Bandelin, J. (80%)**; Lippert, T. (10%); Drewes, J. E. (5%); Koch, K. (5%)

3.3.1 Graphical abstract

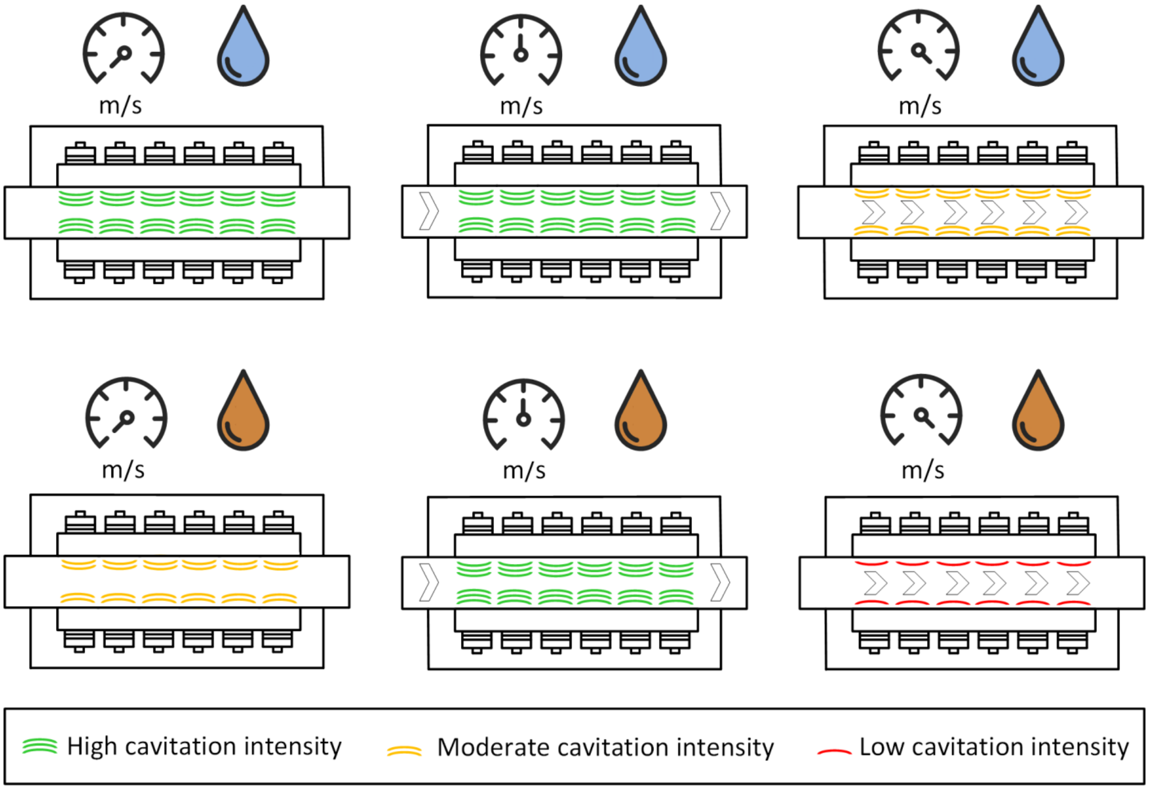

Figure 25: Graphical abstract of Paper III.

3.3.2 Abstract

The impact of high flow rates and increased viscosity of digested sewage sludge (DS) on the cavitation intensity in ultrasonic tube reactors was determined by use of a hydrophone. The results indicate a reduction of cavitation intensity in DS by 11% at an average flow velocity of 0.4 m/s compared to batch sonication. Slightly increased flow velocities in the range of 0.8-1.3 m/s resulted in an increase in cavitation intensity in DS of up to 9%. Further acceleration of the flow leads to a substantial reduction of cavitation noise level both in DS and in water of up to 49%. The results provide valuable insights for the design of sonication systems in wastewater treatment plants. It can be concluded that the sonication system must be designed in such a way that the sludge is in motion during sonication, but the flow velocity does not exceed the limit of laminar flow.

3.3.3 Keywords

Sludge disintegration; anaerobic digestion; ultrasonic reactor; flow velocity analysis; hydrophone

3.3.4 Introduction

Wastewater treatment plants (WWTP) are the largest energy consumers in municipalities. Therefore, every improvement in the energy recovery of the wastewater treatment processes can contribute to the worldwide reduction of energy demand. In particular, the utilization of sewage sludge in anaerobic digestion offers a significant potential of additional energy production and a reduction of residual sludge, which is still not fully utilized so far [111,112]. An improvement of the digestion processes can be achieved by the disintegration of the sludge particles, which are thus made available for the fermentation process to enhance biogas production [13,14,113].

Disintegration can be conducted thermally, chemically, or mechanically, but used to be limited by high costs for the equipment and energy demand in full-scale application [41,114,115]. Employing ultrasound, powerful cavitation bubbles, and thus, high shear forces can be generated in the substrate with a comparatively low energy input to efficiently disaggregate microbial cells and sludge flocs [45,46,116]. Ultrasonic treatment of digested sludge (DS) has shown a high potential to improve the anaerobic processes [117]. For instance, the treatment with a specific energy input of 5,000 kJ/kg_{TS} has revealed an increase in the biogas yield of up to 60% in lab-scale experiments [51].

Further experiments focusing on the treatment of waste activated sludge (WAS) with a lower energy input of 200 kJ/kg_{TS} resulted in a smaller increase in gas production of up to 12%, but demonstrated the ability to achieve a positive energy balance of the ultrasonic treatment [70]. In order to transfer the promising results from the laboratory to full-scale application, the treatment intensity of ultrasonic reactors must be further improved [74,118,119].

The cavitation intensity in a sonicated medium is highly dependent on the solids content, which leads to an attenuation of the cavitation field over the reactor cross-section [93]. Previous experiments on sound propagation in solids-rich substrates, such as WAS and DS, demonstrated that a narrow reactor gap of maximum 60 mm had a positive effect on substrate penetration and cavitation field intensity [93]. For a large-scale operation, a serial connection of tube reactors with such a narrow gap size seems to be favorable concerning an intensive sonication, good mixing between the sonication stages and its simple system configuration [98]. However, the increase in flow velocity in a small cross-section must be taken into account with the design of an optimum treatment system.

When selecting the most suitable substrate for ultrasonic disintegration, the choice is between WAS and DS. WAS provides a comparatively high TS in the range of 2 to 10 % [120]. Therefore, low volume flows are possible when aiming for high specific energy inputs despite the high TS content. Previous experiments have shown that a low TS <3% in WAS had a positive effect on the sonication result [13,121]. Unfortunately this TS is not common in many large-scale WWTPs, as the WAS is usually thickend to TS above 4 % [70]. Therefore, a TS as low as 3% can be found when sonicating DS, for which a successful treatment was reported already [49,51,110,122].

However, sonication of DS requires a high volume flow through the sonication system in order to treat a considerable portion of the digester contents at the given retention time. Sonication of an average digester of 2,500 m³ and a hydraulic retention time of 25 days would result in a volume flow of 100 m³/d or 4.2 m³/h, respectively, in full-time operation. With an assumed target energy input of about 8 kW, the question arises whether eight of the examined reactors should be operated in series, or whether a parallel operation with a resulting volume flow of about 0.5 m³/h might result in a better effect on the cavitation intensity.

The objective of a narrow reaction space and a favorable serial connection leads to significantly higher flow velocities in DS than for the sonication of WAS. This raises the question of the influence of the flow velocity on the cavitation intensity.

To identify the limits and effects of a high flow velocity of DS in the reactors, DS was used as substrate for the experiments and tested with a wide range of volume flows.

Previous studies with horn-shaped sonotrodes revealed suppression of bubble growth and a loss in cavitation intensity at a perpendicular flow velocity as high as 0.3 m/s in the acoustic field [72,79]. For the sonication of WWTP sludge at full-scale, these critical flow velocities could also occur in tube reactors. Compared to horn-shaped sonotrodes, there is nothing known about the impact of fluid velocity on the suppression of acoustic cavitation in the structurally different tube reactors. Therefore, it is of great interest at what flow rates a suppression of cavitation intensity can be expected in these systems. Furthermore, it should be scrutinized to what extent the increased viscosity of the sewage sludge influences the flow regime and cavitation field in tube reactors during sonication.

To determine the highest reasonable flow velocity, the cavitation intensity was measured in a 2-inch (2”) ultrasonic reactor with an internal diameter of 53 mm at different average flow velocities of up to 2.1 m/s. The influence of the solids content and the viscosity of the sonicated media on the cavitation intensity were investigated by the application of DS

3.3.5 Material and methods

3.3.5.1 Experimental setup

The experiment was carried out in a circulation system consisting of an ultrasonic tube reactor (chapter 3.3.5.3), a rotary piston pump, and a magnetic inductive flow meter as shown in Figure 26. The closed circular system featured a volume of 47 liters. The volume flow of 0.5 m^3/h, therefore resulted in a 4-fold circulation, and in 123-fold circulation at 16.5 m^3/h during the entire test duration of 21 minutes.

The analysis of the cavitation field was conducted by a hydrophone (chapter 3.3.5.4), centrally positioned in the reactor by a retaining tube, which was fixed to the end cap of the reactor unit. The flow rate was gradually increased by varying the speed of the pump by means of a frequency converter in a range from 0 to 16.5 m^3/h. The performance of the experiment required 30 seconds per measurement point.

With a total number of 14 measurement points (i.e., 14 different flow rates tested), one sequence required 7 minutes. As the analysis was carried out in triplicates, one full experimental run lasted for 21 minutes.

In order to exclude any influence of multiple circulations on the cavitation noise level, an intermittent test scheme was chosen. The measuring points were initially recorded with increasing volume flow. Afterwards, the measured values were re-recorded with decreasing volume flow. Subsequently, a new measurement was carried out with increasing volume flow. The three measured values obtained for each volume flow point represent the triplicates, whose standard deviation is shown in Figure 27.

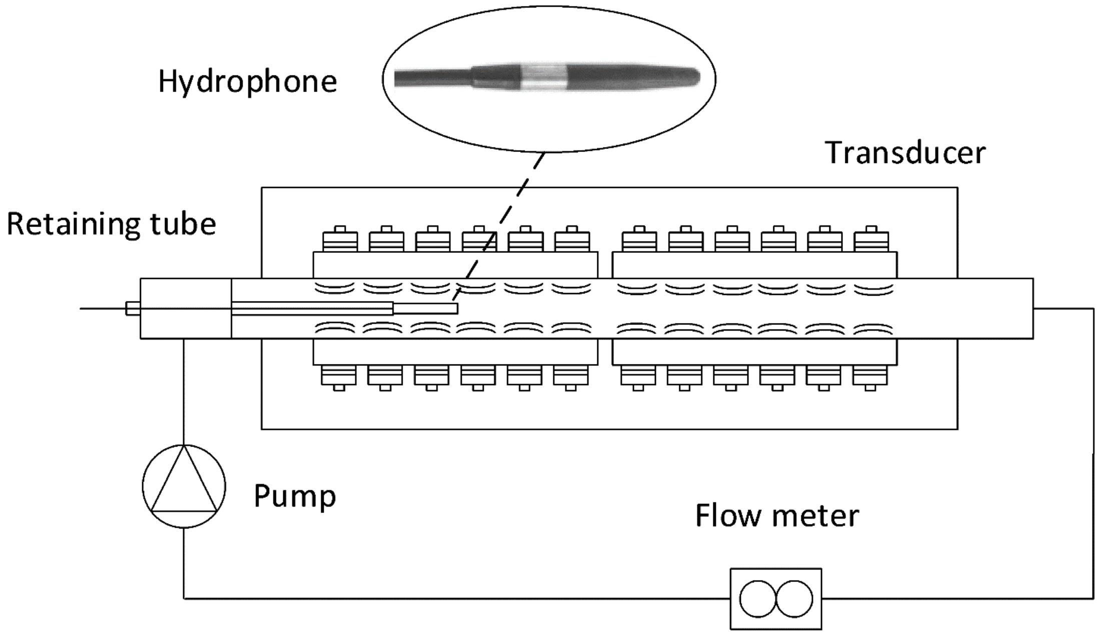

Figure 26: Experimental setup with a 2-inch tube reactor, hydrophone, pump, and flow meter (adapted from Bandelin et al., 2018).

3.3.5.2 Substrates

The examined DS was collected from two different WWTP, namely the Freising WWTP (treatment capacity of 110,000 population equivalents (PE), hydraulic retention time (HRT) of 30 d, 38°C) and from the Traunstein WWTP (treatment capacity of 68,000 PE, HRT of 18 d, 38°C), both located in southern Germany. The sludge samples were taken from the recirculation loop of the anaerobic digesters. The sludge from Freising WWTP was characterized by total solids content (TS) of 2.78 ± 0.01% and volatile solids (VS) content of 1.64 ± 0.01% relative to fresh matter weight.

The sludge from Traunstein WWTP provided a TS content of 2.52 ± 0.02% and VS content of 2.06 ± 0.03% relative to fresh matter weight. Although both WWTPs differed also related to the applied treatment schemes, the resulting TS of DS was in a similar range. This range aligns well with an international interlaboratory test reporting that the TS of digested sludge from WWTPs applied as inoculum for biochemical methane potential tests was in a range of 2.5 % (median) to 3.0 % (average) [123]. Regular tap water was used as a reference for the experiments.

3.3.5.3 Ultrasonic tube reactor

To evaluate the influence of the flow rate on the cavitation intensity, the ultrasonic tube reactor SB 8 (BANDELIN electronic GmbH & Co. KG, Berlin, Germany) with a nominal power of 1,000 W and a constant frequency of 25 kHz was used. The reactor consisted of a 2” stainless steel tube with 24 piezo-ceramic ultrasonic transducers attached on the tube with a wall thickness of 3.65 mm. The alignment of the transducers resulted in a sonication area with a length of 920 mm and a sonicated volume of 2 liters.

3.3.5.4 Cavitation measurement

The intensity of the cavitation bubbles in the reactor was measured utilizing a hydrophone according to standardized procedures [99]. The application of a TC4034-1 hydrophone (RESON A/S, Slangerup, Denmark) in the present study allowed the separate detection of the sound pressure of the ultrasonic frequency (f_0) and the emitted noise of imploding cavitation bubbles. The control unit detected the sound peak pressure in a frequency range of $2.15 \cdot f_0$ to $2.35 \cdot f_0$ and determined the cavitation noise level in decibels (dB).

The radial-sensitive hydrophone, with a diameter of 11 mm, was positioned in the reactor chamber and the medium was circulated during the measurement. Up to a flow rate of 3,000 L/h, the flow rate was increased in 500 L/h increments. Measurements above this level were conducted in increments of 1,500 L/h, whereas all recordings were conducted in triplicates.

