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Sewage sludge disintegration using innovative ultrasound reactors with surface transducers - Performance assessment and optimization of operating conditions

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Abstract

Ultrasound can be used in wastewater treatment for sewage sludge disintegration. The mechanical disintegration method is based on the violent collapse of transient acoustic cavitation bubbles in the sludge, which facilitates the de-agglomeration of sludge flocs and the lysis of bacterial cells. Hereby, the biological degradability of the sludge can be increased, and the efficiency of anaerobic sludge digestion can be enhanced. The reported main merits of sewage sludge disintegration include accelerated degradation kinetics, increased biogas production, intensified solids removal, and improved digestate dewaterability.

Despite the multitude of advantages, the application of ultrasonic disintegration in wastewater treatment plants is still limited. The limited use is mainly due to operational difficulties of currently used sonotrode and radial horn reactors, which feature a high risk of clogging and a pronounced susceptibility to cavitation erosion.

Therefore, the overarching research goal of this dissertation was a performance assessment and further development of a novel, low-maintenance tubular ultrasound reactor with surface transducers. The potential advantages of the new reactor design include an unobstructed reaction chamber for minimized clogging risk and low ultrasound intensity (defined as electrical power per surface area, W/cm^2) for low susceptibility to cavitation erosion. Possible disadvantages are a potentially reduced performance due to the low ultrasound intensity and a more difficult refurbishment of eroded reactors since the ultrasound transducers are firmly fixed to the reactor housing. The following specific research objectives were defined as milestones for the research program:

- Optimization of the specific energy input for most energy-efficient reactor operation
- Characterization of the fluid dynamics of sewage sludge within US reactors
- Optimization of the reactor design for intensive and homogeneous sludge treatment
- Full-scale assessment of the novel reactor type
- Investigation of an alternative *co-treatment* scheme for digested sludge

The examination of the effect of specific energy inputs on sonication efficiency demonstrated that low, industrially feasible energy inputs (for instance, 200 kJ per kg of total solids, TS) enable equally high percentage increases in biochemical methane potential (up to 12%) as high, lab-scale energy inputs of 2,000 kJ/kg$_{TS}$ or 3,000 kJ/kg$_{TS}$. Intermediate inputs between 400 kJ/kg$_{TS}$ and 1,000 kJ/kg$_{TS}$, on the other hand, could not realize significant increases in methane potential. Furthermore, results demonstrated that only the lowest tested energy input of 200 kJ/kg$_{TS}$ allowed for an energy-neutral operation of the ultrasonic reactor, for which the energy content of the additionally generated methane was greater than the energy consumption of the ultrasonic reactor. The finding emphasizes the high potential of low, industrially feasible energy inputs and confirmed the general capability of the novel ultrasonic reactor for sewage sludge disintegration (proof-of-concept).

The investigation of the fluid dynamics in ultrasonic reactors revealed that the high viscosity of sewage sludge largely suppresses cavitation-induced mixing. Hence, sludge flow in ultrasonic reactors was shown to remain largely laminar, despite the exposure to ultrasonic irradiation. As a consequence, the important role of the reactor design was emphasized, since potentially inhomogeneous cavitation fields cannot be compensated for by cavitation-induced mixing of the fluid.

Based on these results, the influence of the reactor design on the homogeneity and the intensity of ultrasound treatment was determined. For this purpose, a size-adjustable flatbed reactor was used, which enabled the investigation of reaction chamber heights ranging from 20 mm to 100 mm. The results revealed that an increase in reaction chamber height leads to both an increase in treatment inhomogeneity and a decrease in treatment intensity. The reason for the different sonication efficiencies despite a constant energy input of 200 kJ/kg$_{TS}$ was found to be strong sound field attenuation, which becomes more and more influential with increasing reaction chamber height. For instance, while no notable sound wave attenuation was discernible at a reaction chamber height of 20 mm, both primary and secondary treatment effects (i.e., cavitation noise and chemical oxygen demand solubilization, respectively) were totally absent in the channel center at a reaction chamber height of 100 mm. To further account for the impact of fluid flow, the flow of the sludge through the reaction chamber was approximated by means of a laminar flow simulation. The results of the simulation suggest that treatment inhomogeneity increases even further in flow-through reactors, since the highest volume flows prevail in the regions with the lowest cavitation intensity. As a rule of thumb, it was concluded that ultrasonic reactors for sewage sludge treatment should be *as small as possible and as large as necessary* (for minimized risk of clogging).

Based on the previous studies to optimize the energy input and the reactor design, a large-scale ultrasonic reactor for the pre-treatment of waste activated sludge was designed and tested at a full-scale wastewater treatment plant. For the trial, a typical wastewater treatment plant with two digesters operated in parallel was selected in order to generate results that are both scientifically reliable and transferable to other plants. The pre-treatment of waste activated sludge with a solids content of around 6% was conducted in full-stream mode with an energy input of 200 kJ/kg_{TS}. Treatment effects were analyzed with regard to methane production, sludge viscosity, (volatile) solids removal, and digestate dewaterability. The results of the full-scale test demonstrated that the pre-treatment (contrary to the promising laboratory results at low energy input) had only moderate effects. Only the viscosity of waste activated sludge was significantly reduced by approximately 6% as a result of the treatment, which did not result in any significant savings, however. Methane production was elevated by a maximum of 6%, albeit only by trend. For all other parameters (i.e., viscosity of digested sludge, solids removal, and digestate dewaterability), the treatment did not achieve significant improvements. Hence, based on the only moderate effects and the insignificant savings, the full-scale pre-treatment was found to be not economical.

The alternative co-treatment scheme for digested sludge was analyzed in the lab using a continuously operated digester system with a non-sonicated control and a sonicated test digester. Despite a relatively high specific energy input of 2,000 kJ/kg_{TS}, the treatment led to an only moderate improvement in methane yield (+ 9%) and solids removal (+5%). As the energy costs of ultrasonic treatment were almost twenty times higher than the cost savings from additionally generated methane gas and reduced amounts of sludge for disposal, the process was also found to be uneconomical under the conditions investigated. To render co-treatments economically viable, future research exploring the potential of low energy inputs is required.

In summary, the results suggest that the potential of ultrasonic sewage sludge treatment using tubular reactors with a surface transducers is limited. The main reason for the moderate performance was possibly the strict orientation of the experimental settings to full-scale conditions (including low energy inputs, high solids content of the treated substrates, and long hydraulic retention times in the digester), which suggest that ultrasonic pre-treatment is not effective or economically viable for typical, well-performing wastewater treatment plants. Future studies should, therefore, put a clear focus on plants with underperforming digesters with short hydraulic retention times and substrates with a lower solids content. Besides, also alternative treatment strategies with potentially more favorable boundary conditions should be explored, such as ultrasound-assisted enhancement of biological nitrogen elimination, post-treatment of digestate for enhanced dewaterability, or pyrolytic destruction of organic pollutants in industrial wastewater treatment.

Kurzzusammenfassung

Ultraschall kann in der Abwasserbehandlung zur Klärschlammdesintegration eingesetzt werden. Das Verfahren basiert auf transienter akustischer Kavitation, welche eine mechanische Zerkleinerung von Schlammflocken, sowie den Aufschluss von Bakterienzellen ermöglicht. Durch die Desintegration kann die biologische Abbaubarkeit des Schlamms gesteigert werden, was die Leistungsfähigkeit der anaeroben Schlammfaulung erhöht. Zu den wichtigsten in der Literatur beschriebenen Effekten der Klärschlammdesintegration zählen ein beschleunigter biologischer Abbau, eine erhöhte Biogasproduktion, ein intensivierter Feststoffabbau, sowie eine verbesserte Restschlammentwässerbarkeit.

Trotz der Vielzahl an Vorteilen ist der Einsatz der Ultraschalldesintegration auf Kläranlagen bislang noch begrenzt. Der limitierte Einsatz ist vornehmlich auf betriebliche Schwierigkeiten derzeit eingesetzter Sonotroden- und Hornreaktoren zurückzuführen, welche ein hohes Verstopfungsrisiko, sowie eine ausgeprägte Anfälligkeit gegenüber Kavitationserosion aufweisen.

Ziel der vorliegenden Dissertation war deshalb die Leistungsbeurteilung und weitere Entwicklung eines wartungsarmen, rohrförmigen Ultraschallreaktors mit Flächenschwingsystem. Zu den potenziellen Vorteilen der neuen Reaktorbauform zählen eine unverbaute Reaktionskammer für ein minimiertes Verstopfungsrisiko, sowie eine geringe Ultraschallintensität (definiert als elektrische Leistung pro Fläche, W/cm^2), für eine niedrige Anfälligkeit für Kavitationserosion. Mögliche Nachteile sind eine potenziell verringerte Leistungsfähigkeit infolge der geringen Ultraschallintensität, sowie eine erschwerte Wiederinstandsetzung erodierter Reaktoren, da die Ultraschallschwinger fest mit dem Rohrreaktor verbaut sind. Für die Durchführung des Forschungsprogramms wurden folgende Forschungsziele definiert:

- Optimierung des Energieeintrags für einen möglichst energieeffizienten Reaktorbetrieb
- Untersuchung der Fluiddynamik von Klärschlamm in der Reaktionskammer
- Optimierung des Reaktordesigns für eine intensive und gleichmäßige Beschallung
- Großtechnische Erprobung des neuen Reaktortyps
- Untersuchung eines alternativen *Co-Behandlungskonzepts* für Faulschlamm

Die Untersuchung zur Optimierung des Energieeintrages konnte zeigen, dass niedrige, großtechnisch anwendbare Energieeinträge (z.B. 200 kJ pro kg Trockensubstanz, TS) eine ebenso hohe Steigerung des biochemischen Methanpotentials von Überschussschlamm von bis zu 12% hervorrufen konnten, wie hohe labortechnische Energieeinträge von 2.000 kJ/kg_{TS} und 3.000 kJ/kg_{TS}. Mittlere Einträge (400 kJ/kg_{TS} bis 1.000 kJ/kg_{TS}) konnten hingegen keine signifikante Steigerung des Methanpotentials erzielen. Des Weiteren ging aus den Ergebnissen hervor, dass nur der niedrigste getestete Energieeintrag von 200 kJ/kg_{TS} einen energieneutralen Betrieb des Ultraschallreaktors ermöglichte, d.h. der Energiegehalt des zusätzlich erzeugten Methangases war nur bei einer Behandlungsintensität von 200 kJ/kg_{TS} größer als der Energieverbrauch des Ultraschallreaktors. Damit konnte das hohe Potential niedriger, großtechnisch anwendbarer Energieeinträge sichtbar gemacht, und die grundlegende Leistungsfähigkeit des neuartigen Ultraschallreaktors bestätigt werden.

Die Untersuchung der Fluiddynamik in Ultraschallreaktoren ergab, dass die hohe Viskosität von Klärschlamm kavitations-induzierte Mikroturbulenzen weitgehend unterdrückt. Damit verbleibt die Schlammströmung in Ultraschallreaktoren weitgehend laminar. In der Konsequenz kommt dem Reaktordesign eine wichtige Funktion zu, da potenziell inhomogene Kavitationsfelder nicht durch eine ausreichende Durchmischung des Fluids ausgeglichen werden können.

Basierend auf diesen Ergebnissen wurde der Einfluss des Reaktordesigns auf die Homogenität und die Intensität der Beschallung bei konstantem Energieeitrag (200 kJ/kg_{TS}) ermittelt. Hierzu wurde ein höhenverstellbarer Flachbettreaktor verwendet, welcher die Untersuchung von Reaktionskammerhöhen von 20 mm bis 100 mm ermöglichte. Die Ergebnisse konnten zeigen, dass eine Vergrößerung der Reaktionskammerhöhe eine Steigerung der Behandlungsinhomogenität und eine Verringerung der Behandlungsintensität hervorruft. Grund für die unterschiedliche Leistungsfähigkeit bei gleichbleibendem Energieeintrag ist die starke Schallfeldabschwächung, welche mit zunehmender Reaktionskammerhöhe immer stärker zum Tragen kommt. Bei einer Reaktionskammerhöhe von 20 mm konnte beispielsweise keine nennenswerte Schallfeldabschwächung ermittelt werden, während bei einer Reaktionskammerhöhe von 100 mm eine völlige Absenz von Beschallungseffekten (wie z.B. Kavitationsrauschen oder Solubilisierung des chemischen Sauerstoffbedarfs) in der Mitte der Reaktionskammer festgestellt wurde. Des Weiteren wurde die Strömung des Schlamms durch die Reaktionskammer mittels einer laminaren Strömungssimulation angenähert. Die Strömungssimulation zeigte auf, dass die Inhomogenität der Beschallung für Durchflussreaktoren noch weiter zunimmt, da die höchsten Volumenströme in den kavitationsärmsten Regionen im Reaktor vorliegen. Als Faustformel konnte damit abgeleitet werden, dass Ultraschallreaktoren für die Klärschlammbehandlung *so klein wie möglich und so groß wie nötig* (für ein minimiertes Verstopfungsrisiko) dimensioniert werden sollten.

Basierend auf den Studien zur Optimierung des Energieeintrags und des Reaktordesigns wurde ein großtechnischer Ultraschallreaktor zur Vorbehandlung von Überschussschlamm konzipiert, und auf einer Kläranlage erprobt. Hierbei wurde eine möglichst typische Kläranlage mit zwei parallel beschickten Faulbehältern ausgewählt, um sowohl wissenschaftlich belastbare, als auch möglichst übertragbare Ergebnisse zu generieren. Die Vorbehandlung des Überschussschlamms mit einem Feststoffgehalt von rund 6% fand im Vollstrom-Verfahren bei einem Energieeintrag von 200 kJ/kg_{TS} statt, und die Behandlungseffekte wurden im Hinblick auf Methanproduktion, Schlammviskosität, Feststoffabbau und Restschlammentwässerbarkeit analysiert. Die Ergebnisse des großtechnischen Versuchs suggerieren, dass die Vorbehandlung entgegen der vielversprechenden Laborergebnisse nur moderate Effekte hervorrufen konnte. Einzig die Viskosität des Überschussschlamms wurde durch die Behandlung signifikant um rund 6% herabgesetzt, was jedoch keine nennenswerten Einsparungen nach sich zog. Die Methanproduktion konnte maximal um rund 6% gesteigert werden, was aber statistisch nicht signifikant war. Für alle anderen Parameter (d.h., die Viskosität des Faulschlamms, den Feststoffabbau und die Restschlammentwässerbarkeit) führte die Beschallung zu keinen messbaren Verbesserungen. Im Hinblick auf die nur minimalen Effekte der Behandlung und die damit insignifikanten Einsparungen war die durchgeführte Überschussschlammvorbehandlung insgesamt unwirtschaftlich.

Die alternative Co-Behandlung von Faulschlamm wurde labortechnisch mithilfe eines Kontroll- und eines Testfermenters in einem kontinuierlichen Versuch analysiert. Trotz eines relativen hohen spezifischen Energieeintrags von 2.000 kJ/kg_{TS} führte die Behandlung nur zu moderaten Verbesserung der Methanausbeute (+9%) und des Feststoffabbaus (+5%). Da die Energiekosten der Ultraschallbehandlung jedoch beinahe zwanzig Mal höher lagen als die Kosteneinsparungen durch zusätzlich erzeugtes Methangas und reduzierte Schlammmengen, war das Verfahren unter den gewählten Versuchsbedingungen ebenfalls unwirtschaftlich. Für einen wirtschaftlicheren Einsatz von Co-Behandlungen sollten zukünftige Studien daher das Potential von Niedrigenergiebeschallungen untersuchen.

Zusammenfassend suggerieren die Ergebnisse, dass das Potential der Ultraschallbehandlung von Klärschlamm mittels rohrförmiger Reaktoren mit Flächenschwingsystem relativ limitiert ist. Der Hauptgrund für die nur moderate Leistungsfähigkeit war möglicherweise die konsequente Orientierung der Versuchsbedingungen an großtechnische Realisierbarkeit (einschließlich niedriger Energieeinträge, hoher Feststoffgehalte der behandelten Substrate, und langer Verweilzeiten im Faulbehälter), was im Umkehrschluss jedoch bedeutet, dass die Ultraschalldesintegration für gut funktionierende Kläranlagen mit typischen Betriebsbedingungen keine effektive oder wirtschaftliche Behandlungsstrategie zu sein scheint. Zukünftige Studien sollten daher den Fokus auf Kläranlagen mit Faulbehältern an der Kapazitätsgrenze und Substraten mit niedrigen Feststoffgehalten legen. Für eine Erprobung der Ultraschalltechnologie unter möglicherweise günstigeren Rahmenbedingungen sollten jedoch auch alternative Behandlungsstrategien untersucht werden, wie z.B. die Unterstützung der biologischen Stickstoffelimination, die Nachbehandlung des Gärrests für bessere Entwässerbarkeit, oder die pyrolytische Entfernung von Schadstoffen in der Industrieabwasserreinigung.

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Contents

Abbreviations

AMPTS	Automatic Methane Potential Test System
ANOVA	Analysis of variance
AOB	Ammonia-oxidizing bacteria
AOP	Advanced oxidation process
BMP	Biochemical methane potential
CFD	Computational fluid dynamics
CNL	Cavitation noise level
COD	Chemical oxygen demand
CST	Capillary suction time
CV(RMSE)	Coefficient of variation (of the root mean square error)
DD_{COD}	Degree of disintegration
DS	Digested sludge
EAC	Exposure to acoustic cavitation
EPS	Extracellular polymeric substances
FM	Fresh matter
GCI	Grid convergence index
HRT	Hydraulic retention time
HSD	Honestly significant difference
NCST	Normalized capillary suction time
NOB	Nitrite-oxidizing bacteria
OLR	Organic loading rate
PE	Population equivalent
PFAS	Poly- and perfluorinated alkyl substance
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctanesulfonic acid
PIV	Particle image velocimetry
PS	Primary sludge
RCH	Reaction chamber height
RCL	Reaction chamber length

RCW	Reaction chamber width
RS	Raw sludge
RSD	Relative standard deviation
sCOD	Soluble chemical oxygen demand
SE	Specific energy
SMP	Specific methane production
SMY	Specific methane yield
THP	Thermal hydrolysis process
TS	Total solids
TSS	Total suspended solids
US	Ultrasound
VS	Volatile solids
VSR	Volatile solids removal
WAS	Waste activated sludge
WWTP	Wastewater treatment plant

CHAPTER 1

General introduction - Current challenges of sewage sludge management

1.1 Sludge production during conventional wastewater treatment

The production of vast amounts of sewage sludge as an inevitable by-product of conventional wastewater treatment has become a major challenge for wastewater treatment plant (WWTP) operators worldwide, both from an economical and an environmental perspective [1]. Over 10 million tons (dry weight) of sewage sludge are, for instance, produced each year in the European Union [2], while similar annual production rates can be found for the United States (8.2 million tons of dry sewage sludge), or China (40 million tons of dewatered sewage sludge) [3]. In the future, the global sewage sludge production is projected to even rise due to the continued commissioning of new WWTPs, especially in developing countries, or the increase of effluent quality standards [4].