3.3.5.5 Rheological analysis and flow calculation

The dynamic viscosity of the DS was determined by a rotational rheometer (Viscotester VT 500, HAAKE Messtechnik GmbH & Co., Karlsruhe) in combination with a cylindrical spindle of the type MV-DIN No. 7. All investigations were conducted in triplicates at a constant temperature of 20°C. Reynolds numbers (Re) were calculated for the adjusted flow rates to classify the flow regime in water. For DS, generalized Re numbers (Re_{gen}) were calculated, owing to its shear thinning flow behavior [124]:

$$Re = \frac{\rho \cdot \bar{u} \cdot D}{\eta} \quad [5]$$

$$Re_{gen} = \frac{\rho \cdot D^{n} \cdot \bar{u}^{2-n}}{K \cdot \left(\frac{3n+1}{4n}\right)^{n} \cdot 8^{n-1}} \quad [6]$$

Where ρ is the density of the fluid [kg/m^3], ū is the average flow velocity [m/s], D is the hydraulic diameter [m], η is the apparent viscosity [Pa·s], n is the power-law index [–], and K is the consistency index [Pa·s^n]. Density of the sludge was determined using a pycnometer and was found to be 1.00 ± 0.01 kg_{FM}/L for both DS samples. For the determination of K and n, the viscosity functions of each triplicate of the DS were approximated by the Ostwald-de-Waele model according to Ratkovich et al. [125] :

$$\tau = K \cdot \gamma^{n} \quad [7]$$

Where τ is the shear stress [Pa] and γ is the shear rate [1/s]. Consistency index K and power-law index n were finally obtained by fitting the model to the experimental data by use of the least-squares method.

3.3.6 Results and discussion

Cavitation intensity was measured in 42 different test configurations using a 2” tube reactor for the sonication of water and DS from two origins at 14 different flow rates. The results, displayed in Figure 25, indicate that the cavitation intensity in water and DS was significantly reduced at increased flow velocities. An average flow velocity of 0.2 m/s reduced the cavitation intensity in water by 6% compared to a sonication without perpendicular flow. Even higher reductions of 10% and 32% were observed by the sonication of $DS_{Freising}$ and $DS_{Traunstein}$ at the same flow rate.

In the present experiment, the solids content of 2.78% in $DS_{Freising}$ led to a reduction of the cavitation intensity of 38% for the case of an inactivated pump compared to the sonication of water. Regarding the large-scale application of ultrasonic disintegration of DS, a small reactor gap size is reasonable to provide an effective sonication [13], but can result in high flow velocities at high volume flows. To investigate the superposition of a high average flow speed with an increased solids content of the substrate in the present study, the sonication of DS with an average flow velocity of up to 2.1 m/s was conducted in the experiment. These operation conditions resulted in a nearly complete suppression of cavitation in the sonicated liquid.

Although the results on the cavitation-reducing effect of high flow rates could limit the design of large-scale sonication systems for continuous treatment, a positive influence of a medium flow rate on the cavitation field in DS was observed. Slightly increased flow velocities in the range of 0.8-1.3 m/s resulted in an increase in cavitation intensity of up to 9% in $DS_{Freising}$ compared to the cavitation intensity in the stationary medium. A similar effect was observed in the higher viscous $DS_{Traunstein}$, where a local maximum was observed in the range of 8.6-9.7 m/s. This can be possibly explained by the shear-thinning behavior of the DS, which leads to a reduction in viscosity when the flow velocity in the tube is increased. The cavitation-inhibiting effect of a high viscosity compared to water can thus be reduced in the DS by slightly increased volume flows.

When considering the Reynolds numbers (Re) in the DS for the present test setup and the shear rate dependent viscosity, a change of the flow regime to turbulence can be expected at much higher flow rates compared to water; i.e., at a flow rate of about 8.3 m^3/h (Table 3). In this context, it is noticeable that the highest cavitation intensity was measured in the transition range to turbulent flow (Re_{crit}). It can be concluded that the cavitation-inhibiting influence of the increased solids content in $DS_{Freising}$ can be narrowed from 38% to 19% compared to water by the operation of the sonication in this Re range.

However, a further increase of the volume flow in the turbulent range with Reynolds numbers of 3,800 to 7,300 led to a strong reduction of the cavitation intensity in $DS_{Freising}$ (-49%) as observed simultaneously at this flow rate in water (-19%) and $DS_{Traunstein}$ (-99%). For the application of the investigated 2” tube reactor for the disintegration of $DS_{Freising}$, an optimal flow rate of about 6 m^3/h and 9 m^3/h for $DS_{Traunstein}$, respectively, could be determined. When using the ultrasonic reactor for sonication of water, the volume flow rate should be kept as low as possible, since the cavitation intensity is significantly reduced by more than 13% at an average flow velocity of 0.4 m/s already. Nevertheless, in comparison to DS, there is only a slight attenuation of cavitation intensity at very high flow rates in water, so that even at average flow velocities of 2.1 m/s up to 80% of the cavitation intensity compared to the batch sonication is achieved.

Results of this study imply that in full-scale systems the volume flow in reactors with a small cross-section should be limited below 1.5 m/s when DS with a TS between 2 and 3 % is treated. For the sonication of full-scale volume flows, which would entail exceeding the turbulence threshold, a parallel operation of several sonication units is recommended.

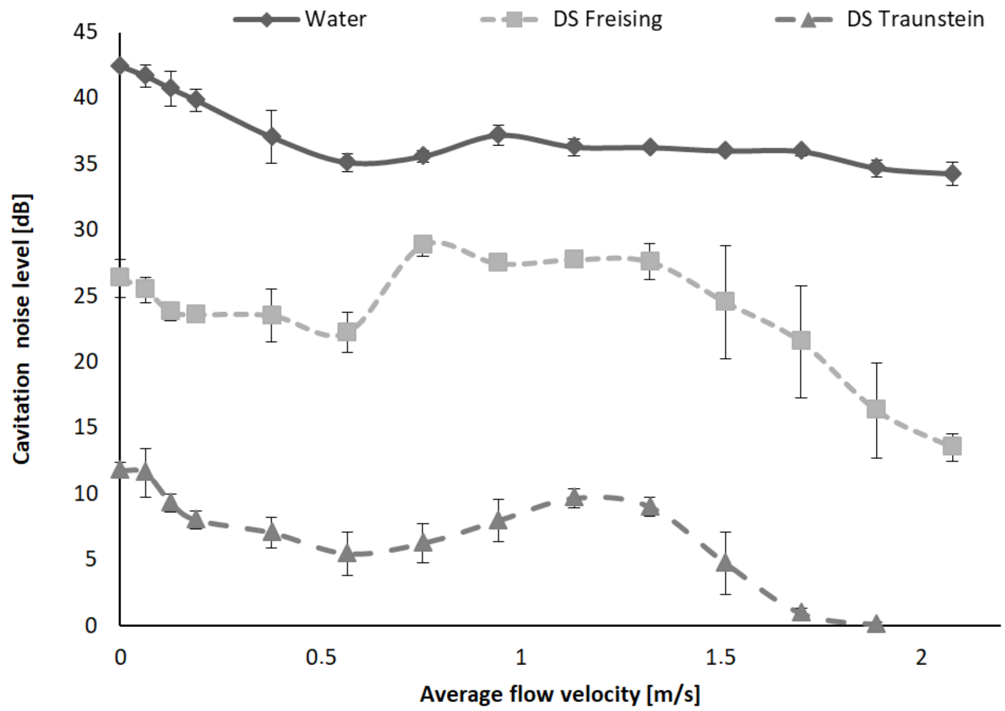

Figure 27: Cavitation noise level in water and digested sludge from Freising and from Traunstein in relation to the average flow velocity. Error bars denote standard deviation.

Table 3: Average flow velocities and Reynolds numbers at different flow rates of water and digested sludge in a 2-inch-tube reactor. The grey marked fields indicate the area with laminar flow regime in the tube.

Flow rate [m³/h]	Average flow velocity [m/s]	Re in Water [-]	Re_{gen} in $DS_{Freising}$ [-]			Re_{gen} in $DS_{Traunstein}$ [-]		
0	0	0	0	±	0	0	±	0
0.5	0.06	421	1	±	0	0	±	0
1.0	0.13	841	8	±	0	2	±	0
1.5	0.19	1262	24	±	1	8	±	0
3.0	0.38	2523	146	±	7	53	±	2
4.5	0.57	3785	421	±	21	164	±	7
6.0	0.76	5046	889	±	42	368	±	14
7.5	0.94	6308	1589	±	74	687	±	25
9.0	1.13	7570	2554	±	118	1146	±	40
10.5	1.32	8831	3815	±	173	1766	±	59
12.0	1.51	10093	5399	±	243	2569	±	83
13.5	1.70	11354	7336	±	326	3574	±	111
15.0	1.89	12616	9649	±	425	4803	±	144
16.5	2.08	13878	12365	±	540	6276	±	183

3.3.7 Conclusions

This study investigated the influence of volume flow on the cavitation intensity in an ultrasonic reactor for the sonication of DS for the first time. The results indicate that the cavitation intensity is weakened at high Reynolds numbers; i.e., turbulent flow conditions in the medium. However, it should be noted that the flow of DS through the test chamber in the transitional range led to an increase in cavitation intensity. The results provide valuable insights for the design of sonication systems intended for the ultrasonic treatment of DS in WWTP.

3.3.8 Declaration of interests

The authors declare that they have no known competing financial interest or personal relationships that could have appeared to influence the work reported in this paper.

3.3.9 Acknowledgments

The authors gratefully acknowledge the funding of this study by the Federal Ministry of Economics and Technology (BMWi), Grant 03ET1396A.

4 Overall discussion and full-scale application

The experimental results from **Paper #1** were used to determine a suitable reactor concept for sonication of large quantities of sludge. The main finding from **Paper #1** that a limited gap size provides a positive effect on the cavitation intensity suggests the use of a box-shaped reactor, which is characterized by a large cross-section and a small gap at the same time (Figure 27).

The inlet and outlet could be implemented by an attached collector tube, which provides the same cross-section as the reactor chamber. The passages should be slightly inclined to the reactor chamber to achieve a favorable, even flow distribution. This concept for an ultrasonic reactor was pursued and developed further by the use of cross-shaped reinforcement by steel profiles, welded to the reactor outside to strengthen the sound emitting surface against pressure loads. The remaining surface was equipped with 30 piezoceramic oscillation systems on each side. The reactor thus achieved a nominal ultrasonic power of 3,000 W.

The alternating combination of two reactors results in a symmetric shape and would allow for installation in an existing, straight pipeline without any additional tubing. The test procedures to adjust the resonant frequency of the ultrasonic generators revealed a high cavitation intensity and reliable operation of the new reactor system, in terms of mechanical stability, with the sonication of water. During sonication of WAS in a suitable test setup, only a slight pressure increase of less than 0.4 bar was observed with a volume flow of 1 m^3/h and an open outlet without counter pressure. In order to fulfill the further precondition of an intensive sonication from **Paper #1**, the increase of the static pressure to about 1 bar in the reaction chamber, precautionary force simulations have been performed with the enforced surface (Figure 29).

With regard to the use of the reactor in the main line of WAS in the Starnberg WWTP, a further increase in pressure of up to 4 bar had to be assumed. Therefore the completed reactor system was subjected to an enhanced pressure test, in which cracks appeared at the reinforcement angles.

With respect to the static pressure sensitivity of a flat surface, the box-shaped reactor concept was therefore no longer considered for use on an industrial scale.

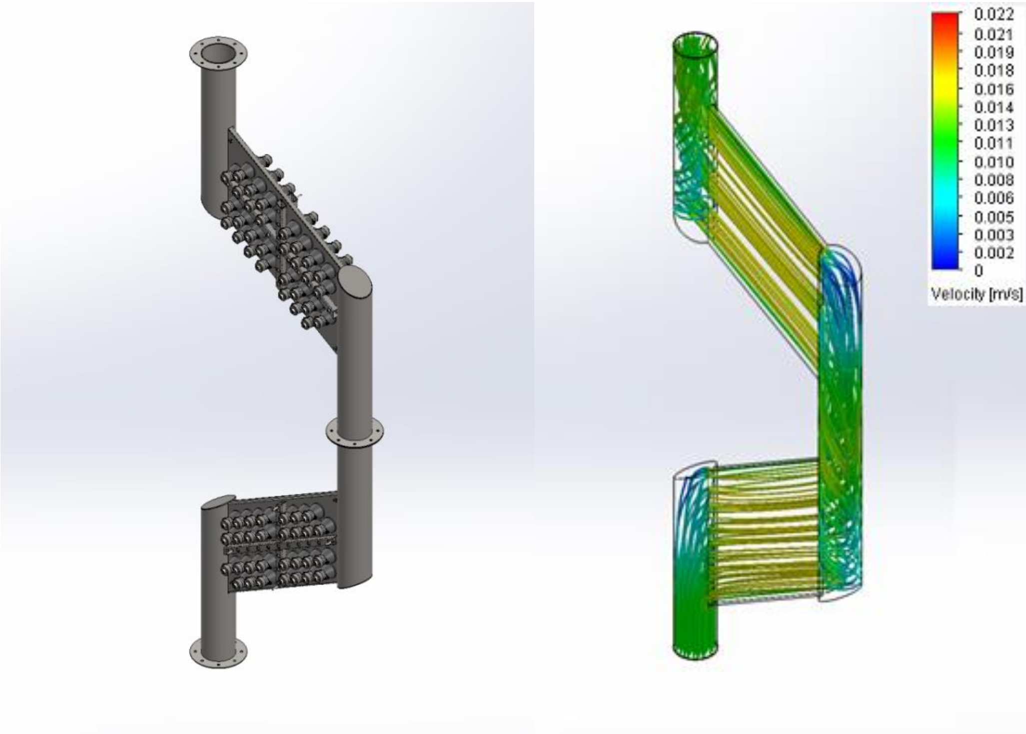

Figure 28: Drawing of two assembled flatbed reactors (left) and flow simulation (right).

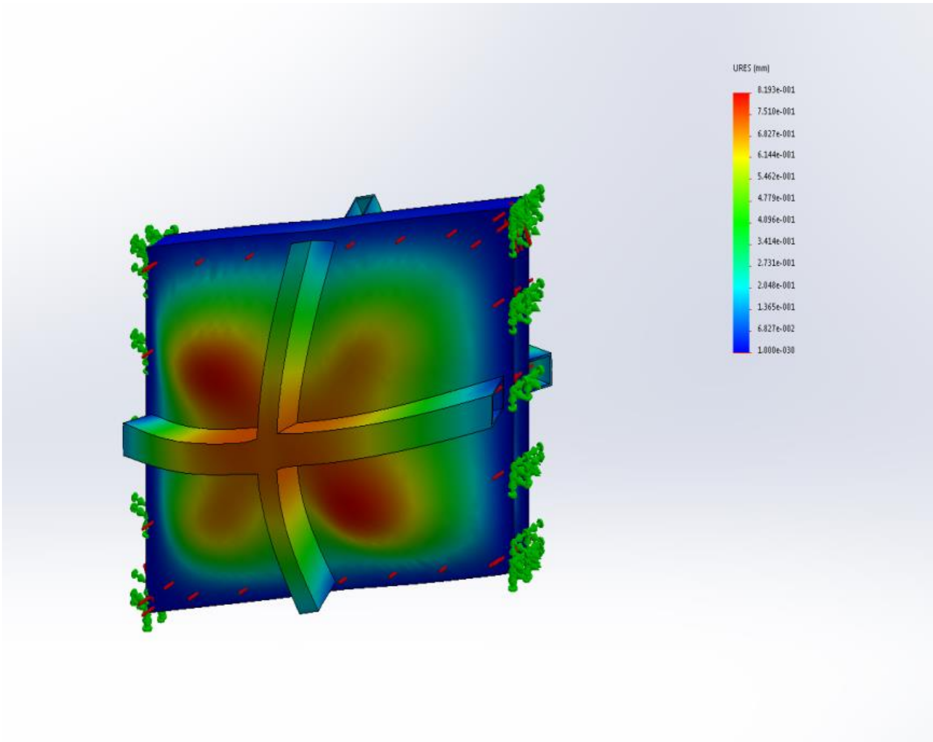

Figure 29: Pressure and deformation analysis for a reinforced flatbed reactor.

To avoid permanent deformation caused by overpressure during operation, a circular design was therefore considered in further studies. In order to ensure a small gap distance of less than 60 mm for a large tubular cross-section, a 164 mm strong tube was combined with a 50 mm strong inner tube. This inner tube was subjected to excitation by two 750 W oscillation systems and increased the cavitation field within the reactor.

The installation of tube elbows at the end of the tube allowed the central positioning of the inner oscillation system, which resulted in a symmetric space between the sound emitting surfaces. Due to the meandering arrangement of the inlet and outlet, a uniform flow profile could be achieved over the length of the reactor (Figure 31). The sonication volume of the double-tube reactor was determined to be 20 Liters and resulted in a sonication intensity of 222 W/l. When sonicating sewage sludge, gases can be expelled and lead to the dry running of the ultrasound emitting surface. This can result in an overheating of the transducers and result in damage to the piezoceramic disks.

To ensure the reliable removal of gases, the reactors were aligned in a vertical position by a rectangular support frame (Figure 30). The Starnberg WWTP was selected to evaluate the novel ultrasonic system at full-scale operation due to the parallel operation of two digesters of which one could be supplied with the treated substrate, and the remaining digester could serve as an untreated control. In this plant system, the flow rate of WAS is up to 4.5 m^3/h during the daily operation of 8 hours, and the two digesters are fed in alternating order with sequences of two hours. The operation of the dewatering system is limited to the daily opening hours of the WWTP to allow for intervention in case of malfunction of the belt press.

In order to achieve a specific energy input of 200 kJ/kg_{TS}, which turned out to be expediently in preliminary tests, three of the designed reactors were connected in parallel, resulting in an average flow rate of 1.5 m^3/h within each reactor. Operation conditions of the digestion process, as well as the TS and VS of the inserted WAS, and primary sludge, were monitored during the test course. The TS, viscosity, dewaterability, and the specific gas production of each digester were analyzed and compared, with a sample rate of one week. To identify the possible differences in the digestion efficiency of each reactor, the test procedure was started two hydraulic retention times earlier than the beginning of the sonication. Detailed results of the operation of the ultrasonic system over a period of 292 days, including more than 7 hydraulic retention times of the digesters, were published in [126] and can be found in the appendix.

In the course of the experiment, it became apparent that the sonication of WAS at full-scale could only have a minor impact on the gas yield, and the promising batch test results could not easily be transferred to large-scale by increasing the size of the reactor. A number of factors were taken into account as an explanation of this observation. First of all, the sonication of WAS reaches only 1/3 of the material stream inserted into the digester, and the ultrasonic treatment cannot affect the primary sludge. An additional factor for the limited effect of the sonication of WAS is the short operating time of the feed pump and thus of the ultrasonic system of 8 h per day. This limits the specific energy input to the sludge, while the system uses only a third of its theoretical daily capacity.

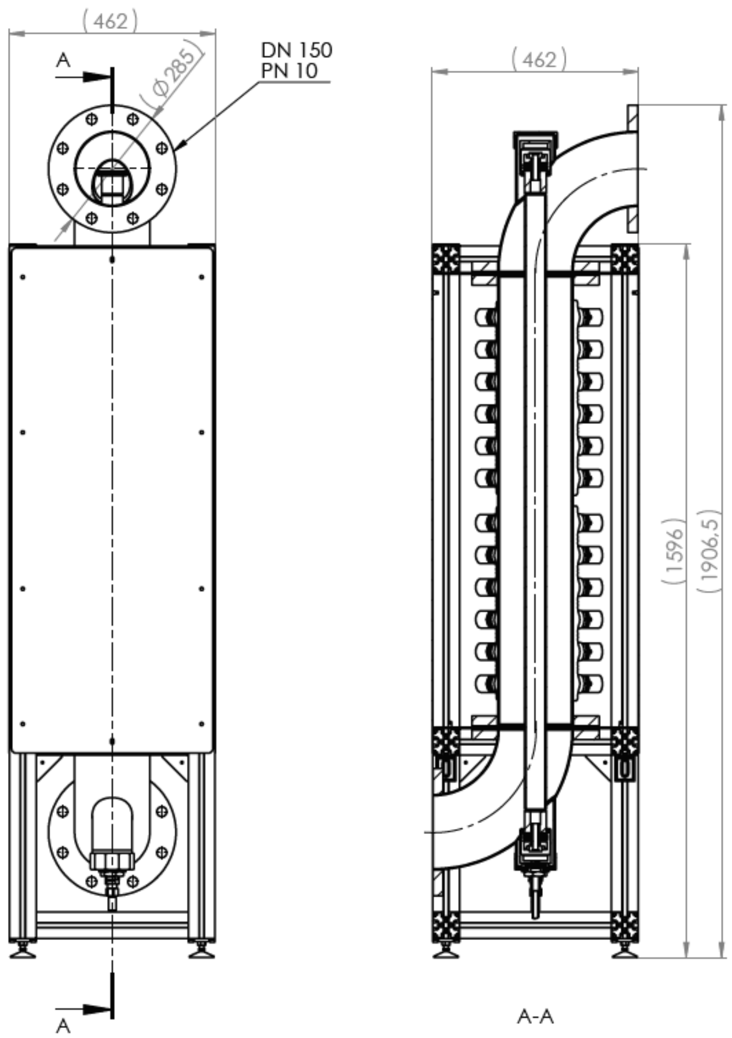

Figure 30: Scheme of the double tube reactor.

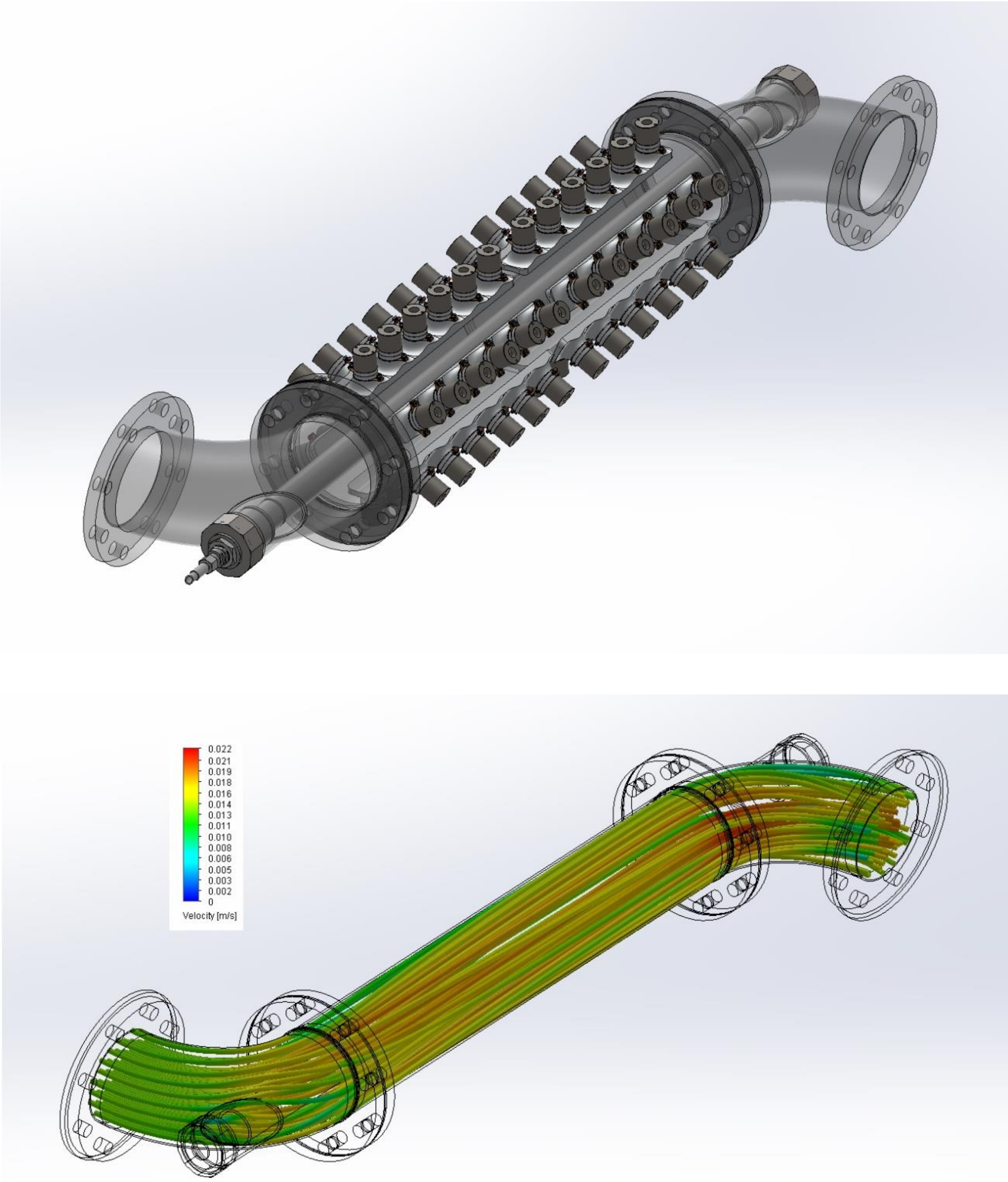

Figure 31: Drawing and flow simulation of the double tube reactor.

Sonication of WAS was therefore compared in **Paper #2** with the sonication of the possible alternative digested sludge at a technical-scale. In addition, different reactor concepts were taken into account to examine the influence of certain aspects of the sonication system on treatment efficacy. The results demonstrated that the sonication of DS has a number of advantages over the sonication of WAS. These include the significantly lower TS, which leads to a significantly higher cavitation intensity at the same energy input. In addition, the sonication of digested sludge revealed that a significant relative increase in gas yield could be achieved in the batch test even at low energy inputs of about 300 kJ/kg_{TS}. Additional tests in a lab-scale digester system with the ability to run in the continuous operation confirmed the positive effect of sonication of DS [73]. The significantly lower TS of DS results in a possible volume flow in the reactor of almost twice as high as in WAS at the same specific energy input. Additionally, the test results from **Paper #1** and **Paper #2** demonstrated the positive influence of small gap dimensions in the sonication of media containing solids.

The experimental reduction of the gap size and simultaneous increase of the reactor capacity led to a number of design challenges. These included the reduced compressive strength when using box-shaped sonication systems, as well as an increase in the mass of the reactors, which made production and assembly more complicated. In addition, the increase in the size of the sonication systems led to a deterioration in maintainability because if a particular oscillation system became defective or worn out, the entire sonication system had to be replaced.

The use of a large number of smaller tube reactors solves the difficulties described above and additionally simplifies the standardization and series production of the ultrasonic systems. On the other hand, the use of several small ultrasonic systems increases the number of connected pipes and installation work for the power supply. If a large number of small reactors are connected in parallel, the sludge must be distributed evenly over the reactors used. Due to the inhomogeneity and the risk of deposits, the volume flow of each line would have to be measured and permanently readjusted by automatically controlled control flaps. A much simpler approach is to install the reactors in series, which results in the same energy input and retention time as a parallel connection.

In addition, with the sonication of digested sludge on a laboratory scale and low flow rates, the series connection showed no difference to sonication in parallel connection concerning the increase of the methane yield [51]. On the other hand, this setup leads to an increase in the average flow velocity in the reactor with the sonication of larger amounts of sludge, and the cavitation intensity was reported to be disturbed by the introduced flow in previous experiments [72].

In order to examine further optimization of the plant design by connecting a large number of small reactors with a diameter of 50 mm in series (Figure 32), the damping of the cavitation intensity at a wide range of different flow rates was investigated in **Paper #3**.

The results revealed that the cavitation intensity in water is reduced with an increasing flow rate. Reduction in intensity was also observed in the sonication of DS, but an average flow velocity in the range of 0.8 to 1.3 m/s revealed a positive influence on the cavitation intensity, which can be explained by the shear-thinning properties of the sludge. Very high flow rates in the turbulent range (Re > 2,300) led to a strong reduction of the cavitation intensity in the reactor, with all media examined. With regard to sonication at the full-scale range, the results imply that sonication of up to 8 m^3/h with a specific energy input of 300 kJ/kg_{TS} can be realized with a series connection of 20 of the investigated 2-inch reactors to a simultaneously intensive and efficient sonication of a large quantity of DS.

Based on the positive results, an ultrasonic system was installed in the circulation loop of the digester at the Traunstein wastewater treatment plant. For this purpose, a side branch and an additional pump were installed on the suction side of the circulation pump. The medium was passed through four reactors with a diameter of 150 mm each and additionally through four reactors with an inner diameter of 50 mm. The nominal power of the setup reached 20 kW and was continuously operated with a volume flow of 5 m^3/h, which resulted in a specific energy input of 300 kJ/kg_{TS}. Operation of the described setup in Traunstein WWTP revealed a stable and continuous operation of the system, but due to technical failures of digester components, no statement about the effects of the ultrasonic treatment can be made yet.

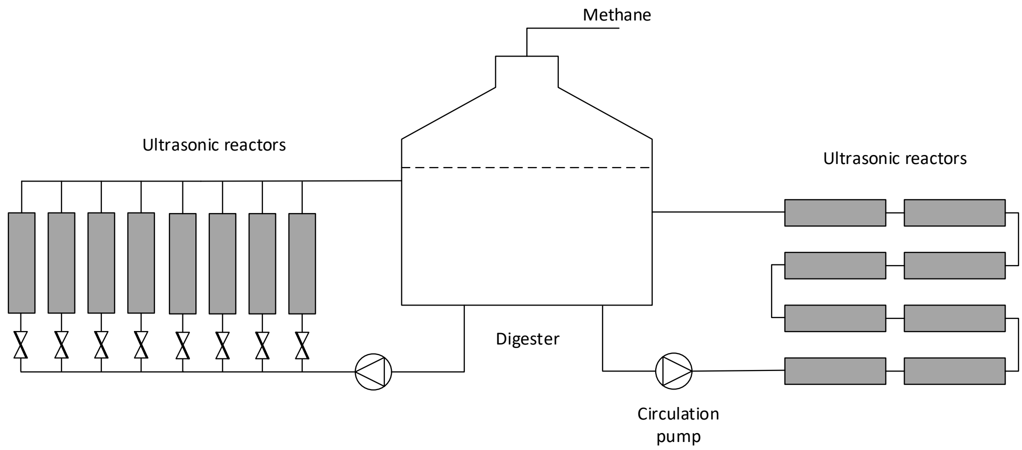

Figure 32: Setup of multiple ultrasonic reactors in parallel (left) and serial connection (right)

Overall it can be concluded that the application examined in this study, consisting of the sonication of WAS and DS, requires a high energy input as a consequence of the comparatively high solids content. The tolerance of the process regarding a reduction of the power density, especially in the sonication of WAS, was observed in **Paper #1** to be very limited. Also, a reduction of the specific energy input by shortening the sonication time in **Paper #2** had a significant influence on the desired increase of the methane yield and other parameters investigated. The reduction of the TS of the medium to be sonicated by sonication of DS instead of WAS led to a higher cavitation intensity documented in **Paper #2** but could not show a long-term increase of the gas yield at lab-scale.