On the one hand, the produced sewage sludge poses a severe environmental concern, by containing a multitude of different contaminants, including pathogenic microorganisms (bacteria, viruses, and protozoa), trace organic chemicals (for instance, pharmaceutical residues), heavy metals, and various other anthropogenic pollutants such as microplastics [5, 6]. Owing to the high organic content, the sludge is furthermore characterized by high biological activity and strong odor emissions. On the other hand, the sludge is also a valuable and energy-rich resource due to the contained nutrients (such as phosphorus) and its high biochemical methane potential (BMP), which can be used to recover a significant part of the energy required for wastewater treatment [7, 8].

The two main sludge types produced during conventional wastewater treatment are primary sludge (PS) and waste activated sludge (WAS) [9]. PS is formed during primary sedimentation, where roughly 50% - 60% of the suspended solids present in the raw wastewater is settled to form a highly putrescible sludge with a solids content of typically 1% - 3% [9]. WAS originates from the secondary clarifiers, where activated sludge from the biological treatment step is sedimented to be either recycled to the activated sludge tank (for maintaining a sufficient biomass concentration and sludge age) or removed from the process as excess sludge. The sedimented WAS reaches a solids content of around 1% - 2% and mostly comprises aerobic microbial biomass, extracellular polymeric substances (EPS), and wastewater compounds that were not degraded during the biological treatment step [10]. Due to the recalcitrant nature of microbial cells and EPS, WAS is characterized by a comparably low biodegradability [11]. The combination of PS and WAS is termed raw sludge (RS).

1.2 Sewage sludge stabilization via anaerobic digestion

Owing to its adverse properties and poor dewaterability, RS requires further treatment before it can be disposed [12]. In most modern WWTPs, such treatment (or stabilization) is conducted via anaerobic digestion of the RS [11, 13, 14]. During the four-stage digestion process (comprising hydrolysis, acidogenesis, acetogenesis, and methanogenesis), the organic sludge substances are successively degraded to yield energy-rich biogas with a methane content of 55% - 70% and biologically stabilized digestate [15, 16]. The generated biogas is typically valorized on-site for electricity and heat production by the use of combined heat and power plants, while the digestate is disposed after a final dewatering (and sometimes drying) step [11, 13, 17].

In addition to electricity production and sludge stabilization, further merits of anaerobic sludge digestion are (i) better dewaterability of the digestate, (ii) mass reduction of biosolids, (iii) improved storability and transportability of the sludge residues, and (iv) removal of pathogens [6, 18]. Hereby, anaerobic digestion enables significant cost savings at WWTPs and is considered an essential part of modern wastewater treatment [11]. An example of a full-scale anaerobic digestion system with four digesters at the Gut Großlappen WWTP in Munich, Germany, is presented in Figure 1.1.

1.2.1 Remaining (economic) challenges of anaerobic digestion

Despite the aforementioned advantages of anaerobic digestion, the process still faces several impediments. A major drawback is the slow reaction rate, which requires long hydraulic retention times (HRTs) in the digesters of 20 d - 50 d for sufficient sludge stabilization. Nonetheless, the degradation of organic sludge substances is still incomplete, and typically, only about half of the organic matter is removed and valorized to biogas during anaerobic digestion [19]. As a consequence of the slow and incomplete digestion process, the construction of huge digesters is necessary, which entails considerable construction and maintenance costs. For instance, the four digesters depicted in Figure 1.1 comprise a total

Figure 1.1: Anaerobic digesters at the Gut Großlappen WWTP in Munich, Germany (photographed by Philipp Weigell[1]).

volume of 58,000 m^3 and caused project costs of roughly 63 million € [20].

Besides the costs for the digester infrastructure, the management costs of the remaining digestate represent another considerable financial burden and can account for up to 50% of the operating costs of a WWTP [11, 21]. These costs mostly comprise dewatering, drying, transport, and disposal costs, whereas especially the latter is expected to rise further due to increasingly stricter legal requirements for sludge disposal [6]. For instance, the comparably cheap land application of sewage sludge will be inadmissible in Germany for all WWTPs larger than 50,000 population equivalents from the year 2032 onward, so that more and more plant operators need to switch to (currently) more expensive disposal routes, such as co- or mono-incineration. Currently, approximately 65% of the sewage sludge produced in Germany is already disposed via incineration [6].

Such trend towards incineration also has critical economic implications on anaerobic digestion. On the one hand, thorough stabilization remains important regarding sufficient digestate dewaterability and, thus, minimum sludge masses for disposal. On the other hand, an overly pronounced organic matter degradation and biogas production during anaerobic digestion might even be somewhat counterproductive due to the decreased calorific value of the digestate [22, 23]. Thus, for most economic sludge management, on-site benefits of anaerobic digestion such as biogas production also need to be balanced against potential cost increases during sludge disposal.

[1] Image available at: https://de.wikipedia.org/wiki/Datei:Faulturmanlage_im_Kl%C3%A4rwerk_Gut_Gro%C3%9Flappen.jpg

1.2.2 Hydrolysis - the bottleneck of anaerobic sludge digestion

The main reason for the slow degradation kinetics of anaerobic digestion (and hereby for most of the remaining economic drawbacks) is the initial hydrolysis step, which often becomes rate-limiting when particulate or colloidal substances such as sewage sludge are digested [24]. During hydrolysis, particulate, insoluble polymers (such as carbohydrates, proteins, and fats) are transformed into soluble di- and monomers (such as monosaccharides, amino acids, and fatty acids) through the enzymatic activity of hydrolytic bacteria [4]. This already complex process that requires specific bacterial exoenzymes for hydrolyzing specific polymers (for instance, proteases and lipases for the degradation of proteins and lipids, respectively [18]) is further impaired by the recalcitrant characteristics of sewage sludge. Especially thickened WAS features a distinct floc structure, which reduces surface area and shields the biomass within the flocs from hydrolytic microorganisms [25]. In addition to the poor morphological availability, WAS comprises mostly bacterial biomass with intact cell walls, which constitute of recalcitrant peptidoglycans and complex polysaccharides, thus resembling an effective barrier against hydrolytic degradation. Furthermore, EPS are recalcitrant by nature, by being predominantly constituted of complex biopolymers such as proteins and humic substances [26–28]. A schematic of a WAS floc demonstrating its unfavorable morphology and recalcitrant features is depicted in Figure 1.2.

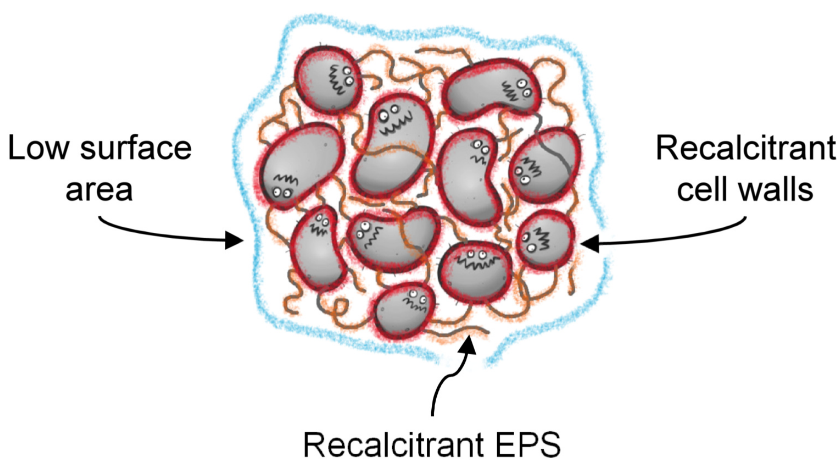

Figure 1.2: Illustration of a waste activated sludge floc and its recalcitrant properties, with EPS = extracellular polymeric substances.

1.3 Sewage sludge pre-treatment - a viable solution for enhanced sludge management?

A technological solution to enhance the initial hydrolysis step of anaerobic digestion is sewage sludge pre-treatment. Through pre-treatment, floc agglomerates and bacterial cell walls can be ruptured, thus aiding the rate-limiting hydrolysis step by an increase in surface area and a release of intracellular matter [29, 30].

1.3.1 Pre-treatment goals

Naturally, the paramount goal of applying pre-treatments is the acceleration of the digestion process [4, 12, 19]. Once reaction kinetics are increased, the same digestion performance (for instance, regarding methane production, biosolids degradation, or dewaterability enhancement) can be attained at shorter HRTs, thus increasing the capacity of existing digesters or allowing to build smaller, more cost-effective digesters. For digesters operating at capacity limit, pre-treatments may even help to avoid the construction of an additional digester.

Other, sometimes equally important pre-treatment goals can be foaming prevention in the digester [31, 32], a further improvement of digestate dewaterability [33, 34], or the reduction of sludge viscosity [35, 36]. Especially an additional improvement of dewaterability is highly auspicious, as it allows for reduced residual sludge masses with a higher calorific value and, ultimately, for reduced disposal and incineration costs [23]. A reduced sludge viscosity, on the other hand, may entail savings due to lower costs for stirrers and pumps or may similarly contribute to an enhanced digestion process, due to better and faster mixing of the feed sludge with the anaerobic biomass [37].

Regarding the current trend towards incineration as the main disposal method [6], an intensification of methane production and organic matter removal through pre-treatment must also be seen as critical, since a lower organic content in the digestate results in a lower calorific value and a decreased incineration efficiency. In this case, pre-treatments would only be economically feasible once the benefits (such as increased electricity production through additional methane or improved digestate dewaterability) are able to compensate both the pre-treatment and the increased disposal costs.

1.3.2 Pre-treatment methods

Numerous pre-treatment methods exist for enhancing sludge hydrolysis, including chemical, biological, thermal, and mechanical methods [19]. For municipal sewage sludge treatment, thermal hydrolysis processes (THPs, belonging to the thermal methods) and ultrasonic treatment processes (belonging to the mechanical methods) resemble by far the most applied and researched pre-treatment strategies [10].

1.3.2.1 Thermal hydrolysis

THPs rely on heating the sludge to 160°C - 190°C for 20 min - 30 min, which causes pronounced solubilization of chemical oxygen demand (COD), a decrease of sludge viscosity, and a reduction of pathogens [10]. The main merits include enhanced reaction kinetics, increased biogas yield, and an improved dewaterability of the digestate [10, 26]. A drawback is the potential formation of persistent compounds due to so-called Maillard reactions at temperatures above 170°C, which may render parts of the organic material inaccessible for biodegradation and methane production [38]. THPs were commercialized by several companies (such as Cambi or Veolia) and are installed at several WWTPs worldwide [26].

From a practical point of view, THPs represent a rather complex pre-treatment strategy, and, for instance, the Cambi THP can easily entail investment costs of several million € and cause considerable space requirement of hundreds of square meters [39]. As complete heat integration (i.e., utilization of the heat produced during the THP for digester heating) is furthermore essential for an economical use of the technology, considerable structural alterations at WWTPs might be required [40].

1.3.2.2 Ultrasonic sludge disintegration

From the available mechanical pre-treatment technologies (including ultrasonic treatment, lysis-centrifuges, shear shredders, and collision plates), ultrasonic methods resemble by far the most applied in full-scale WWTPs [26, 41].

The reported main merits of ultrasound (US) pre-treatments are (i) high efficiency, (ii) environmentally friendly operation with no added chemicals, and (iii) easy retrofitting of existing WWTPs due to very compact reactor dimensions [10, 19, 42]. The treatment method relies on the phenomenon of transient acoustic cavitation, i.e., the formation and violent collapse of micro-bubbles present in the sludge. Due to the violence of the bubble implosion, powerful shear forces are released into the sludge which mechanically disintegrate both sludge flocs and bacterial cell walls.

Despite the advantages of the technology, the large-scale use of ultrasonic reactors in WWTPs still faces several obstacles, which is mostly related to operational challenges or the high maintenance requirements of US reactors [43]. Especially conventional radial horn and sonotrode systems were reported to be susceptible to clogging and fast erosion of the sound-emitting surfaces, thus causing limited reactor lifetime, troublesome operation, and necessitating frequent and costly service cycles [31, 44–46].

In the light of these remaining challenges, the overarching objective of this dissertation was the investigation and further optimization of novel tube-shaped US reactors with surface transducers, which are hypothesized to exhibit higher erosion-resistance and minimized clogging risk.

CHAPTER 2

State-of-the-art of ultrasonic sewage sludge disintegration

2.1 Theory

Ultrasonic sewage sludge disintegration relies on the phenomenon of transient acoustic cavitation, which is defined as the consecutive formation, growth, and violent implosion of acoustic micro-bubbles in a liquid medium [47]. A short introduction to all processes involved in acoustic cavitation is given in the following.

2.1.1 Bubble formation

According to the above definition, the starting point of acoustic cavitation is the formation of acoustic bubbles in a liquid. In practice, an abundance of microscopic bubbles already exists in the liquid, which particularly applies to pre-aerated media such as WAS. When entrapped in crevices of the sludge flocs or when covered by a stabilizing skin of surfactants or hydrophobic impurities, some of these microscopic gas bubbles may withstand buoyancy effects or natural dissolution processes and remain available for the acoustic cavitation process [48]. Another source for such pre-existing micro-bubbles is the fragmentation of large cavitation bubbles during collapse events into clouds of microscopic bubble nuclei [49]. Yet, most of these microscopic bubbles are relatively stable due to their small size and the accordingly high influence of the confining surface tension pressure (or *Laplace pressure*), which means that, at this stage, the bubbles are not likely to implode in transient cavitation events [48].

2.1.2 Bubble growth

To become active transient cavitation bubbles, a growth of the bubbles to a critical size is necessary, which can be initiated by imposing an acoustic field on the liquid. Due to the varying pressure of the sound field, the micro-bubbles will start oscillating, thus commencing a process termed rectified mass diffusion [50]. During the rarefaction phase, the reduced liquid pressure

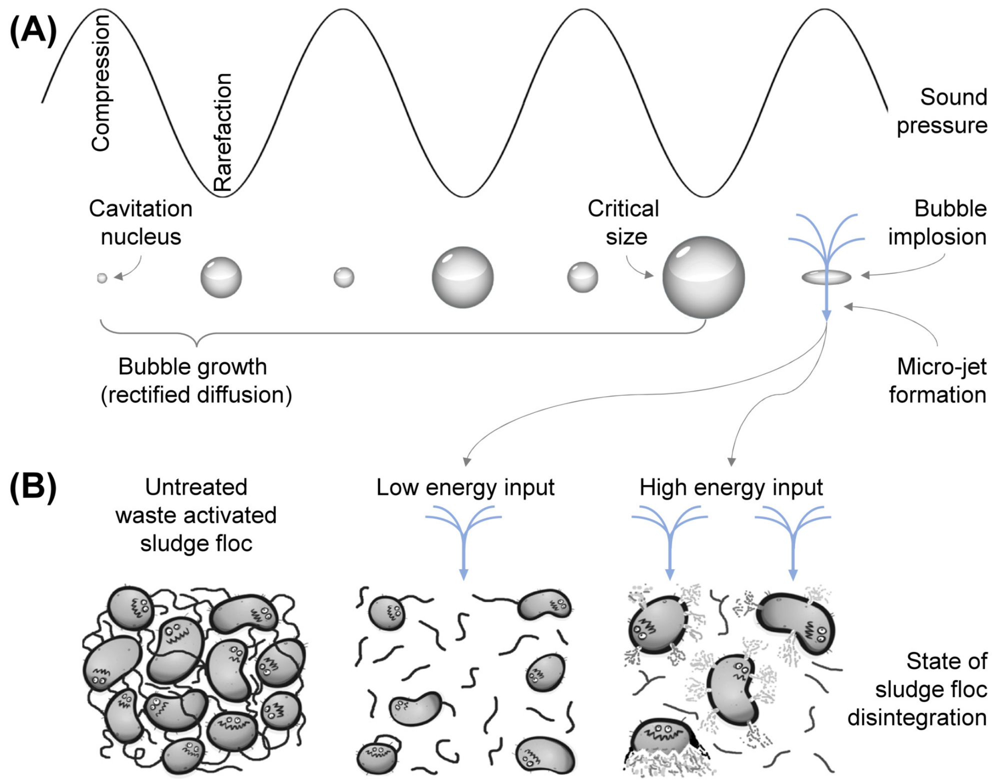

Figure 2.1: Principle of rectified mass diffusion (subfigure A) and different stages of sludge disintegration (subfigure B), including an intact sludge floc, a de-agglomerated floc after sonodispergation (low energy input treatment) and a completely disintegrated floc with severe cell lysis (high energy input treatment).

causes an expansion of the bubble. Through the volume increase, the concentration of gas and water vapor inside the bubble declines, and an inward concentration gradient between bubble and liquid develops. Thus, dissolved gas and solvent vapor from the surrounding liquid will enter the bubble. In the subsequent compression phase, the volume of the bubble decreases again, thus reversing the concentration gradient and causing a back-diffusion of mass into the liquid. However, this efflux is slightly lower than the previous mass influx so that the bubble experiences a net increase in mass and, thus, a net growth [48, 50]. A schematic representation of the process is displayed in Figure 2.1A.

The two mechanisms responsible for the directionality of the diffusion process are the "area effect" and the "shell effect" [50]. The area effect refers to the increased surface area (or diffusion boundary) of the bubble during rarefaction, which allows for higher mass diffusion rates than in the compression cycle. The shell effect refers to a squeezing of the shell of fluid surrounding the bubble during expansion, causing an increase in dissolved gas concentration in the shell. Thus, concentration gradients (and hence, diffusion rates) are larger during rarefaction than during compression. Due to both effects, a continuous growth of the bubble takes place over several acoustic cycles. Naturally, this growth cannot proceed indefinitely, and at a certain point, the bubble reaches an unstable (resonant) size, where the natural oscillation frequency of the bubble is equal to the frequency of the driving US field. At this point, the bubble will grow explosively to a maximum, highly unstable size within one acoustic cycle [49].

2.1.3 Bubble collapse and its effects on sewage sludge

Due to the rapid bubble growth at resonance size, the pressure in the bubble is strongly reduced. In consequence, water from the surrounding shell of fluid evaporates into the bubble until the growth is terminated in the subsequent compression phase. Due to the imposition of elevated pressure, the water vapor condensates again, thus causing an instant and dramatic volume reduction of the bubble. In consequence, the surrounding liquid is forcefully sucked into the collapsing cavity [51].

When collapsing bubbles are attached to solid surfaces (which is a regular case when solids-rich substrates like sewage sludge are sonicated), the water typically enters the bubble at the opposite side of the solid surface. Hence, the bubble is not experiencing a concentric collapse but is rather pierced by a powerful jet of water that is directed towards the solid particle [51]. Depending on the bubble size, such micro-jets can easily reach velocities of over 100 m/s [52, 53], causing a tremendous release of shear forces, heat, and pressure. Local temperatures as high as 4,000 K - 15,000 K [54] and pressures of up to 1.3 GPa [55] were measured during the cavitation bubble collapses.

Through these extreme physical forces, the sludge flocs are disintegrated in a two-stage process, including sonodispergation (i.e., the de-agglomeration of the floc structure) and sonolysis (i.e., the rupture of the microbial cell walls) [56]. A schematic illustration of the two-stage disintegration process is given in Figure 2.1B.