The application of the technology at full-scale conditions for sonication of WAS showed that in order to achieve a significant influence on the digester operation, a larger amount of the digester content would have to be treated than possible with the sole and single sonication of WAS. However, the results from **Paper #3** demonstrate that the restrictive conditions revealed in **Paper #1** and **Paper #2,** as well as in the large-scale tests, can be technically addressed. A possible approach recommends the use of a large number of small sonication units in the most favorable series connection for sonication of the digestate content. With this design, a suitable sonication time can be realized with a small reaction gap size and thus high sonication intensity.

5 Recommendations for future studies

5.1 Optimization of sewage sludge treatment

The mechanisms and effects of sonication of WWTP sludge described in this thesis provide an overview of the potential of the treatment and offer suggestions about an increase in treatment efficiency depending on the sludge properties and reactor design. Although an increase of methane yield was observed after the treatment of WAS at lab-scale, the application of the technology at an average WWTP did not result in an economically feasible improvement of the methane production. The small impact of the applied ultrasonic energy can be explained by the high energy absorption of the particles in the substrate due to high dry solids content of up to 7% and a very long HRT of more than 40 days. A reduction of the TS by the adjustment of flocculant dosage would save the cost of the necessary chemicals and improve the cavitation intensity during ultrasonic treatment. On the other hand, the reduction of the TS in the WAS input stream of the digester would increase the amount of water in the digester and reduce the retention time of the sludge particles. This could result in partial digestion and reduction in methane yield, which would oppose the aspired increase in energy recovery.

Sonication of DS with its comparable low TS of 2.5 to 3.0 % could solve the challenge of high ultrasound absorption in WAS. In lab-scale experiments, the sonicated DS revealed an impressive relative increase in gas production. On the other hand, sonication of already pre-digested sludge only resulted in a short term increase, which declined drastically after a few days. Also, the absolute increase of the gas production after sonication of DS was smaller compared to the same volume of WAS. The behavior of a significant increase in the short term offers the potential for an investigation in a wastewater treatment plant with high utilization of the digester capacity, and therefore, short HRT. Another optimization of the sonication systems design could be the combined treatment of WAS and DS by the dilution of WAS with digester content prior to sonication. In many WWTPs, WAS and primary sludge are injected into the circulation line of the digester.

The equipment of the circulation line with the ultrasonic devices would improve the mixing and ensure optimized pre-treatment of the different sludge types immediately before digestion by the combination of the benefits of the different sludge types.

5.2 Improvement in cavitation erosion resistance

The intense implosion of the cavitation bubbles is intended for the mechanical disintegration of the sludge particles and the release of sludge components. On the other hand, however, the sound-emitting surface of the oscillation system is also affected by the cavitation bubbles and is eroded over a longer period of time (Figure 33). While moderate erosion on the surface of an oscillation system is part of the usual wear and tear [127], the media impacts the degree of erosion of the oscillating surface. Reasons for premature wear of the oscillating system can be aggressive components of the sonicated medium, such as chloride ions [128]. Also, the mechanical stress of the surface by particles remaining on the surface can lead to increased erosion in association with the oscillating surface [129].

In the context of the sonication of highly viscous and solid containing media such as sewage sludge, particles also come into contact with the oscillating surface. The increased viscosity compared to water represents an increased resistance of the abrasive medium. During the practical testing of the double-tube reactor in the Starnberg WWTP, a pronounced cavitation erosion was observed on the sound-emitting surface. The tested tube reactor, with a thickness of 2.6 mm, was exposed to severe damage during the given period of operation. However, this observation could not be reproduced with a similar power density in the laboratory when using regular tap water and the observed cavitation erosion was significantly lower in this case. This observation illustrates the increased susceptibility to wear of the sound emitting surface during sonication of WAS. The influence of the viscosity, the TS, or the chemical composition should therefore be investigated in further studies. One approach to prevent excessive cavitation erosion is to vary the intensity distribution in order to achieve a uniform wear and tear of the reactor surface. Another attempt consists in coating the reactor surface to increase the strength of the sound emitting surface against erosion.

For this purpose, hard chromium plating represents a target-oriented approach, which must, however, be considered against the additional costs for the coatings [130]. The reduction of the TS and thus improvement of the sound field distribution is additionally suggested as a process engineering option for excessive cavitation erosion.

Figure 33: Example of cavitation erosion in a rectangular ultrasonic reactor with a gap size of 25 mm.

5.3 Alternative sonication objectives

5.3.1 Sonication for decreased sludge disposal costs

Additionally to the increase in methane yield examined in this study, the reduction of the residues to be disposed of by improved anaerobic digestion represents a potential application scenario for ultrasonic disintegration [131]. Due to the further increase of regulations regarding the maximum permissible content of hazardous substances and the resulting decline in the use of sewage sludge in agriculture, the cost-intensive incineration of the residues of wastewater treatment evolves into a critical cost factor for municipalities [132].

Besides the reduction of biomass as a result of its conversion into methane, the improvement of the dewaterability is the main factor for lowering the costs for transport and disposal of the waste [133]. Separation of the remaining biomass after anaerobic digestion requires effective flocculation, which is supported by the addition of polymers to the digested sludge.

The floc formation in the residual sludge was reported to be additionally improved by the specific use of ultrasound directly before the dewatering process [134]. The result of the ultrasound application is substantially dependent on the energy input and can both improve and deteriorate the drainability of the sludge.

At medium energy input, the cell membrane is perforated to the extent that natural floc forming extracellular polymeric substances (EPS) are released [135,136]. With increased ultrasonic intensity, however, a decline in floc formation was observed, which can be explained by an intensive homogenization of the biomass and destruction of the floc structure [131]. The application of the technology at an industrial scale has not yet been reported and offers the potential for further investigations. The extent to which the optimal sonication intensity can be influenced by the design of large-scale reactors in terms of power density and sonication time has not yet been fully investigated and will be the precondition for further development of the technology.

5.3.2 Sonication for sludge component recovery and removal

While the particular focus of this study was on the improvement of the energetic use of the residues from wastewater treatment, the separation of the sewage sludge components for recycling is another possible application [137]. This includes the recovery of nutrients through the cultivation of microalgae in the wastewater [138,139]. The subsequent utilization of the produced biomass is performed by the disintegration of the cells and the release of its constituents containing lipids, proteins, and carbohydrates. This process has already been improved by the use of ultrasonic disintegration at a lab-scale. By the single-use of ultrasound, a release of proteins of about 99% from algae called *Scenedesmus obliquus* could be achieved with an energy input of 200 kWh/kg_{TS} [138].

Also, the removal of phosphorus from the wastewater by enhanced biological phosphorus removal (EBPR) was reported to be enhanced by the use of low-intensity ultrasound [140]. Due to the presence of heavy metals such as Cu, Zn, and Pb in the municipal wastewater, it is becoming increasingly difficult to recycle sewage sludge in agriculture for fertilization due to concerns about a long term accumulation of the heavy metals in soil [132].

One approach is to separate the hazardous substances from the sludge to prepare reuse in plant cultivation. The extraction of the problematic substances can be fulfilled by acid extraction by the use of nitric acid or citric acid [141]. Due to the slow transfer of the substances into the solution, treatment times of up to 11 days were observed at lab-scale with the extraction of Pb in citric acid with a pH of 2.33 to achieve the highest possible extraction rate of 95% [142]. The extraction rate of Pb with nitric acid has been increased by the use of ultrasound, which would lead to a reduction of the costs for the chemical reactants and treatment time in large-scale industrial applications [143].

Ultrasonic disintegration has also been successfully used for the extraction and recovery of enzymes like lipase and protease from WAS at lab-scale [144]. The regained amounts of enzymes can be used for advanced processes in the transformation of plant oil into biodiesel [145]. Though a number of promising approaches for the recovery or separation of various sludge components are already available, the transfer of these approaches to full-scale requires the solution of various technical challenges and reveals a considerable need for further research.

5.4 Alternative ultrasonic applications

5.4.1 Use of ultrasound in sonochemical organic pollutant removal from water

The use of ultrasonic reactors for sonochemical reactions, especially for the oxidation of pollutants dissolved in water by advanced oxidation processes (AOP), represents a further application of the technology in wastewater treatment [146]. Various mechanisms can be utilized to achieve this objective. These include the generation of $^{-}$OH radicals, which enhance the oxidation of certain organic substances, such as medication residues or pesticides. Also very high-temperature peaks of collapsing cavitation bubbles can improve the degradation of temperature-sensitive substances. This is particularly relevant for per- and polyfluoralkyl substances (PFAS), which are very difficult to degrade in nature and were used in the past for the production of coatings or as foaming agents [147].

While the use of intensive ultrasonic cavitation enhances the degradation of organic substances in laboratory, the very low concentrations and correspondingly low energy efficiency of an industrial-scale implementation represent an obstacle to an application at full-scale wastewater treatment plants [148,149]. Possible approaches to a successful implementation consist of the concentration of the pollutants by upstream cleaning processes such as reverse osmosis or the use of the technology in applications with a very high pollutant concentration, for example, in the treatment of industrial wastewater or in the remediation of contaminated sites.

A further use case is the combination of ultrasonic induced cavitation with other oxidative processes such as photocatalysis [150]. Here, the solution to be treated is passed over a catalyst layer, e.g., of titanium oxides or zinc oxides. The catalyst is exposed to intensive ultraviolet radiation to trigger the reaction. In order to ensure an optimal mass transfer at the catalyst interface, the reaction volume is additionally exposed to ultrasound so that an optimal flow of the reactive catalyst surface with a reactive medium is enabled. For the development of suitable ultrasonic systems, the relationships examined in this thesis can be relevant for the achievement of an optimal ultrasonic intensity with regard to an enlargement of the reactor systems and an increase in the flow rates.

However, further investigations are necessary to determine which cavitation intensities can achieve the highest energy efficiency for the respective degradation process.

5.4.2 Ultrasonic treatment of nitrifying bacteria

While the sonication of WAS and DS was intensively investigated in this study, a further step of wastewater treatment comes into closer focus for the use of ultrasonic reactors. This concerns the removal of ammonium (NH_4^+) in the incoming wastewater. The most common process for this treatment step is the oxidation of ammonium to nitrite (NO_2^-) by ammonia-oxidizing bacteria (AOB) and subsequent oxidation to nitrate (NO_3^-), which is conducted by nitrite-oxidizing bacteria (NOB) under aerobic conditions [151]. The generated nitrate is transferred in a microbial denitrification step to gaseous nitrogen and released from the wastewater cleaning process to the atmosphere.

Sidestream sonication of the circulating sludge led to an increase in the efficiency of the process and could be an option for further optimization [152]. On the other hand, the process of nitrification and denitrification requires high degree of aeration, which represents a large part of the energy demand of conventional wastewater treatment [153]. In addition, the emission of nitrous oxide, which is considered a highly intensive greenhouse gas, as a side product of the microbial process is gaining more and more attention in the efforts to reduce the total climate-impacting emissions [154].

Autotrophic nitrogen removal is, therefore, a promising alternative to conventional nitrification/denitrification [151]. In this process, AOB convert only a part of the ammonium in the wastewater into nitrite. In the anaerobic ammonium oxidation (ANAMMOX) reaction, the remaining ammonium is subsequently oxidized by the nitrite [153]. Although this process has significant advantages in terms of energy consumption and climate gas emissions, the startup and stable operation of the process was a major challenge for large-scale application due to the slow growth of the bacteria and long setup times of up to one month [151].

The main difficulty is to promote the growth of AOB and, at the same time, suppress NOB, which would lead to the undesired oxidation of nitrite. The specific influence on the bacterial composition during the startup phase can be achieved by generating mesophilic temperatures between 30-40 °C, low oxygen, and pH control [155].

However, this process takes place at a limited speed and makes it difficult to restart the process after technical failures at full-scale application. The use of ultrasound with a frequency of 20-40 kHz showed a very effective stimulation of the growth of the desired AOB and suppression of NOB in a series of experiments [155,156].

This mechanism resulted in a reduced startup time of the partial nitrification process within 18 days and improved the stability against temperature variation due to changing external conditions [157]. Using ultrasound technology to supress the growth of NOB offers a number of advantages over the disintegration of WAS and DS considered in this work. First, a comparatively low energy density of 0.2 W/cm² was observed in the laboratory experiments to achieve the effect [152].

Second, the medium to be sonicated has a lower solid content and thus lower attenuation of the sound waves, which improves the generation of cavitation bubbles.

A further advantage of the application is the only temporary need for sonication in the starting phase [158]. When the desired process parameters are reached, a reduction of the use of ultrasound and partial load operation for process stabilization can, therefore, be assumed as suitable process control in large-scale implementations [158]. This can help to reduce the required energy consumption as well as the wear of the ultrasonic system.

The knowledge gained and methods developed in this work for measuring and optimizing the intensity of ultrasonic reactors can also be used for the development of large-scale reactors for sonication of NOB. This concerns, for example, the variation of the gap size of ultrasonic reactors as well as the direct measurement of the ultrasonic intensity in the respective medium. For an optimal adjustment of the cavitation intensity and retention time in the ultrasonic sonication system, further investigation of the sonication parameters required for the process is recommended.

5.4.3 Sonication of suspensions

Observations on the optimized generation of intensive cavitation fields in media with solid particles can be transferred to further process engineering applications with the use of ultrasonic reactors as well. A relatively new approach is the ultrasonically assisted recycling of slag from waste incineration plants [159]. After the incineration process and the separation of the metallic components, as well as glass, an inorganic residue of gravel and sand remains in the slag. The material is contaminated on the surface with chloride, sulfate, and heavy metals [160]. Contaminants restrict the further use of the material in road construction due to the threat of leaching and groundwater contamination. On the other hand, cleaning and reuse of the material as a base material for concrete instead of landfilling can reduce both the need for landfill capacity and the consumption of globally limited gravel sources [161,162]. The precondition for the reuse of the material is an effective disintegration of the agglomerated particles and removal of the pollutants by the transfer in a washing solution [160].

The dissolved substances can subsequently be separated and disposed from the washing solution in a closed circuit.

The use of ultrasound-induced cavitation represents a solution approach for the effective disintegration and cleaning of the slags [159]. An intensive ultrasonic field has to be applied to sedimenting suspensions of different particle size distributions. At the same time, the energy requirement and investment volume needed for an economically attractive implementation must be as modest as possible. The experiments carried out in this study reveal fundamental effects in the sonication of suspensions and can also be applied to a certain extent to the processing and reuse of slags. These effects include the adjustment of the reactor space, which allows for a concentration of the ultrasonic power and compensation of the absorption of the particles. Also, the increase in static pressure to 1 bar and the associated increase in cavitation intensity are not limited to sewage sludge treatment but can also be transferred to this application, where intensive treatment is necessary.

6 Overall conclusion and outlook

The experiments performed within the scope of this thesis demonstrate both the potential and the limitations of sonication of WWTP sludge to increase the methane yield. Sonication of WAS with a low energy input of 300 kJ/kg_{TS} showed a positive additional methane yield in the batch test, with all ultrasonic devices investigated. However, transferring these observations to large-scale involves several limiting conditions as the side stream sonication of only one part of the digester's feed stream, leaving out the primary sludge as well as the only temporary operation of the system during the daily operating hours of the dewatering application, which limited the impact of the technology.

In particular, the high TS of WAS leads to an intensive absorption of the ultrasonic power. This requires focused reactor concepts designed for the properties of the treated substrate. The sonication of DS, on the other hand, demonstrated a significantly higher relative increase in the methane yield in the short term of 5 days after sonication, but it has to be admitted that this advancement was largely diminished after a longer retention time in the batch test of up to 30 days. These observations allow for the conclusion that digester with high capacity utilization and, therefore, short hydraulic retention time would be more amenable to ultrasonic treatment. The resulting large flow velocities in focussed ultrasonic reactors were considered and investigated, and the observations imply that the sonication efficiency is increased with moderate flow velocity and the flow regime should be below the turbulence threshold.

However, in plants with high capacity utilization and correspondingly short hydraulic retention times, sonication of DS could make a positive contribution to increasing the energy production.

Overall, the experiments carried out in the study provided insight into the fundamental interrelationships, but opened up new questions such as how to effectively improve the durability of the sound emitting surface and how to increase the impact of the sonication and the treated substrate. These topics as well as the use of ultrasound disintegration for additional applications should be part of further investigations.

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Appendix

Full-scale assessment of ultrasonic sewage sludge pre-treatment using a novel double-tube reactor

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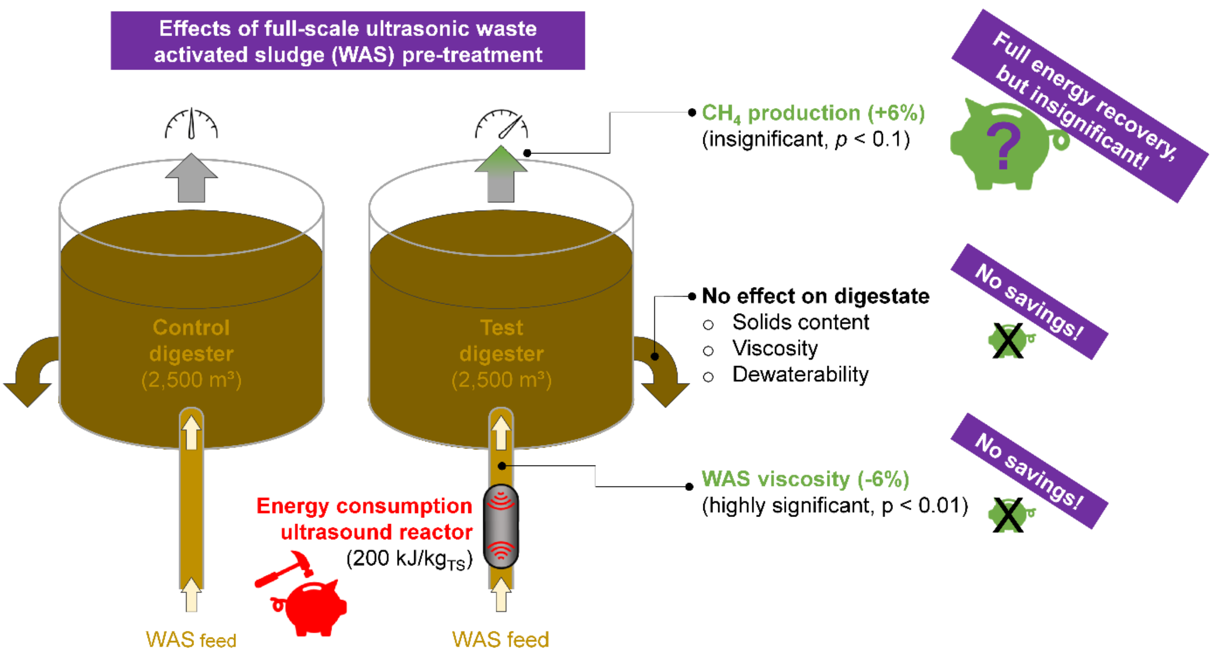
Keywords

Ultrasound pre-treatment, sewage sludge, biogas production, sludge viscosity, full-scale assessment

Highlights

- Full-scale assessment of ultrasound (US) pre-treatment of waste activated sludge
- Enhancement of CH_4 production by trend (by a maximum of 6.2%)
- Significant reduction of waste activated sludge viscosity (by 5.8% on average)
- No effect on dewaterability, solids content, or viscosity of the digestate
- Already small gains in CH_4 production compensate the US reactor´s energy demand

Graphical abstract



Abstract

The performance of a novel double-tube ultrasound (US) reactor for waste activated sludge (WAS) pre-treatment was assessed at a full-scale wastewater treatment plant (WWTP). For high transferability of the results, a well-performing WWTP with industry-typical operating conditions was selected for the test. The effects of the treatment (conducted at a specific energy input of approximately 200 kJ per kg of total solids) were monitored regarding improvements in (i) sludge viscosity, (ii) methane production, (iii) biosolids removal, and (iv) digestate dewaterability. The pre-treatment caused a significant reduction of WAS viscosity (-5.8% on average, at $p < 0.01$) and a maximum, yet insignificant increase in methane production by 6.2% (at $p < 0.1$). No effect was observed for solids content, viscosity, or dewaterability of the digestate. The economic benefit of the reduced WAS viscosity was insignificant, as the reduced pump energy demand was less than 1‰ of the energy consumption of the US reactor. Surplus methane production, on the other hand, was able to overcompensate the energy demand of the US reactor already at low percentage increases. However, considering improved methane production in the economic assessment remains somewhat speculative given the only insignificant increase.

1. Introduction

Ultrasound (US) can be utilized in wastewater treatment as a mechanical pre-treatment method for sewage sludge disintegration [1,2]. Hereby, the phenomenon of transient acoustic cavitation is utilized, which is defined as successive formation, growth and implosion of acoustic micro-bubbles present in the sludge [3]. Through the severe violence of the bubble collapse (creating hot spots with temperatures of 4,000 - 15,000 K [4] and pressures of up to 1.3 GPa [5]), a shear-induced disintegration of the sludge is attained, which results in a de-agglomeration of sludge flocs (sonodispergation) and a lysis of cell walls (sonolysis) [6,7]. In consequence, the initial and often rate-limiting hydrolysis step of anaerobic sludge digestion can be accelerated, leading to faster reaction kinetics of the whole process, enhanced methane production, and increased organic matter degradation [8–11].

Further reported merits of ultrasonic pre-treatment are, for instance, a reduced sludge viscosity and improved digestate dewaterability [12,13].

Given the various benefits, a large body of research was devoted to further advance ultrasonic pre-treatment technologies (see review papers by [8], [9], [2], or [10]). However, despite considerable achievements in the field, most of the studies were limited to lab-scale environments, which can differ considerably from the conditions prevailing in full-scale wastewater treatment plants (WWTPs). For instance, specific energy inputs for full-scale sonication at WWTPs rarely exceed 1,000 kJ per kg of total solids (TS) [14], while for lab-scale studies, a range of energy inputs between 1,000 - 16,000 kJ/kg_{TS} is generally recommended as most effective range (Carrere et al., 2016; Tyagi et al., 2014). The considerable offset is a result of the large sludge volumes that are to be treated in full-scale plants, which typically curtail treatment times in the US reactors to seconds or minutes at best [14–16]. High energy inputs of several thousand kilojoules per kilogram TS are, therefore, merely of academic interest but have not much relevance for full-scale WWTPs. For instance, when assuming a rather moderate sludge flow rate of one m^3/h and a sludge TS of 5%, an energy input of 10,000 kJ/kg_{TS} would already require an US reactor with an electrical power of 138 kW. However, such power rating would entail investment costs of roughly one million € (based on reactor costs of 7,500 €/kW, according to the manufacturer BANDELIN electronic GmbH & Co. KG, Berlin, Germany) and monthly electricity costs of over 21,000 €, when assuming an industrial electricity price of 21 Cent/kWh and 24 h of reactor operation per day. Therefore, existing full-scale reactors typically feature much lower electrical power, such as 4 kW in [17], 10 kW in [16], 15 kW in [18], and 30 kW in [15], thus leading to rather moderate specific energy inputs of 288 kJ/kg_{TS}, 720 kJ/kg_{TS}, 1,080 kJ/kg_{TS}, and 2,160 kJ/kg_{TS}, respectively, in the given example. As treatment effects typically increase with increasing energy input [2,19], transferability of lab-scale studies to full-scale applications is, hence, limited. A further difference relates to test methodology, as many lab-scale studies use biochemical methane potential (BMP) tests in batch mode to assess the effects of sludge pre-treatments. However, full-scale digesters are almost unanimously designed as continuous systems [20] so that the transferability of BMP tests to full-scale digesters is again limited

[21,22]. Last but not least, the important assessment of treatment economics and operational stability (including clogging behavior and lifetime of the sound-emitting surfaces) is difficult to obtain in lab-scale studies and requires full-scale testing.

Despite the research need for full-scale trials, only few studies are available to date. One of the best documented full-scale studies was performed by [15]) at the Ulu Pandan Water Reclamation Plant in Singapore, where raw sludge (RS) with an average TS of 1.6% (comprising primary sludge, PS, and waste activated sludge, WAS) was pre-treated before anaerobic digestion using a 20 kHz radial horn US reactor with an electric power of 30 kW. Despite a specific energy input of only 300 kJ/kg_{TS} and a relatively long hydraulic retention time (HRT) of 30 d of the sludge in the digester (potentially cancelling out kinetic effects of the treatment ([10]), the authors reported an increase in methane production between 13% and 58% relative to a control digester and a positive energy recovery of over 300%. Similarly promising results were reported by [23]), who summarized findings of several full-scale trials conducted in the UK, Sweden, and the USA. Most details were provided for a full-scale trial at the Orange County Sanitation District, USA, where pre-treatment of WAS using a radial horn reactor led to an increase in gas production by up to 50% and a slight, but significant improvement of digestate dewaterability (TS of filter cake increased by 1.6 percentage points on average). However, no information on specific energy input, sludge characteristics or digester performance before the treatment was provided. Another full-scale test was conducted by [16]) at the Bamberg WWTP in Germany by the use of two sonotrode reactors (5 kW each) installed for part-stream pre-treatment of 80% of the accruing WAS. The objective of the pre-treatment was to cope with an increase in the WWTP´s design capacity, which has led to a critical drop in HRT to only 18 days. During the full-scale trial conducted from 2004 to 2011, a constant increase in biogas production (from about 1.5 million m^3/a to 2.2 million m^3/a) and volatile solids (VS) degradation (from 34% to 50%) was observed, despite a verifiably low energy input of only 148 kJ/kg_{TS}. As no control digester was used, however, it was difficult to ascertain that the positive development was solely related to the US pre-treatment.

In contrast to the positive experiences, also information on unsuccessful trials can be found. A report from the Swedish Water and Wastewater Association [24] stated that

US pre-treatment of WAS at the Gässlösa WWTP (city of Borås, Sweden) had only negligible effect on biogas production or organic matter degradation. However, as WAS accounted for only 15% of the total sludge volume, test conditions might have been challenging. A second full-scale trial at the Ernemar WWTP (city of Oskarshamm, Sweden) did not result in a noticeable change in methane production or organic matter degradation as repeated malfunctions of the US reactor prohibited reliable test conditions [24]. Similar operational challenges were reported by [25], who stated that the lifetime of the radial horns employed at the full-scale test at the Ulu Pandan Water Reclamation Plant amounted to less than 2,000 h (or 83 d in 24/7 operation). In addition to the fast erosion, the reactor exhibited considerable clogging susceptibility, thus necessitating manual flushing and cleaning cycles every second day. [16] confirmed the limited lifetime of sound-emitting surfaces and stated that the sonotrodes employed at the full-scale test at the Bamberg WWTP (Germany) required annual replacement, thus entailing maintenance costs of several thousand euros per year.

Besides the somewhat miscellaneous experiences, it should be noted that the results of previous full-scale trials are not always published in peer-reviewed journals and sometimes challenged by inconsistent methodologies. For instance, start-up phases for verifying comparable performance between control and test digesters before the treatment were performed only rarely. Also, in some studies, no control digester was employed or experimental descriptions (for instance, regarding specific energy inputs, sludge characteristics, or feeding regimes) were incomplete or missing. Hence, a scientifically meaningful interpretation and comparison of currently available information on full-scale sonication tests is difficult, which seems a general issue for pre-treatments and has also been observed other for methods such as thermal hydrolysis [26,27].

Thus, in order to obtain a reliable assessment whether ultrasonic WAS pre-treatment can positively affect anaerobic sludge digestion under full-scale conditions or not, the effects of WAS sonication were examined at a municipal WWTP with two digesters operated in parallel (one used as control, and one used as test digester) after verifying comparable digester performance. Treatment effects were examined by monitoring

(i) sludge viscosity,
(ii) methane production,
(iii) biosolids degradation, and
(iv) digestate dewaterability.

To minimize clogging risk (as observed for sonotrode and radial horn reactors), a novel double-tube reactor with an unobstructed reaction chamber was employed for the first time. For obtaining results that can also be transferred to other plants, a WWTP with industry-typical operating conditions was selected (for instance, concerning substrate TS, or WAS to RS ratio). For a preliminary assessment of treatment economics, the electricity costs of the US reactor were balanced against potential benefits through, for instance, decreased sludge viscosity, additional methane production, improved biosolids degradation, or better digestate dewaterability.

2. Materials and methods

2.1 Selection and operational characteristics of the Starnberg WWTP

Based on a preceding field survey among 17 municipal WWTPs in southern Germany, the Starnberg WWTP (located approximately 20 km southwest of Munich, Germany) was selected for the full-scale trial. Hereby, the main selection criterion was to find a largely regular, well-performing plant rather than a plant with most amenable, but potentially non-representative pre-treatment conditions (such as very short HRTs in the digester or low substrate TS content as in previous full-scale tests [15,16]). The goal of selecting such typical, or even challenging plant was to obtain results that are also transferable to other WWTPs (according to the New-York-Principle: *"If I can make it there, I'll make it anywhere").* Further selection criteria were the availability of two comparable digesters operated in parallel for a setup comprising a control and a test digester, a high degree of controllability of the plant (especially regarding feeding regimes), and motivated plant personnel; every researcher who ever conducted a study on a full-scale plant will likely agree that the latter one is maybe the most important.

Key characteristics of the selected plant and the corresponding median values from the conducted field survey are summarized in Table 1.

Table 1: Average characteristics of the Starnberg wastewater treatment plant (WWTP) and median values from the survey among 17 WWTPs in southern Germany.

Parameter	Starnberg WWTP	Median of the 17 WWTPs examined
Design capacity [PE]	70,000	100,000
Daily PS production [m³]	69	108
Daily WAS production [m³]	30	59
TS of PS [% of FM]	3.8	3.9
VS of PS [% of TS]	86	80
TS of WAS [% of FM]	6.3	6.7
VS of WAS [% of TS]	74	73
Digester volume [m³]	2 · 2,500	5,000
Digester temperature [°C]	37	38
Average HRT [d]	42	32
Share of WAS relative to RS [% of VS_{fed}]	38	43

WWTP = Wastewater treatment plant, PE = Population equivalent, PS = Primary sludge, WAS = Waste activated sludge, TS = Total solids, FM = Fresh matter, VS = Volatile solids, HRT = Hydraulic retention time, RS = Raw sludge.

The comparison demonstrates that the Starnberg WWTP features rather representative characteristics regarding substrate TS (6.3% vs. a median value of 6.7%) and the share of WAS relative RS (38% vs. a median value of 43%, based on VS_{fed}). A larger deviation, on the other hand, was observed for the HRT, which amounted to 42 d on average at the Starnberg WWTP (ranging from 39 d to 45 d, due to the slightly fluctuating sludge production) as compared to 32 d for the median WWTP from the field survey. Such increased HRT represents a challenging boundary condition, however, as potentially accelerated reaction kinetics due to the US treatment may no longer be detectable [10]. At the same time, such constraint might have applied for most other plants, too, owing to the still quite high median and average HRTs of 32 d and 33 d, respectively. Therefore, the Starnberg WWTP seemed overall a good choice for the conducted full-scale trail. Full information on the operational characteristics of the 17 plants examined is given in the supplementary material (Table S1).