2.1.4 Employed frequencies

Among various factors such as power input, operating pressure, or substrate characteristics (including pH, temperature, and solids content [4]), ultrasonic frequency represents an especially important governing factor and typically, frequency ranges of of about 20 kHz to 40 kHz were found to be most suitable for efficient sewage sludge sonication [12, 19]. At such ranges, relatively large cavitation bubbles are formed, because the resonant bubble size is inversely proportional to the driving US frequency [57]. As collapse velocity (and, hence, collapse intensity) is a function of resonant bubble size, mechanical effects can be maximized at such low US frequencies [51]. This is advantageous for sewage sludge sonication, as sludge disintegration was shown to be mainly caused by mechanical forces rather than by free radicals, which are predominantly formed at higher frequencies (above 100 kHz [19, 25, 58]).

2.2 Ultrasonic sludge treatment in lab-scale environments

Literature on lab-scale US pre-treatment of sewage sludge is extensive, and numerous reviews are available which provide a detailed overview of the methods applied and results obtained in laboratory experiments (see Pilli et al. [4], Carrère et al. [10], Le et al. [12], Tyagi et al. [19], Carrère et al. [26], Khanal et al. [27], Carlsson et al. [59], Elalami et al. [60], Zhen et al. [61]). A brief overview of the current state of lab-scale research (which can never be exhaustive given the large number of articles published) is presented in the following.

2.2.1 Experimental conditions

2.2.1.1 Treated sludge types

Traditionally, US is applied as a pre-treatment for WAS due to its especially recalcitrant character [12, 19]. Accordingly, pre-treatment of the comparably well-degradable PS is only seldomly conducted [26, 62]. An exception is RS pre-treatment, where a mixture of WAS and PS is sonicated [63]. A further, relatively new treatment scheme is co- or inter-treatment, where digested sludge (DS) is sonicated in the recirculation loop of an anaerobic digester or the overflow from a first to a second digester, respectively. In contrast to WAS pre-treatment, however, co- or inter-treatment is not well researched yet, and only a few studies have addressed this alternative treatment scheme to date [43, 64–67]. Further substrates that are sometimes treated with US are agricultural sludges or manure [43, 64, 68].

2.2.1.2 Laboratory reactor configurations

In laboratory studies, US is usually induced into the sludge through sonotrode tips that are submerged into a sludge containing vessel [4, 69–75]. Alternative reactor configurations with externally mounted transducers such as ultrasonic baths [56], tube-shaped flow-through reactors [43, 76], or flatbed reactors [77] are employed less frequently.

Hereby, it should be noted that generally only little attention has been paid in laboratory studies how reactor configuration affects sonication performance. At the same time, the results of several studies indicated that reactor design has a defining impact on sonication efficiency

[42, 56, 78]. Nonetheless, a clear understanding of how reactor design affects sonication efficiency has not been obtained yet and an in-depth discussion on the role of (geometric) US reactor design on disintegration efficiency is not provided in any of the above-listed review papers.

2.2.1.3 Applied energy inputs

Several parameters are available for describing the energy input of US treatments, including specific energy input (Joule per kilogram of total solids, [kJ/kg_{TS}]), ultrasonic dose (Joule per liter of sludge volume, [J/L]), ultrasonic density (Watt per liter of sludge volume, [W/L]), or ultrasonic intensity (Watt per square centimeter of transducer surface, [W/cm^2]) [12]. Among these expressions, the specific energy input is the most frequently used one, as it allows to account for both the electrical power of the US reactor and the properties of the treated sludge:

$$SE = \frac{P \cdot t}{TS \cdot V \cdot \rho} \tag{2.1}$$

where SE is the specific energy [kJ/kg_{TS}], P is the electrical power of the applied US system [kW], t is the treatment duration [s], TS is the total solids content [kg_{TS} per kg of fresh matter, FM], V is the tallied volume [L] and ρ is the density [kg_{FM}/L].

A typical range of specific energy inputs applied in the lab is 1,000 - 16,000 kJ/kg_{TS} [19, 26], while several studies also applied much higher inputs (for instance, 27,000 kJ/kg_{TS} in Ruiz-Hernando et al. [36], 30,500 kJ/kg_{TS} in Neumann et al. [79], or even 108,000 kJ/kg_{TS} in Salsabil et al. [80]). Such inputs are, however, extremely energy intensive and in order to treat, for instance, only one liter of sludge with a TS content of 5% with an energy input of 100,000 kJ/kg_{TS}, a US reactor with an electrical power of 1,000 W and a treatment time of approximately 83 min (= 5,000 s) would be necessary. Assuming that all ultrasonic energy is ultimately transformed into heat, such 100,000 kJ/kg_{TS}-treatment would heat up the sludge by almost 1,200 K (assuming a specific heat capacity of 4.18 kJ/(kg·K)), which clearly illustrates the extraordinary energy intensity of such treatment setting.

A specific energy input of 1,000 kJ/kg_{TS} is often reported as *cell lysis threshold* [25, 72, 81], thus marking the difference between the two disintegration mechanisms of sonodispergation (i.e., a de-agglomeration of sludge flocs) and sonolysis (i.e., a rupture of the microbial cell walls and a release of intracellular matter) [56]. As cell lysis is generally seen as the predominant functional principle of ultrasonic sludge treatment [4, 12, 19], only few studies addressed energy inputs below the cell lysis threshold to date [29, 72, 82, 83]. However, with respect to full-scale applications which are typically limited to rather short treatment times, specific energy inputs below this threshold of 1,000 kJ/kg_{TS} are by far the most relevant range [45, 84, 85].

2.2.2 Treatment effects

2.2.2.1 Effects on sludge solubilization

The degree of sludge solubilization is often seen as a general performance indicator of US pre-treatment [10, 12, 19]. Typically, solubilization is expressed as degree of COD disintegration (DD_{COD}):

$$DD_{COD} = \frac{sCOD_{US} - sCOD_0}{sCOD_{NaOH} - sCOD_0} \tag{2.2}$$

where $sCOD_{US}$ is the concentration of soluble COD (sCOD) after sonication, $sCOD_0$ is the initial concentration of sCOD of the untreated sludge sample, and $sCOD_{NaOH}$ is the sCOD concentration of the sludge sample after alkaline hydrolysis.

Several authors reported a quasi-linear relationship between specific energy input and DD_{COD} [81, 82, 86], and accordingly, DD_{COD} values can range from 5% - 10% at low energy inputs below 1,000 kJ/kg_{TS} [82, 83] to almost 100%, when extremely high energy inputs (such as 100,000 kJ/kg_{TS}) are employed [81, 86].

Although DD_{COD} is often used as a key indicator for the efficacy of US pre-treatment, several authors state that an increased DD_{COD} value does not necessarily lead to improved digestion performance. On the one hand, the solubilized COD may constitute recalcitrant organic substances which are not contributing to enhanced digestion performance. On the other hand, the treatment may solubilize compounds that are already easily soluble so that the treatment would not have been necessary in the first place [43, 87]. Moreover, it should be noted that the de-agglomeration of sludge flocs at low specific energy inputs does not lead to notable COD solubilization [81], but can already positively affect anaerobic digestion performance due to increased surface area [25, 72].

2.2.2.2 Effects on methane production

Effects of US treatment on methane production were mostly investigated by the use of BMP tests in batch mode [29, 43, 68, 70, 88, 89], but several studies also employed (semi-)continuous digestion tests to examine pre-treatment effects [71, 74, 90, 91]. Hereby, batch tests were shown to somewhat overrate pre-treatment effects as compared to continuous digestion tests [92–94]. For instance, reported percentage increases in methane production range from 20% to 140% in batch tests [73, 80, 95–98], and from 10% to 40% in continuous tests [25, 63, 64, 91]. A potential explanation for the considerable offset may be adverse long-term effects caused by the pre-treatment (such as the release of inhibitory compounds) that can only be detected in continuous digestion tests [43, 64]. Regardless of the offset between batch and continuous tests, previous results generally indicate US pre-treatment significantly enhances methane production, albeit at quite high ranges of specific energy input (1,000 - 16,000 kJ/kg_{TS}).

2.2.2.3 Effects on reaction kinetics

Several studies addressed the impact of US pre-treatment on the reaction kinetics of anaerobic digestion by analyzing VS removal under varying HRTs. Apul and Sanin [70] demonstrated that US pre-treatment of WAS enabled to reduce the HRT of a semi-continuous digester from 15 d to 7.5 d without causing a notable drop in VS removal efficiencies (55% VS reduction at 15 d vs. 53% VS reduction at 7.5 d). For the untreated control reactor, on the other hand, VS reduction dropped to 38% once HRT was shortened. Hence, results suggest that US pre-treatment is able to maintain stable digestion performance even with a drastically reduced digester volume.

Neis et al. [99] achieved similar results for WAS pre-treatment and preserved the same VS degradation despite a massive reduction in HRT from 16 d to 4 d. While such result is highly promising, it should also be noted that the sonicated feed sludge featured verifiably low TS contents of 0.7% to 2.6% and that high energy inputs ranging from about 9,700 kJ/kg_{TS} to 36,000 kJ/kg_{TS} were employed.

Braguglia et al. [91] performed a similar experiment at specific energy inputs of 2,500 kJ/kg_{TS} and 5,000 kJ/kg_{TS}. At an HRT of 20 d, the destruction of VS amounted to 36% (relative to VS_{fed}) for a non-sonicated control digester, and to 39% for a sonicated digester. When reducing HRT to 10 d, VS removal efficiency dropped to 30% for the control digester, while for the test digester, VS removal remained at 35%. Thus, VS removal of the test digester at an HRT of 10 d was almost equally high as for the non-sonicated control digester at an HRT of 20 d.