2.2 Digester operation and experimental schedule

For the experiment, the operation of the two digesters was synchronized with respect to heating, mixing, and feeding regimes. Heating and mixing regimes were already comparable, as both digesters are stirred by recently renewed, identical stirrers and heated (and further mixed) through the same recirculation loop. For synchronizing feeding regimes, digester feeding with PS was alternated every quarter of an hour, i.e., after the control digester was fed for 15 minutes, feeding changed to the test digester for the next 15 minutes and so on. Hereby, settling of the PS in the storage pit (potentially leading to different organic loading rates despite equal pumping times) could be minimized. Feeding with thickened WAS was conducted for eight hours per day, while the control and the test digester were fed for 2 times 2 hours each, respectively. Due to constant belt filter settings and flocculant supply, WAS characteristics remained more or less constant throughout a working day so that equally short feeding changes as for PS were not necessary.

To verify whether the synchronization was successful, the performance of the two digesters was monitored in a start-up phase for two HRTs (or 82 days) without US pre-treatment. After completing the start-up, the experiment was started, and the US reactors were activated for 4 hours per day (during WAS feeding of the test digester). The test phase of the experiment was conducted for seven consecutive HRTs (or 292 days), thus fully complying with the suggestion of the VDI guideline 4630 to monitor changes to continuous digesters for at least three to five HRTs [28].

2.3 US reactor and operating conditions

WAS pre-treatment was conducted using a novel double-tube US reactor, comprising an inner tube oscillator with a diameter of 50 mm, and a surrounding outer tube with a diameter of 164 mm. Through the double-tube layout, an unobstructed, circular reaction chamber with a gap distance of 57 mm was formed between the two tube surfaces. Such gap distance was selected to balance the requirements of operational stability (large gap distances desirable for low clogging risk) and cavitation field intensity (small gap distances desirable to minimize cavitationally inactive zones due to sound wave attenuation), according to previous reactor design optimizations for sewage sludge pre-treatment [29,30]. Both tubes resemble surface oscillation systems with a driving frequency of 25 kHz, whereas the inner tube is powered by two US convertors (750 W each, one at both ends of the tube) and the outer tube is powered by an array of 72 externally mounted US transducers (up to 50 W each). The tallied electrical power of one reactor unit amounts to 4.5 kW, with a resulting US density of 222 W per liter reactor volume. A 3-D schematic of the novel reactor system is depicted in Fig. 1A.

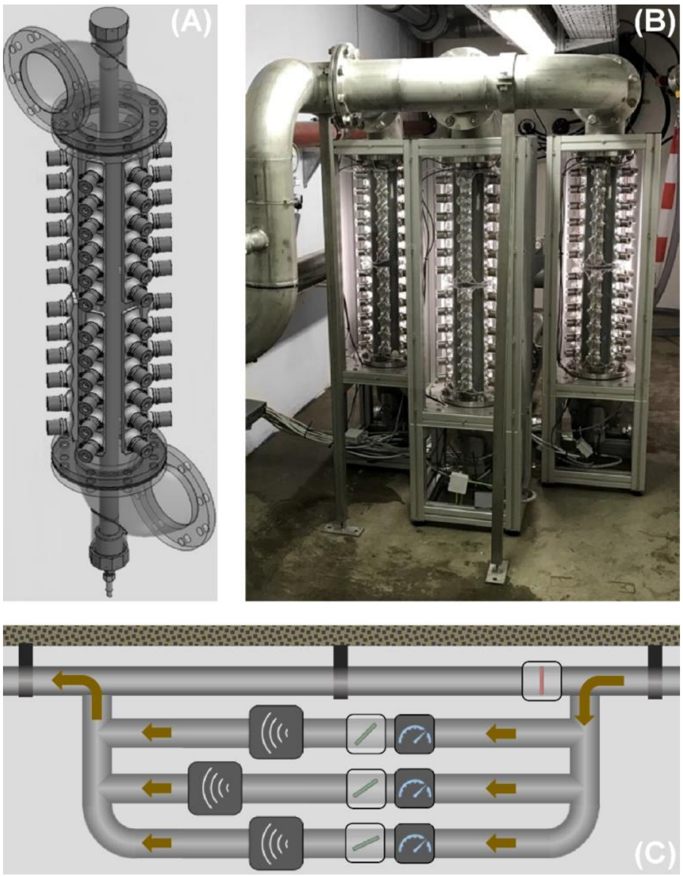


Fig. 1: 3-D schematic of the employed double-tube reactor (A), photograph of the whole reactor setup (B), and top view assembly plan of the reactor installation (C). The closed valve in the main WAS pipe is indicated by a red bar, while the partially opened valves in the inlet pipes to the US reactors are indicated by green bars. WAS flow is indicated by brown arrows.

For the full-scale trial, a total of three double-tube reactors (including additional inlet and outlet pipes, sludge flow meters, and flow valves) with a total electrical power of 13.5 kW were operated in parallel. Aluminum support frames allowed for a vertical positioning of the tube reactors, to ensure a reliable discharge of entrapped air pockets (potentially causing severe damage to the US reactor due to local overheating) and a homogeneous sludge flow over the sound emitting surface. A photograph and an assembly plan of the tri-partite reactor system is depicted in Figs. 1B and 1C, respectively.

The reactors were installed next to the main conveyance pipe for thickened WAS and were equipped with inlet and outlet pipes to lead the WAS flow from the main pipe through the reactor system. To facilitate full-stream treatment, the main pipe was blocked by a closed valve (see Fig. 1C). For ensuring equal distribution of the sludge flow over the three reactors, each inlet pipe was equipped with a flow meter and an electronic valve. Flow meters and valves were interconnected using the automation technology Loxone (Loxone Electronics GmbH, Kollerschlag, Austria). The control program recorded the individual flow rates in each line and calculated the targeted volume flow for each inlet pipe by dividing the total volume flow by three. Based on the calculated volume flow, the valve positions were dynamically adapted by the software, until the flow rate in each inlet pipe was equal to the desired flow rate. Resulting average flow rates for each pipe and reactor were approximately 1.25 m^3/h. Based on this volume flow, the electric power of the US reactors, and the TS content of WAS (6.3% on average), a specific energy input of about 200 kJ/kg_{TS} was attained, similar to previous full-scale trials [15,16].

In order to assess whether the novel double-tube reactor features improved clogging resistance or not, reactor cleaning or backwashing was omitted during the entire experimental phase of 374 days.

To ensure safe reactor operation nonetheless and to prevent damage to the US reactor in case of a clogging event, volume flow and temperatures of both sludge and reactors were logged every minute by the central automation system. Once the flow rate dropped below 0.3 m³/h or the temperature rose above 70°C at any point in the setup, US reactors were automatically deactivated.

2.4 Monitoring of treatment effects and digester performance

2.4.1 Sludge viscosity

Apparent sludge viscosity was determined once a week for thickened WAS before and after US pre-treatment and for DS sampled from both the control and the test digester. Viscosity measurements were conducted in triplicate at ten different shear rates (ranging from 6.45 1/s to 644.4 1/s) at a controlled temperature of 20°C using a rotational viscometer (Viscotester VT 500, HAAKE Messtechnik GmbH, Karlsruhe, Germany).

Based on the obtained viscosity curves of untreated and pre-treated WAS, potential reductions in frictional head loss (leading to savings in pump energy demand) in the WAS pipe were calculated according to [31]:

$$\Delta h = \left(\frac{4LK}{D}\left[\frac{8Q}{\pi D^3}\frac{(3n+1)}{n}\right]^n\right)\cdot\frac{1}{10}$$

(Eq. 1)

where Δh is the head loss [m], L is the length of the WAS pipe [= 45 m], *K* is the consistency index [Pa·s^n], D is the inner pipe diameter [= 0.15 m], Q is the flow rate [= 0.001 m³/s, or 3.75 m³/h], and *n* is the power-law index [-]. Consistency index *K* and power-law index *n* were determined for both untreated and pre-treated WAS by fitting the Ostwald-de-Waele (or power-law) model to the obtained viscosity curves by the use of the least squares method, according to [32,33]. Calculations of frictional head loss were cross-checked using an online calculator available at https://checalc.com/fluid_flow_power_law.html.

The pump energy savings due to the reduced head loss were calculated according to Eq. 2:

$$E_s = tQ\rho g\Delta h\frac{1}{\eta}$$

(Eq. 2)

where E_s is the saved shaft energy [kWh], t is the operating time [h], ρ is the sludge density [= 1,000 kg/m³], g is the acceleration of gravity [= 9.81 m/s²], Δh is the head loss [m], and η is the assumed pump efficiency [= 0.6].

2.4.2 Methane production

The volume of the produced biogas and its methane content were continuously measured using two identical ultrasonic flow volume and gas quality meters (OPTISONIC 7300 Biogas, KROHNE Messtechnik GmbH, Duisburg, Germany), installed at the gas outlets of both digesters. The recorded gas volume was automatically converted to standard conditions (dry gas, temperature of 0°C, pressure of 101,325 Pa). Both gas volume and quality were logged by the WWTP´s process control system every 60 minutes.

2.4.3 Sludge solids content

To monitor the temporal development of the feed sludge characteristics and to identify potential reductions in the (organic) solids content of the digestate, TS and VS of thickened WAS, PS, and of digested sludge (DS) of both digesters was determined in triplicate once a week according to standard methods [34].

2.4.4 Digestate dewaterability

The dewaterability of the digestate of both digesters was determined once a week through capillary suction time (CST) tests, using a CST test apparatus (101/A, Axchem Deutschland GmbH, Erzhausen, Germany) and Axchem filter paper. CST measurements were conducted in quintuplicate at a controlled temperature of 20°C.

A detailed description of both the test apparatus and the test procedure can be found elsewhere [35].

2.5 Statistical analysis

According to the decision diagram for statistical tests in anaerobic digestion experiments [36], differences between the control and the test digester were assessed using a Student's *t*-test, with p values < 0.05 indicating significant differences and p values < 0.01 indicating highly significant differences. To ensure compliance with the *t*-test requirements (i.e., normal distribution of data and homogeneity of variance), data sets were previously analyzed using the Shapiro-Wilk test and the F-test, respectively. The additional requirement of independent measurement data (control vs. test digester) was met through the selected experimental setup [36].

3. Results and discussion

3.1 Operational aspects

Despite the omission of cleaning and backwash, no clogging events were encountered throughout the entire experimental phase of 374 days. Hence, from an operational point of view, the novel reactor design seems well-suited for full-scale application. However, for future trials, it is recommended to switch from the parallel operation of the reactors (causing increased material costs and system complexity) to a simpler and more cost-effective serial arrangement of the reactors. It was furthermore shown by [37] that an increased flow velocity in US reactors enhances cavitation intensity, which is a result of the shear-thinning flow behavior of sewage sludge and the reduced sound wave attenuation at lower viscosities.

3.2 Effects of the pre-treatment on sludge viscosity

The apparent viscosity of WAS before and after the pre-treatment is displayed in Fig. 2, for a shear rate of 644 1/s. This shear rate was selected for data analysis, as it exhibited the lowest result variability among the ten shear rates tested. On average, US treatment reduced the viscosity of WAS by 5.8%, while the strongest average decrease (per HRT) of almost 8% was observed in HRT #6. The same qualitative difference was observed for the other recorded shear rates (data not shown). When comparing the data sets of the individual HRTs, viscosity reduction was significant for HRT #4 and HRT #6 (at $p < 0.05$), and highly significant for HRT #8 (at $p < 0.01$). For the lumped data sets from the entire test phase (HRTs #3-9), the overall viscosity reduction due to US pre-treatment was found to be highly significant (at $p < 0.01$). A discussion on the economic implications of the decreased viscosity is presented in section 3.6.

The achieved reduction in viscosity is in line with previous studies [12,38], similarly reporting reduced WAS viscosity after US pre-treatment. Yet, the obtained viscosity reduction of around 6% is considerably lower than typically reported decreases. For instance, [38] achieved a reduction in viscosity of over 70%, albeit by the use of a specific energy input as high as 27,000 kJ/kg_{TS}. To implement such energy input at the Starnberg WWTP, a full-scale reactor with an electric power of about 1,800 kW would have been necessary, thus entailing investment cost of roughly 13 million € and monthly electricity costs of about 90,300 € assuming 8 h of reactor operation per day an industrial electricity price of 21 Cent/kWh. The example calculation again emphasizes that promising lab-scale results obtained at high energy inputs require critical interpretation and are not directly transferrable to full-scale applications.

In addition to WAS viscosity, the viscosity of DS in both the control and the test digester was analyzed (see Fig. S1 in the supplementary material). In contrast to WAS, no measurable impact on DS viscosity could be discerned, which is plausible given the only moderate reduction of WAS viscosity of around 6% and the relatively low share of WAS in the RS stream (approximately 69 m^3 of PS and 30 m^3 of WAS per day). Savings due to reduced energy demand for stirrers or the pumps installed in the recirculation loop of the digesters are, therefore, not expected.

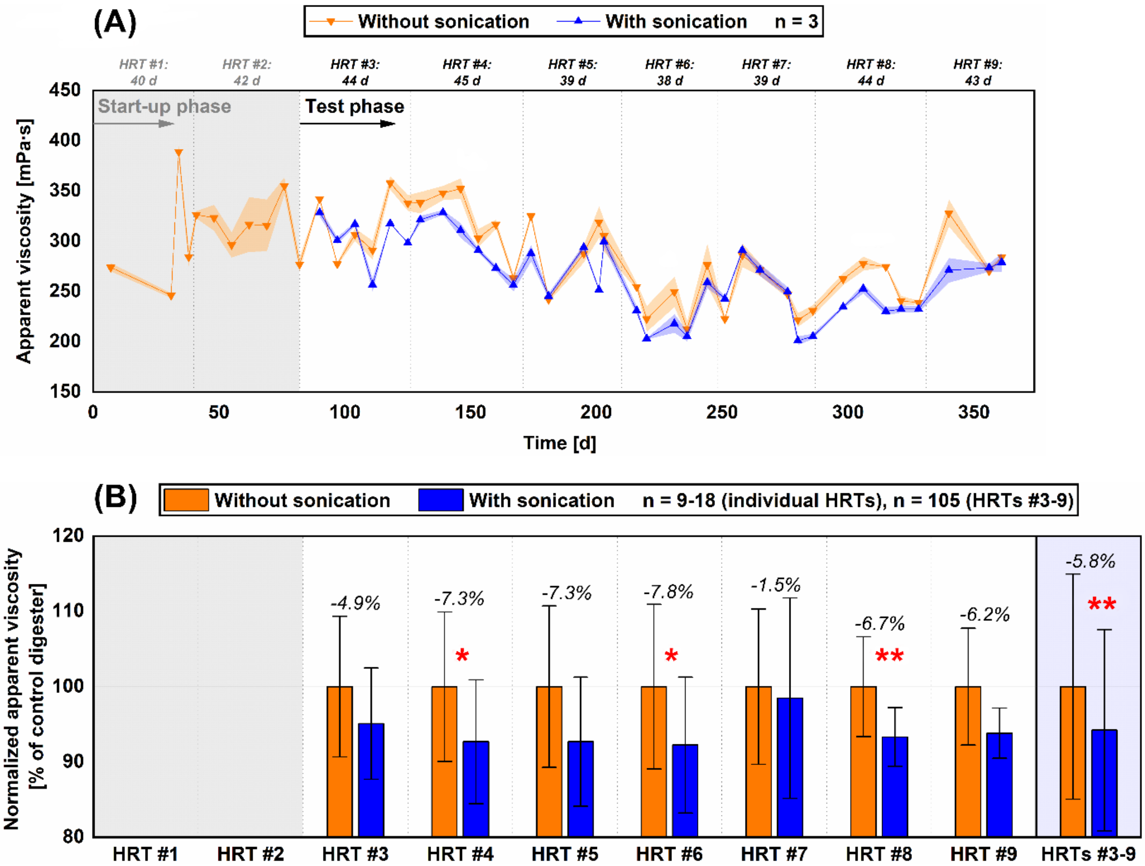


Fig. 2: Development of apparent viscosity of waste activated sludge with and without sonication at a shear rate of 644 1/s (A) and normalized apparent viscosity relative to the control digester (B). Significant and highly significant differences between two data sets are denoted with one, or two asterisks, respectively. Error bands and error bars in denote the standard deviation of the mean.