Overall, the results clearly indicate that US pre-treatment can strongly accelerate reaction kinetics and reduce HRTs (and thus digester volumes), while most of the employed energy inputs again appear quite high.

2.2.2.4 Effects on viscosity

Sludge viscosity is typically reduced through US pre-treatment. For instance, Ruiz-Hernando et al. [100] examined the impact of specific energy inputs ranging from 3,000 kJ/kg_{TS} to

30,000 kJ/kg_{TS} on WAS viscosity and found that increasing energy input causes a constant decrease in viscosity. However, the relationship was not linear and above 18,000 kJ/kg_{TS}, only small incremental reductions of sludge viscosity were observed when further increasing energy inputs. Similar findings were reported by Ruiz-Hernando et al. [36] and Pham et al. [101].

2.2.2.5 Effects on dewaterability

Regarding dewaterability, US can entail both positive and negative effects. Generally, there is consensus that low energy inputs can contribute to improved dewaterability, while high energy inputs tend to deteriorate digestate dewaterability [12, 19, 83].

For instance, Feng et al. [102] reported an improved sludge dewaterability for energy inputs below 2,200 kJ/kg_{TS}, but a severely decreased dewaterability above an energy input of 4,400 kJ/kg_{TS}. It was hypothesized that at low inputs, the interstitial water content can be released which improves dewaterability, while at high energy inputs, the decrease of particle size and the release of intracellular matter causes an increased adsorption of water to the solid particles, and thus, a deteriorated sludge dewaterability [102, 103].

2.2.3 Remarks on transferability of lab-scale results to full-scale applications

While the above-listed lab results appear quite promising, their direct transferability to full-scale applications may be limited. A prime reasons for the limited transferability is the distinct offset between energy inputs applied in laboratory studies (several thousand kJ/kg_{TS} [10, 19]) and full-scale studies (typically much less than 1,000 kJ/kg_{TS} [45, 84, 85]). As specific energy input and several performance indicators (such as DD_{COD} or methane production) were shown to correlate [4, 12, 19], an extrapolation to full-scale tests is, hence, difficult. Second, the transferability of batch tests to continuous systems such as full-scale digesters is limited, as batch tests were shown to overrate pre-treatment effects [92–94, 104]. Third, the efficiency of different reactor geometries and lab- and full-scale US reactors was reported to be quite different [42, 56, 78, 97] so that the effects for a particular specific energy input might strongly depend on the employed reactor type. For instance, Nickel and Neis [78] reported that a full-scale reactor had twice the disintegration efficiency of a lab-scale reactor, despite the same specific energy input and the same sonotrode used. Fourth, most lab-scale studies were conducted with rather dilute WAS samples with average TS contents of about 2% (see review papers by Tyagi et al. [19] and Le et al. [12]), while TS concentrations of thickened WAS at WWTPs typically amount to 5% - 6% [36, 105]. As the difference in solids content has a significant impact on sound wave attenuation and disintegration efficiency [14, 99], comparability may again be hampered.

2.3 Ultrasonic sludge treatment in full-scale environments

2.3.1 Reactor configurations

In industrial practice, ultrasonic sludge treatment is commonly conducted by the use of flow-through US reactors. Hereby, a distinction is generally made between US reactors with rod-shaped transducers (sonotrodes and radial horns) and US reactors with surface transducers. In the case of rod-shaped transducers, US is induced into the sludge through comparably small sonotrode tips or horns [12], while in flat transducer systems, large oscillating surfaces (such as the reaction chamber housing) are responsible for sound induction [106].

For sludge treatment in full-scale WWTPs, the most commonly applied reactor types are sonotrode or radial horn systems [12], which is mostly due to their high ultrasonic intensity (25 - 50 W/cm^2 for sonotrodes [45], and 13.7 W/cm^2 for radial horns [84]), allowing for a strong cavitation field and, thus, intense sludge disintegration [99].

In sonotrode reactors, the sludge is conveyed through a series of tubular reaction chambers (similar to a marble run layout), while at the end of each chamber, a sonotrode is immersed into the flow path for sound induction (Figure 2.2A). Thus, the sludge is exposed to high-intensity cavitation fields for multiple times when passing through the reactor. However, due to strong sound wave attenuation, cavitation hotspots are limited to the close vicinity of the sonotrode tips so that cavitational activity within large-scale sonotrode reactors is rather inhomogeneous [106].

Radial horn reactors, on the other hand, feature torus-shaped sonotrodes that enclose the flow path of the sludge within a tubular reaction chamber, i.e., the sludge is basically flowing through the sonotrode. Yet, the conveyance of the sludge through the comparably small horn diameters results in narrow, operationally challenging flow paths. Typical reactors feature several radial horns in series [12, 84, 107], which are agitated by external transducers commonly arranged in a V-shaped layout (Figure 2.2B).

A main drawback of the high-intensity sonotrode and radial horn reactors is the fast erosion of the sound-emitting surfaces. As a consequence, both sonotrodes and radial horns require frequent replacement, which entails considerable maintenance costs [31, 44]. On the other hand, the performance of the reactor systems can be fully restored by a new set of sonotrodes or radial horns so that the much more expensive replacement of the entire reactor can be avoided.

Besides erosion susceptibility, a further hindrance is the considerable clogging risk of the two reactor types, which is a result of the obstructed and narrow flow paths [31, 44, 46].

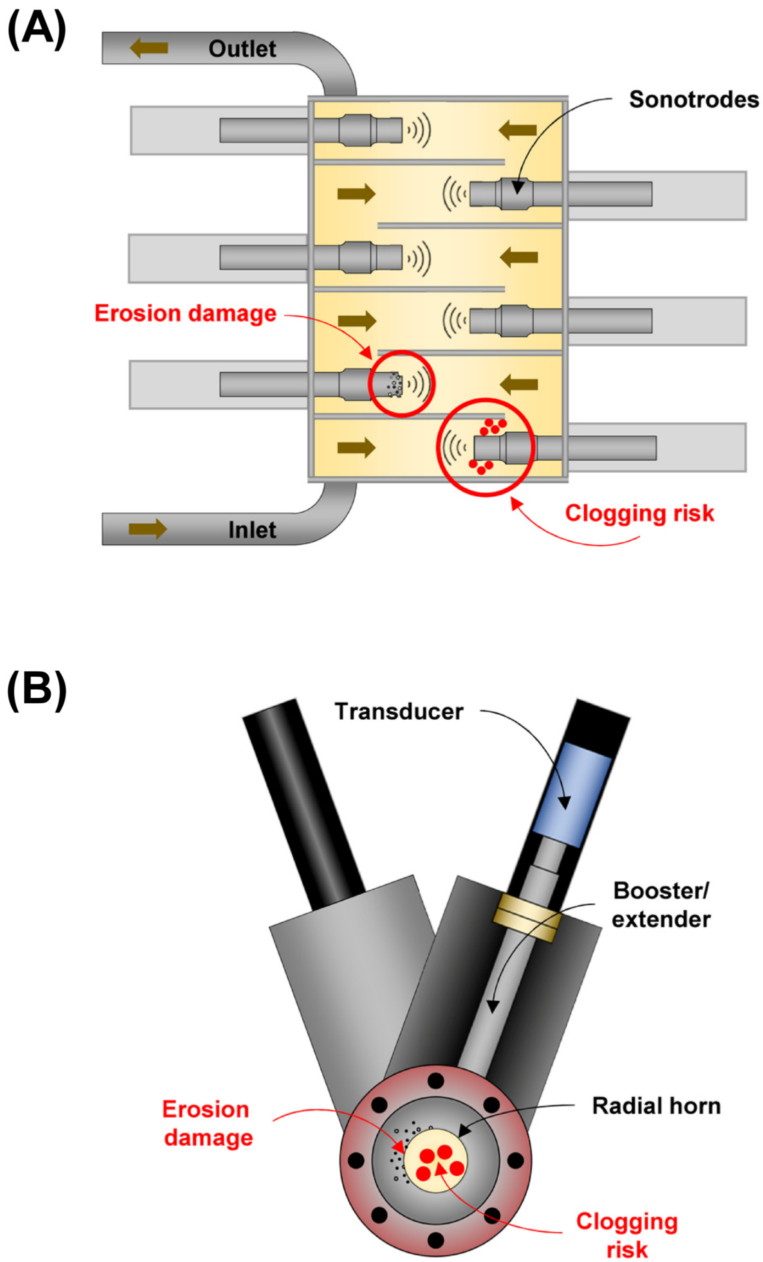

Figure 2.2: 2-D schematics of a conventional sonotrode reactor (A, side view) and a radial horn reactor (B, front view), showing areas of increased clogging risk and erosion damage.

To avoid clogging events, a common strategy is the installation of a macerator before the US reactor [108], which, however, increases investment and electricity cost. Moreover, as maceration is a mechanical pre-treatment method in itself, a clear distinction between the effects of US treatment and the effects of maceration might be difficult to attain [41].

An alternative and relatively novel reactor concept is resembled by US reactors with surface transducers. The main difference to sonotrode reactors is that the US transducers are mounted externally at the reaction chamber [106] so that an entirely unobstructed flow path through the reactor can be realized. Thus, when designed with a diameter similar to a pre-existing sludge pipe, such reactor configuration does not introduce any additional clogging risk. A schematic of such reactor type is presented in Figure 2.3.

Due to the widely distributed array of US transducers over the reactor housing, US intensity is lower than for sonotrode or radial horn reactors and typically amounts to less than 1 W/cm^2 [76]. As a consequence, the mechanical stress on the sound emitting surfaces is reduced, which potentially reduces erosion susceptibility and prolongs the reactor lifetime. In case erosion damage occurs nonetheless, a potential downside is that the US transducers are firmly fixed to the outside of the reactor wall. Thus, the replacement of the whole reactor might be necessary, which presumably entails much higher costs than the exchange of sonotrodes or radial horns.