3.3 Effects of the pre-treatment on methane production

The development of the methane production of both digesters is displayed in Fig. 3. The performance of the digesters largely equalized during the start-up phase (average deviation in HRT #2 of less than 3%, at $p > 0.09$), thus indicating a successful synchronization of feeding regime and digester operation. Based on these favorable test conditions, the test phase could already be initiated after a start-up phase of only two HRTs.

The test phase was conducted for a total of seven HRTs (or 292 days). Yet, despite the long testing time, the conducted US pre-treatment did not result in a significant enhancement of methane production in the test digester. Statistical analysis revealed that the difference between the control and the test digester remained insignificant, both for the comparison of the individual HRTs and for the comparison of the lumped data sets from the full test phase. Only by trend, methane production of the test digester seemed slightly enhanced from HRT #7 onward, with a maximum, HRT-averaged percentage increase of 6.2% relative to the control digester in HRT #9 (at $p < 0.1$).

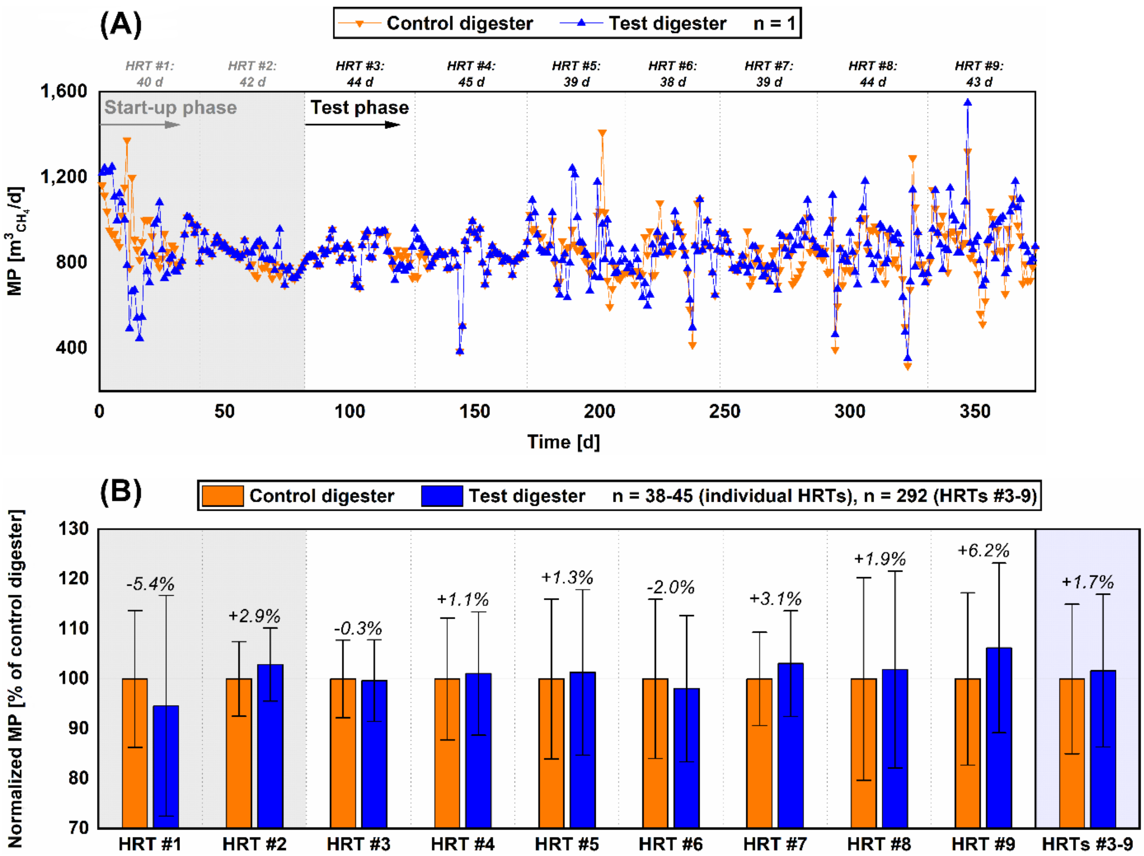


Fig. 3: Development of daily methane production (MP) of the control and the test digester (A) and normalized MP relative to the control digester (B). Error bars denote the standard deviation of the mean.

The finding of an only insignificant enhancement of methane production is in contradiction to most previous full-scale trails, where considerate increases in methane production between 13% and 58% were observed [15,16,23]. One possible reason for the distinct difference could be the comparably low ultrasonic density of the novel double-tube reactor (222 W/L) in combination with the high solids content of the WAS

(= 6.3%). Previously reported US densities of sonotrodes and radial horns were higher and amounted to 345 W/L, and 1,714 W/L, respectively [15,16]. The hypothesis that the novel reactor type was challenged by the high TS content of WAS is substantiated by a recent study by [39], who compared the performance of tube and sonotrode reactors and concluded that high-intensity sonotrode reactors were more suitable for solids-rich substrates such as WAS, while tube reactors (with externally mounted surface transducers) were more effective for substrates with a lower TS content (such as DS). Such assessment is further supported by a study of [40], who did not observe notable BMP enhancement when treating thickened WAS (TS = 8%) with a full-scale “double-layer” US reactor similar to the reactor in this study. Generally, [41] demonstrated that disintegration efficiency already starts to decline at TS contents above 3%, thus further confirming that the TS content of the present WAS sample (= 6.3%) was far higher than optimal TS concentrations for US treatment. At the same time, such performance decline at elevated solids contents seems a general issue for WAS pre-treatment, as the sludge typically exhibits TS contents clearly above 5% after thickening (see Table S1 and [38,42]).

Another factor for the only moderate enhancement might be the long HRT of over 40 days at the Starnberg WWTP, which may have rendered a potential acceleration of reaction kinetics difficult to detect [10]. On the other hand, [43] reported an increase in biogas production by 45% following US pre-treatment despite an extremely long HRT of 69 days in a full-scale digester treating WAS. Nonetheless, there is consensus that shorter HRTs offer more favorable pre-treatment conditions [10], and most of the previous full-scale trials were accordingly conducted at shorter HRTs ranging from 12 - 30 days [15,16,23].

Finally, the relatively low share of WAS in the RS (= 38%, based on VS_{fed}) and the accordingly high portion of methane production from PS might have further masked the effects of the WAS pre-treatment. For instance, even when assuming equal methane yield from VS_{WAS} and VS_{PS}, a methane production increase by over 16% from WAS only is necessary to realize the observed total methane production increase of 6.2%. Given the fact that the current share of WAS to RS was found to be only slightly lower than the median value (= 43%, see Table 1), it becomes clear why effects of pre-treatments can be difficult to prove in full-scale trials. In this context it should also be noted that current sludge minimization measures are designed to significantly reduce WAS production so that in the future, the share of WAS to PS may decline further [44,45].

3.4 *Effects of the pre-treatment on the solids content of the digestate*

To elucidate potential effects of the treatment on solids degradation, the TS content of the digestates was monitored weekly (see Fig. 4). As for methane production, the solids contents of the two digestates were highly similar during the start-up phase (average deviation of less than 1% averaged over HRT #1 and HRT #2), again indicating comparable digestion performance.

Just like for methane production, US pre-treatment could not provoke a significant effect on the solids content in the digestate. The only exceptions are HRTs #5 and #6, where the test digester exhibited a significantly increased TS content. However, the result might be still related to natural fluctuations since the difference became insignificant again in the three subsequent HRTs. An increase in the TS content in the sonicated digester is furthermore considered very unlikely. Overall, results suggest that savings due to reduced amounts of residual biosolids are not to be expected. The finding is again in contrast to the majority of the previous full-scale trails (see [15,16,23]), while it confirms the results of the Swedish study on the Gässlösa WWTP in Borås [24]. Besides the reactor type, the previously mentioned factors, such as the long HRTs or the low share of WAS might again be potential causes for the only insignificant effect of the US pre-treatment.

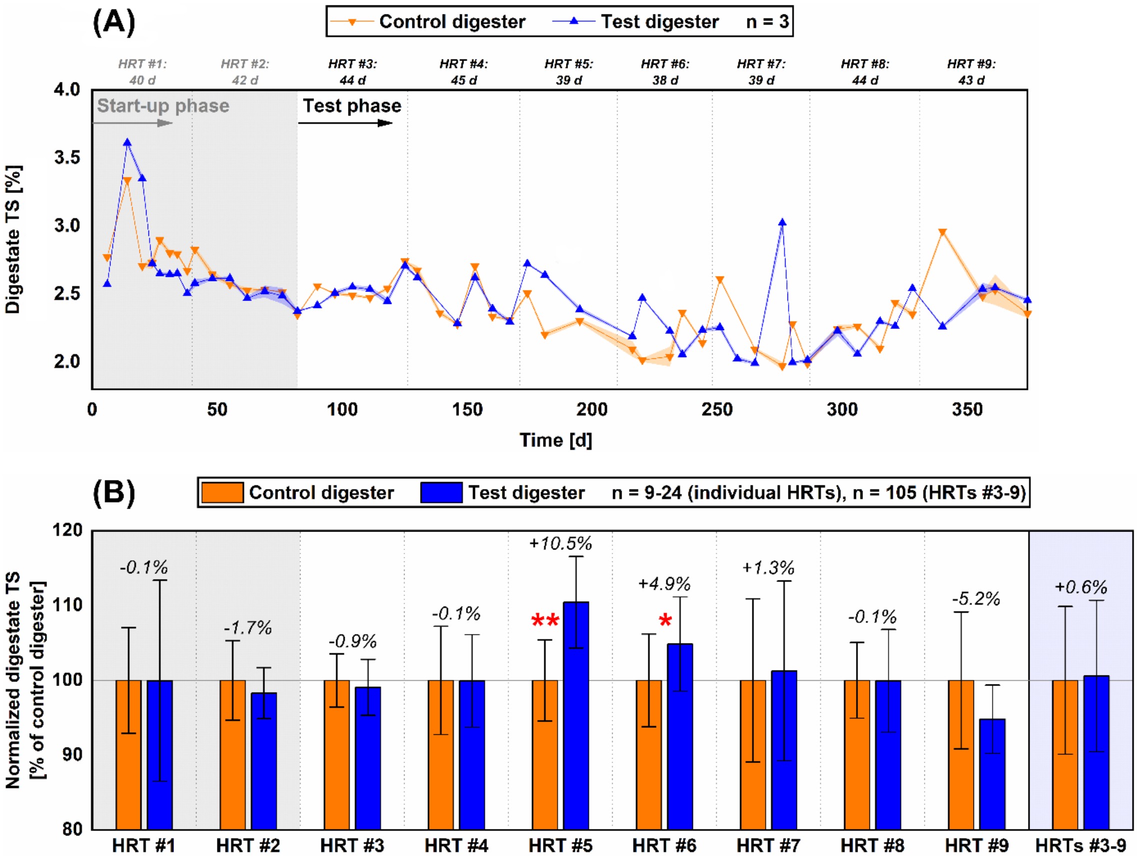


Fig. 4: Development of the total solids (TS) concentration of the digestate from the control and the test digester (A) and normalized digestate TS relative to the control digester (B). Significant and highly significant differences between two data sets are denoted with one, or two asterisks, respectively. Error bands and error bars denote the standard deviation of the mean.

3.5 Effects of the pre-treatment on the digestate dewaterability

CST values for both digesters are depicted in Fig. 5. The differences between control and test digester were insignificant throughout the start-up and the test phase, except for one case in HRT #3, where the test digester exhibited a significantly lower CST, at $p < 0.05$. Overall, the obtained data suggest that the US pre-treatment had no measurable effect of digestate CST, which may be again explained by the above-mentioned reasons. Savings due to enhanced dewatering as observed in a full-scale test by Hogan et al. (2014) could, therefore, not be realized. Given previous findings that US pre-treatments can also deteriorate sludge dewaterability [46,47], the result can also be seen as positive.

At the same time, such effects were mostly observed at high energy inputs so that deteriorated digestate dewaterability may be generally of lesser concern for full-scale sonication [48].

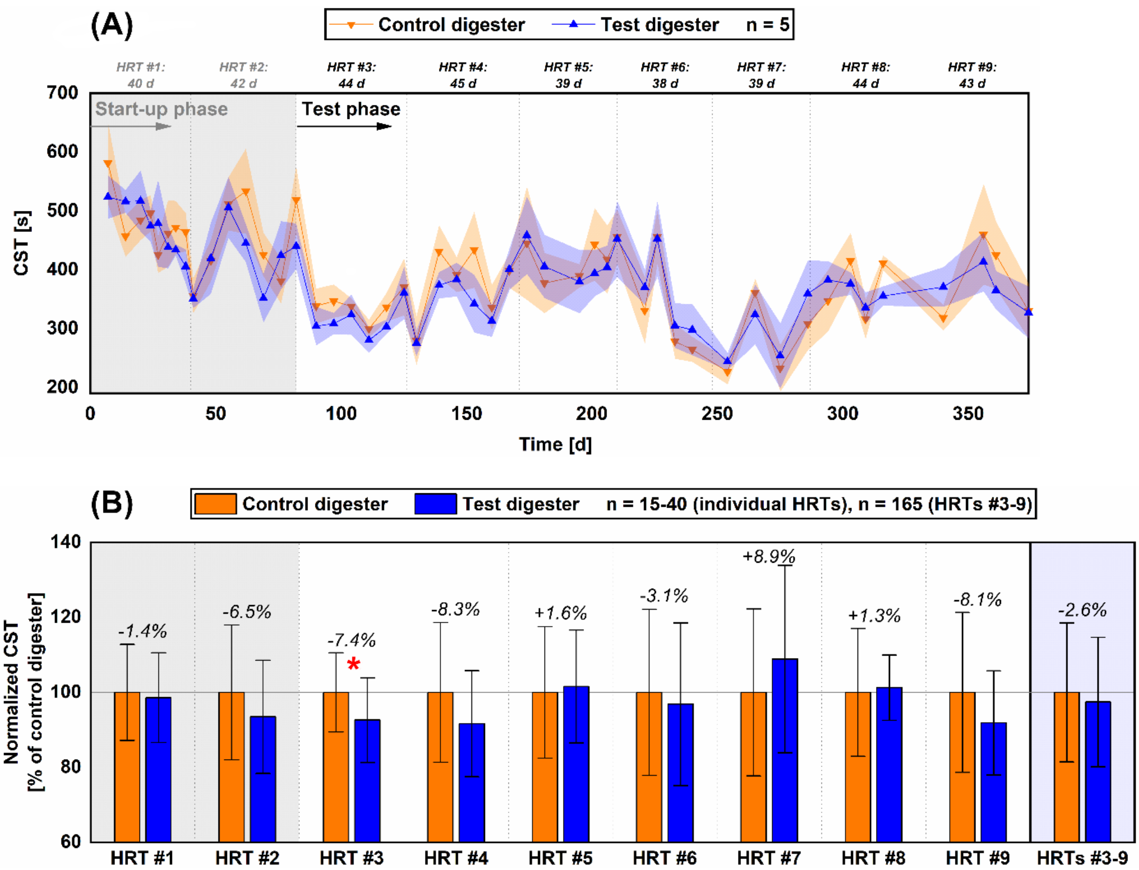


Fig. 5: Development of the capillary suction time (CST) of the digestate from the control and the test digester (A) and normalized CST relative to the control digester (B). Significant differences between two data sets are denoted with an asterisk. Error bands and error bars denote the standard deviation of the mean.

3.6 Economic aspects

For a simplified assessment of treatment economics, the electricity demand of the US reactor was balanced against the energy savings through observed treatment effects, i.e., reduced WAS viscosity and increased methane production (Fig. 6). The achieved viscosity reduction entailed a slightly lower head loss in the WAS conveying pipe (= 0.42 m on average), which corresponded to average pump energy savings of 1.6 kWh per HRT. At the same time, the saved average head loss was less than the additional head loss caused by the US reactor setup (0.70 m on average) so that the full-scale reactor caused a net increase in head loss despite the viscosity reduction. Even when disregarding this additional head loss, the savings due to reduced WAS viscosity would have been negligible. The average electricity demand of the US reactor (~ 2,000 kWh/HRT) was roughly 1,200 times higher than the savings due to the 6%-reduction in WAS viscosity, thus emphasizing that a slightly improved WAS rheology (just like a small added head loss due to the US reactor) is irrelevant concerning treatment economics.

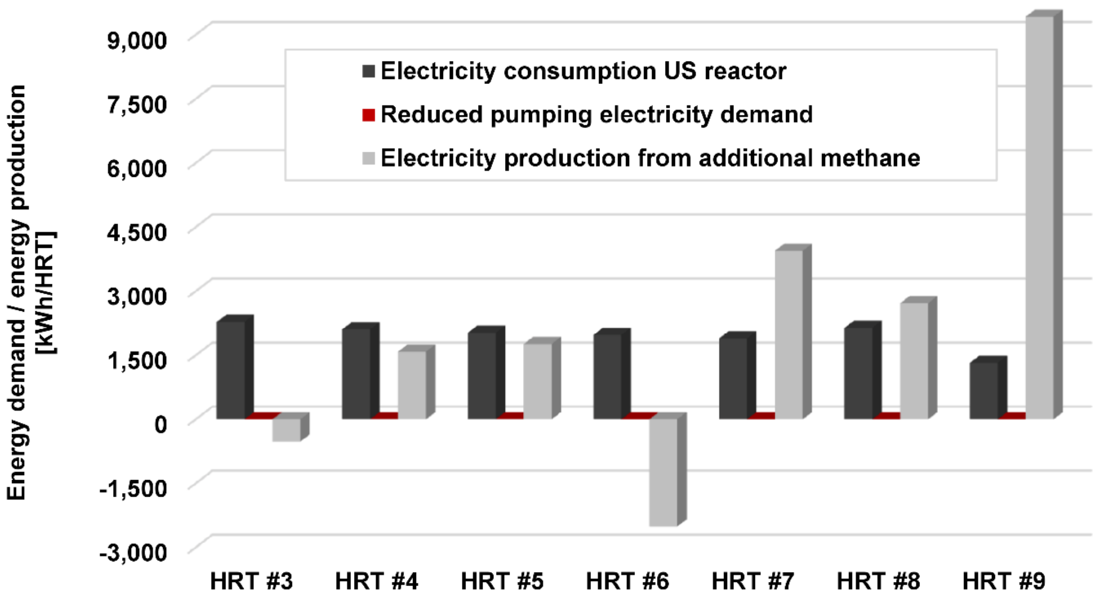


Fig. 6: Energy balance of the full-scale ultrasound (US) pre-treatment assuming an energy content of 10 kWh per m³ of methane and an electrical efficiency of the combined heat and power plant of 40%.

Electricity production from additional methane, on the other hand, was able to clearly overcompensate the electricity demand of the US reactor, at least in the last three HRTs of the test phase. A maximum surplus energy production of about 9,400 kWh/HRT was observed in HRT #9, as opposed by an energy demand of the US reactor of approximately 1,300 kWh/HRT. The low energy demand of the US reactor in HRT #9 was due to a necessary reduction in electric power due to the onset of wear of the transducers. Thus, in the last HRT tested, the energy recovery through additional methane was over seven times higher than the electricity demand of the US reactor. The promising result indicates that even a small increase in methane production (+6.2% in HRT #9) can entail a positive energy balance in full-scale sonication, which is due to the low specific energy input (157 kJ/kg_{TS} in HRT #9) in full-scale tests and the "masking" of the actual pre-treatment effect due to the high share of PS in the RS. At the same time, it must be emphasized that the increase was statistically not significant and that the comparably high methane production in the last HRT tested was most likely also a result of the sonication during previous HRTs with a higher specific energy input. The calculated high energy recovery might therefore resemble an overestimation, while an inclusion of methane production enhancement in the economic assessment remains speculative anyway due to the only insignificant increase.

3.7 Recommendations for future (full-scale) testing

The obtained results suggest that US pre-treatment does not lead to significant (or economically relevant) improvements of anaerobic digestion performance at well-performing WWTPs with industry-typical operating conditions. Hence, it seems advisable to conduct future full-scale trials under more amenable boundary conditions, for instance at WWTPs operating at capacity limit with shorter HRTs. A lower solids content of the substrate and a smaller share of PS might further contribute to more successful full-scale testing. Besides the presumably stronger treatment effects under such more favorable conditions, an economic reactor operation may be easily attained when the pre-treatment is able to prevent the construction of an additional digester. Compared to the construction costs of a digester, the purchase and operating costs of an US reactor are rather moderate so that even a negative energy balance can be economical.

Another possibility to increase the impact of ultrasonic disintegration could be a shift to co- or inter-treatment, i.e., the sonication of the already pre-fermented DS in the recirculation loop of the digester. In laboratory tests, the sonication of DS revealed promising and significantly higher methane yield increase than WAS pre-treatment [39,49–51], which could be explained by the significantly lower TS and the hereby improved formation of cavitation bubbles [29]. Besides, DS sonication would efficiently concentrate treatment effects on the most recalcitrant substances present in the sludge, as readily bioavailable material would already be degraded. A full-scale verification of the promising effects is yet to be conducted.

For future lab-scale studies, it seems advisable to align the experimental settings more closely to conditions feasible at industrial scale. Especially regarding specific energy input, the present study could highlight that pre-treatment conditions in full-scale plants (~ 200 kJ/kg_{TS}) differ considerably from common laboratory settings (1,000 kJ/kg_{TS} - 16,000 kJ/kg_{TS}). For more meaningful and more transferable results, future lab-scale studies should, therefore, put a clear focus on low-intensity US pre-treatments.

4. Conclusion

The results of the conducted full-scale study suggest that ultrasonic WAS pre-treatment does not lead to notable enhancements of anaerobic digestion performance. The only treatment effects observed were a significant reduction of WAS viscosity (-5.8% on average) and a slight but insignificant enhancement of methane production (+6.2% at maximum). While the reduction of WAS viscosity was irrelevant regarding treatment economics, the increase in methane production was able to compensate the electricity demand of the US reactor for three consecutive HRTs, despite the only small percentage increase. As the enhancement was statistically not significant, however, such economic assessment remains somewhat speculative. Regarding operational stability, no clogging events were experienced despite an operation of 374 days without backwash, thus indicating that the double-tube reactor is generally suitable for full-scale WAS treatment. Yet, to ultimately verify the functional capability of the novel reactor type, further full-scale tests under more favorable boundary conditions (such as short HRTs, low substrate solids content, or a larger WAS to RS ratio) or with a different treatment scheme (co-treatment vs. pre-treatment) should be conducted.

Acknowledgements

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Supplementary material

Table S1: Selected characteristics of the 17 wastewater treatment plants examined.

	Design capacity [PE]	**Daily PS production [m^3/d]**	**Daily WAS production [m^3/d]**	**TS of PS [% of FM]**	**VS of PS [% of TS]**	**TS of WAS [% of FM]**	**VS of WAS [% of TS]**	**Digester volume [m^3]**	**Digester temperature [°C]**	**HRT [d]**	**[% of VS_{fed}]**
Average	**154,800**	**163**	**91**	**4.1**	**81**	**6.4**	**73**	**8,100**	**38**	**33**	**43**
Median	**100,000**	**108**	**59**	**3.9**	**80**	**6.9**	**73**	**5,000**	**38**	**32**	**43**
WWTP #1 (Starnberg)	**70,000**	**69**	**30**	**3.8**	**86**	**6.3**	**74**	5,000 (2·2,500)	**37**	**42**	**38**
WWTP #2	600,000	800	380	3.5	83	4.2	81	27,000 (3·9,000)	39	22	36
WWTP #3	75,000	100	n.a.	4.5	75	n.a.	n.a.	4,680 (2·2,340)	38	25	n.a.
WWTP #4	65,000	40	24	4.0	n.a.	6.7	n.a.	1,400 (2·700)	36	22	n.a.
WWTP #5	175,000	115	75	4.9	82	7.7	78	8,250 (2·4,125)	38	40	49
WWTP #6	110,000	85	40	5.0	80	4.7	70	3,600 (3·1,200)	38	30	28
WWTP #7	100,000	90	38	4.5	n.a.	7.7	n.a.	3,600 (2·1,800)	37	30	n.a.
WWTP #8	220,000	286	197	3.5	80	4.3	70	14,250 (3·4,750)	37	32	43
WWTP #9	51,000	78	34	3.0	87	6.1	67	3,100 (2,350 + 750)	40	24	41
WWTP #10	110,000	n.a.	100	n.a.	n.a.	7.0	75	4,950	38	35	n.a.
WWTP #11	230,000	142	40	4.5	69	6.9	71	8,500 (2·4,250)	40	40	31
WWTP #12	400,000	205	160	6.3	79	8.1	69	18,000 (3·6,000)	38	30	47
WWTP #13	192,000	154	96	3.8	79	5.0	76	11,200 (3·1,800 + 2·2,900)	40	48	44
WWTP #14	68,000	30	43	2.6	81	7.1	73	5,000 (2·2,500)	38	30	78
WWTP #15	75,000	180	100	5.5	90	6.8	72	8,000 (2·2,000 + 4,000)	39	38	35
WWTP #16	41,747	57	20	3.1	78	7.5	76	2,800 (2·1,400)	37	36	45
WWTP #17	18,490	180	80	2.5	81	6.0	70	8,000 (2·4,000)	39	32	48

WWTP = Wastewater treatment plant, PE = Population equivalents, PS = Primary sludge, WAS = Waste activated sludge, TS = Total solids, FM = Fresh matter, VS = Volatile solids, HRT = Hydraulic retention time, RS = Raw sludge.

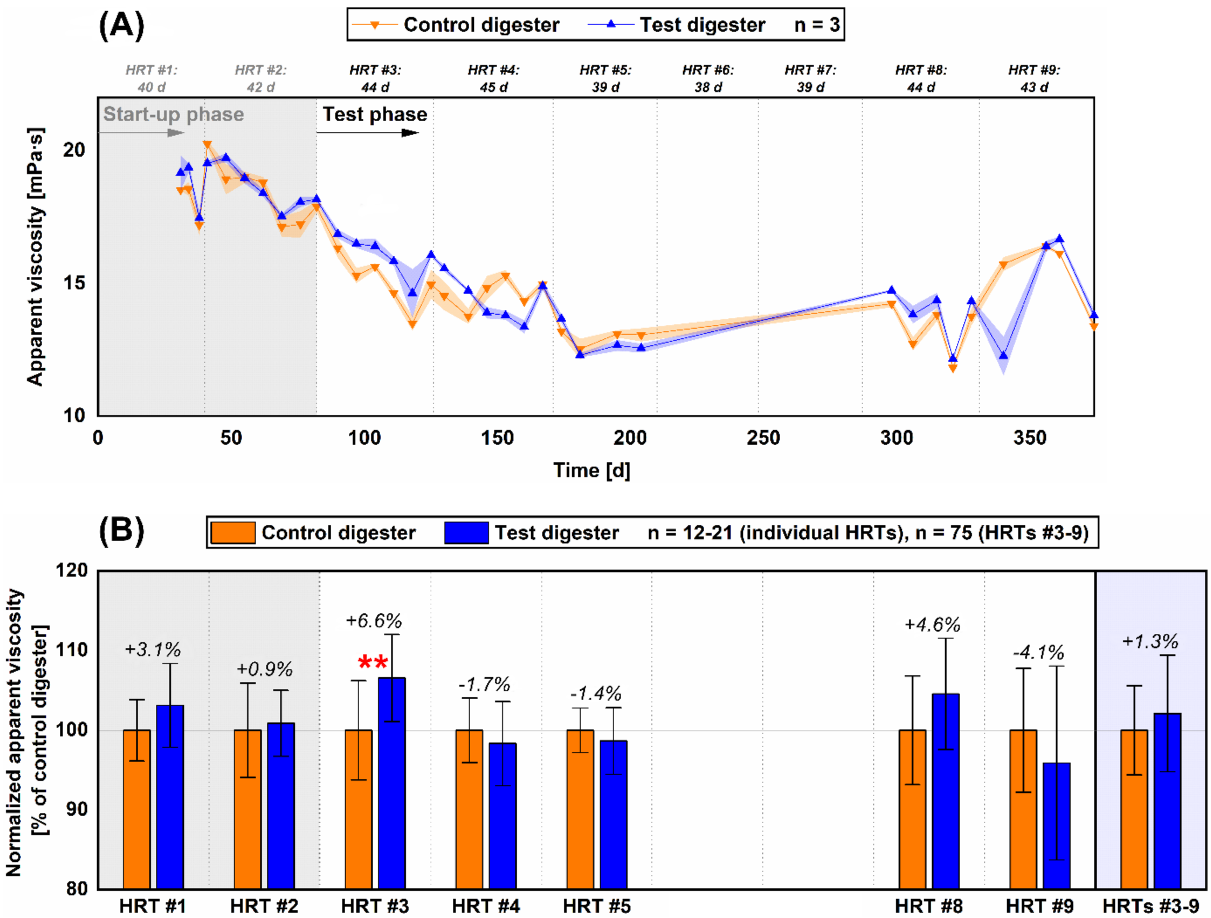

Fig. S1: Development of apparent viscosity of digested sludge from the control and the test digester at a shear rate of 644 1/s (A) and normalized apparent viscosity relative to the control digester (B). Highly significant differences between two data sets are denoted with two asterisks. Error bands and error bars denote the standard deviation of the mean.

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