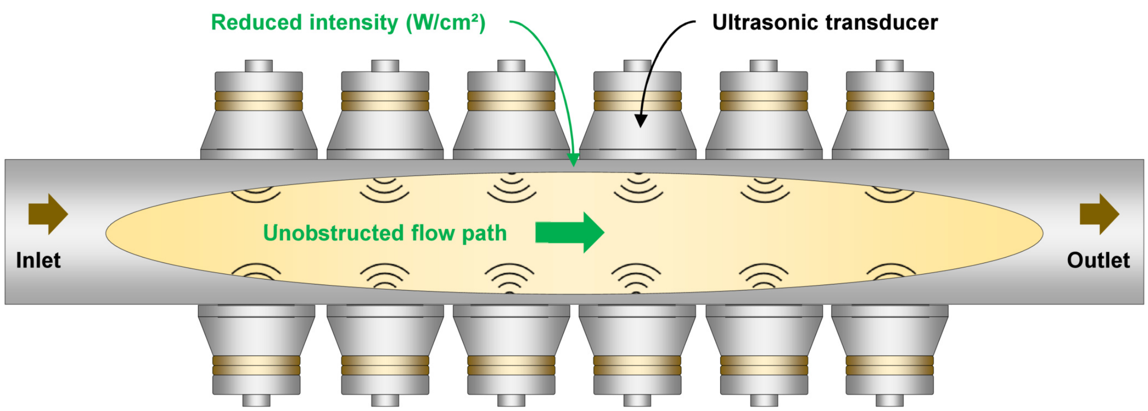

Figure 2.3: Schematic of a tube-shaped flat transducer reactor.

The low intensity could also be disadvantageous, however, as sludge disintegration and US intensity were shown to be strongly correlated [99]. On the other hand, US reactors with multiple surface transducers are generally recommended for full-scale applications due to the more homogeneous cavitation field and, thus, the more homogeneous exposure [106].

Despite the operational advantage with minimal clogging risk and potentially higher erosion resistance, available literature on US reactors with surface transducers is scarce and to date, no full-scale study and only three laboratory studies employed such reactor type for sludge treatment [29, 43, 76].

2.3.2 Experiences from full-scale trials

Ultrasonic sludge treatment has been commercialized by several companies (such as Ultrawaves Water & Environmental Technologies GmbH, Karlsbad, Germany, WEBER ENTEC GmbH & CO. KG, Waldbronn, Germany, or VTA Austria GmbH, Rottenbach bei Haag, Austria) and a number of full-scale US reactors has been implemented in WWTPs worldwide [12, 19]. At the same time, proper company-independent documentation is only available for a few full-scale trials so that it remains challenging to assess whether US is an effective sludge pre-treatment strategy at full-scale WWTPs or not.

Up to now, full-scale US treatment was only studied as a pre-treatment method for WAS and the only reactors investigated were radial horn or sonotrode reactors [45, 46, 84, 107, 109–111]. Most of the existing full-scale studies confirmed the promising treatment effects observed in the lab and, for instance, Xie et al. [84] reported a marked increase in methane production between 13% and 58% after pre-treating RS with a 30 kW radial horn reactor (at approximately 400 kJ/kg_{TS}) at the Ulu Pandan Water Reclamation Plant in Singapore. Similar percentage increases of up to 45% were observed by Neis [45], who increased biogas production at the Bamberg WWTP in Germany from 1.5 million m^3/a to 2.2 million m^3/a through side-stream sonication of thickened WAS by the use of a 10 kW sonotrode reactor (at approximately 150 kJ/kg_{TS}). In accordance to the boosted biogas production, also organic matter degradation was increased from about 34% to 50% [45]. The results are in line with Hogan et al. [111], who summarized the findings of several full-scale trials conducted in the UK, Sweden, USA, and Australia, and reported increases in gas production by up to 50% and improvements of organic matter removal from initially 40% to 70% after the treatment. Also, an improvement of digestate dewaterability was observed at a test at the Orange County Sanitation District (USA), which led to a slight, but significant increase of the TS content of the digestate´s filter cake by 1.6 percentage points [111]. Despite these highly positive results, also negative outcomes were reported and, for instance, Dåverhög and Balmér [107] concluded that US treatment did not lead to measurable improvements of anaerobic digestion performance in two full-scale trails conducted at the Ernemar and Gässlösa WWTPs in Sweden.

While the majority of the above results appears highly promising, it should be noted that some of the studies were not published in peer-reviewed journals and employed somewhat inconsistent test methodologies or provided incomplete experimental descriptions. For instance, the use of a non-sonicated control digester was only conducted rarely, and start-up phases (for monitoring digester performance before the US treatment or for verifying comparable performance between control and test digester) were omitted most of the times. Yet, knowledge on the digester performance before US treatment is of crucial importance to interpret treatment effects. For instance, when methane production in a full-scale digester can be increased by 50%, it seems that the digester performance was highly deficient in the first place so that the result transferability to common, well-performing digesters is very limited. Moreover, experimental descriptions (for instance, regarding specific energy inputs, sludge characteristics, or feeding regimes), which are a key pre-requisite to compare the results among each other, were incomplete or missing completely in most studies. Thus, the scientific value of currently available data on full-scale trials is limited.

Concerning operational aspects, existing full-scale trials confirm the high susceptibility of sonotrode and radial horn reactors towards cavitation erosion and reactor clogging. For instance, Oon et al. [46] experienced frequent clogging of the radial horn reactors used in the previously described full-scale trial at the Ulu Pandan Water Reclamation Plant in Singapore. Thus, manual cleaning and backwash was necessary every second day. Besides the somewhat challenging operation, the lifetime of the horns was found to be quite limited and amounted to less than 2,000 h (or 43 d in permanent operation). The short lifetime of the US-inducing surfaces was confirmed by Neis [45] who stated that the sonotrodes employed at the Bamberg WWTP in Germany required annual replacement. Thus, in addition to the rather troublesome reactor operation, annual maintenance costs of several thousand € seem another hindrance for full-scale US treatment.

For tubular US reactors with surface transducers, no scientifically documented full-scale test is available to date. Yet, three studies investigated the performance of full-scale tube reactors by the use of laboratory BMP tests [29, 43, 76]. Regardless of the lower US intensity of the novel reactor type (< 1 W/cm^2), Appels et al. [29] achieved an enhancement of specific methane yield of a WAS sample by almost 23% already at a low, industrially feasible energy input of 327 kJ/kg$_{TS}$. Koch et al. [43] achieved an increase in methane yield of over 60% when treating DS with a solids content of 2.4%. However, no effect on methane production could be discerned for substrates with higher TS contents such as agricultural sludge (TS = 4.7%) or RS (TS = 3.4%), despite energy inputs as high as 5,000 kJ/kg$_{TS}$. Thus, the scarce existing literature indicates that the low-intensity surface transducer systems are generally capable to successfully pre-treat sewage sludge, while the novel reactor type might be particularly sensitive to higher TS contents. Due to the lack of documented full-scale trials, however, no information on operational aspects is available to date.

CHAPTER 3

Research objectives and hypotheses

Based on the outlined potential of US reactors employing surface transducers, the main research goal of this dissertation was a performance assessment of the novel reactor type and an optimization of treatment conditions. Key research topics were (i) a proof-of-concept of the novel reactor type, (ii) the optimization of operating parameters such as specific energy input, (iii) the examination of the fluid dynamics within the reactor, (iv) an in-depth understanding of the role of the geometric reactor design, (v) the up-scaling and full-scale testing of the newly developed US reactor, and (vi) the exploration of alternative treatment schemes such as DS co-treatment. The key research topics are summarized in Figure 3.1, along with their graphical identifiers and a 3-D schematic of the employed lab-scale flatbed reactor.

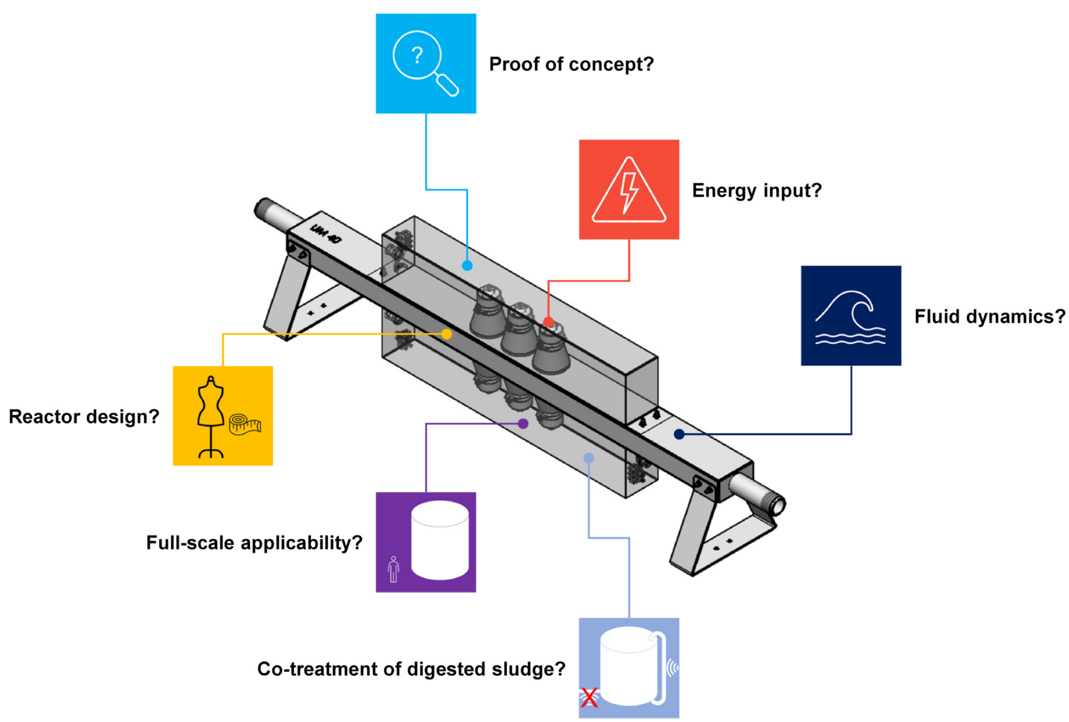

Figure 3.1: 3-D schematic of the novel ultrasonic flatbed reactor and key research topics along with graphical identifiers.

Based on the above research questions, specific research objectives and hypotheses were derived. In the following, all objectives and hypotheses of this dissertation are introduced in detail, along with their respective research significances. A brief overview of the resulting research program is given in Table 3.1.

3.1 Research objective #1

Evaluation of the performance of a novel ultrasonic flatbed reactor for waste activated sludge pre-treatment at industrially feasible specific energy inputs.

In lab- and pilot-scale studies, ultrasonication of sewage sludge is commonly conducted using relatively high specific energy inputs. A frequently reported range of specific energy inputs required to cause notable sludge disintegration and cell lysis is 1,000 kJ/kg_{TS} to 16,000 kJ/kg_{TS} [26, 61], while several studies even applied higher energy inputs, reaching levels of 27,000 kJ/kg_{TS} [36] or even 108,000 kJ/kg_{TS} [80].

However, such levels by far exceed energy inputs feasible at full-scale WWTPs. One of the highest energy inputs reported for a full-scale trial amounted to 1,260 kJ/kg_{TS} at the Monguelfo WWTP in Italy [85]. Typically, much lower energy inputs are attained, ranging from approximately 150 kJ/kg_{TS} (full-scale test at the Bamberg WWTP, Germany, [45]) to approximately 400 kJ/kg_{TS} (full-scale test at the Ulu Pandan Water Reclamation Plant, Singapore, [84]).

Given this critical offset in energy inputs between lab- and full-scale studies, the first research objective was to test whether the novel ultrasonic flatbed reactor is capable of significantly enhancing anaerobic digestion performance at low, industrially feasible energy inputs or not. This proof-of-concept was conducted according to the following hypothesis:

> ***Hypothesis #1:*** *"Ultrasonication of waste activated sludge at low specific energy inputs using a novel ultrasonic flatbed reactor leads to significant enhancement of the sludge´s biomethane potential so that the pre-treatment is energy-positive."*

To test the hypothesis, three different samples of thickened WAS were collected from three different WWTPs to account for the uniqueness of different plants and to obtain results as generally valid as possible. The sampled sludges were subsequently sonicated at seven different specific energy inputs ranging from 200 kJ/kg_{TS} to 3,000 kJ/kg_{TS}. After the treatment, the samples were analyzed for DD_{COD} and tested for their BMP using batch assays.

For an estimation of treatment economics, the energy consumption of the US reactor was balanced against the energy content of the additionally produced methane. The corresponding study is presented in Chapter 4.

3.2 Research objective #2

Evaluation of the impact of fluid dynamics and ultrasonic reactor design on the efficiency of sewage sludge disintegration.

From other realms (such as chemical engineering or industrial wastewater treatment), it is well-known that US reactor design has a major impact on sonication performance [106, 112, 113]. However, for sewage sludge sonication, available literature on the impact of reactor design is scarce and to date, none of the available review papers [4, 10, 12, 19, 26, 61] has addressed the topic at all. A few research papers have dealt with ultrasonic reactor design [56, 78], however, without arriving at a clear relationship between geometric design and disintegration efficiency. At the same time, reactor geometry seems to play a critical role for sludge treatment, due to clogging risk on the one hand (large diameters favorable), and sound wave attenuation on the other hand (small diameters favorable) [43, 114]. To balance these conflicting requirements, a systematic examination of the interplay between reactor geometry and sonication efficiency seems, therefore, crucial.

As full-scale US reactors are typically designed as flow-through reactors, the fluid dynamics in the reaction chamber need to be included in such consideration as well. In a flow regime that is fully-mixed, reactor design might be less important, as all particles experience more or less the same exposure time and the same cavitation intensity. However, in a strictly laminar flow regime, exposure times and treatment intensities might strongly differ among different flow laminae. While it is well-known that US treatment causes pronounced acoustic streaming and thorough mixing in aqueous media [115, 116], it seems unlikely that the same mixing behavior can also be provoked in the highly viscous, non-Newtonian sewage sludge. Thus, to investigate whether the flow in US reactors during sewage sludge pre-treatment remains laminar despite the impact of cavitation-induced micro-turbulences, the following hypothesis was formulated:

> ***Hypothesis #2.1:*** *"Sludge flow in ultrasound flatbed reactors is mostly laminar despite the impact of cavitation-induced micro-turbulences due to the high viscosity of the sludge."*

The hypothesis was tested by visual examination of sludge flow of both WAS and DS in an US reactor with transparent panels. To render the opaque sludge flow accessible for visual inspection, sewage sludge was substituted by two transparent xanthan solutions with similar flow behavior. To visualize flow laminae, dye streams were injected into the solutions, which were pumped through an ultrasound reactor at an industry-typical flow rate. The corresponding study is presented in Chapter 5.

Based on the hereby attained knowledge on the fluid dynamics of sludge flow in US reactors, the relationship between reactor geometry and sludge disintegration was further investigated. Using a size-adjustable reactor setup comprising two opposite plate reactors, reaction chamber heights (RCHs) ranging from 20 mm to 100 mm could be tested. The optimization of the reactor design was conducted in a trifold approach, including cavitation noise measurements with a hydrophone (primary treatment effect), and aluminum foil erosion and COD solubilization analyses (secondary treatment effects). The assumed interplay between reactor geometry and sewage sludge disintegration was examined by the following hypothesis:

> ***Hypothesis #2.2:*** *"Ultrasonic reactor designs with maximum average cavitation noise levels provide both maximum treatment homogeneity and maximum disintegration efficiency."*

The overall disintegration efficiency was assessed by collecting a representative sample of the fully-mixed reactor content of each RCH for the analysis of COD solubilization. The (in)homogeneity of the treatment was assessed by a dense, three-dimensional mapping of cavitation noise, aluminum foil erosion, and COD solubilization throughout the different reaction chamber geometries. The impact of flow on treatment (in)homogeneity was additionally assessed using computational fluid dynamics, based on the knowledge on sludge flow dynamics obtained for Hypothesis #2.1. The corresponding study is presented in Chapter 6.

3.3 Research objective #3

Full-scale assessment of the novel US reactor for waste activated sludge pre-treatment.

Due to the distinct differences between lab- and full-scale environments, a reliable performance assessment of the novel US reactor concept requires a full-scale test at a WWTP. However, for tubular US reactors with surface transducers, no research article on full-scale performance is available to date. To close this research gap, the laboratory flatbed reactor was up-scaled for full-stream WAS pre-treatment based on the knowledge obtained in the previous lab studies and tested at a full-scale WWTP according to the following hypothesis:

> ***Hypothesis #3:*** *"Low energy input sonication of thickened waste activated sludge allows for significantly increased methane production, enhanced organic matter removal, lower sludge viscosity, and improved digestate dewaterability at a full-scale wastewater treatment plant."*

In order to derive results from the full-scale study that are also transferable to other plants, a WWTP with typical (or even challenging) operating conditions (regarding TS content of WAS, HRT of the sludge in the digester, and WAS to RS ratio) was selected according to Frank Sinatra´s 'New-York-Principle' (*"If I can make it there, I´ll make it anywhere."*). For high result reliability, the full-scale test was performed by the use of a non-sonicated control digester and a sonicated test digester after verifying comparable digester performance in a start-up phase. Treatment effects were monitored based on (i) methane production, (ii) organic matter removal, (iii) sludge viscosity, (iv) and digestate dewaterability. For an estimation of treatment economics, potential benefits of the treatment (such as additional methane production, reduced mass of residual biosolids for disposal, lower pumping or stirring costs due to decreased sludge viscosity, or improved dewatering performance) were benchmarked against the electricity and investment costs of the US reactor. In order to test the potentially superior operational performance of tubular US reactors, reactor cleaning and backwash as commonly applied for sonotrode and radial horn reactors was omitted throughout the entire experimental phase. The corresponding study is presented in Chapter 7.

3.4 Research objective #4

Evaluation of the potential of ultrasonic co-treatment (i.e., the sonication of digested sludge during anaerobic digestion) as an alternative to conventional waste activated sludge pre-treatment.

At WWTPs, US is mostly applied as a pre-treatment method for WAS [10, 12, 19]. Yet, co-treatment (i.e., a treatment not before, but during anaerobic digestion) of DS could be a promising alternative particularly for US reactors with lower oscillation amplitude, as DS exhibits a lower, more amenable TS content. Besides, DS is already pre-digested so that the treatment could be efficiently concentrated on the remaining, recalcitrant sludge constituents. Recently, several studies were published to investigate the potential of DS sonication [43, 65, 66, 117]. However, all these studies were conducted using batch tests so that the transferability of their results to continuously operated full-scale digesters is limited [104]. To date, only one study by Azman et al. [64] considered DS sonication in a continuously operated pilot-scale digester. However, as the investigated manure digestate exhibited a high TS content of more than 7%, treatment efficiency might have been strongly compromised by pronounced sound wave attenuation (similar to conventional WAS pre-treatment).

Thus, to assess the performance of DS co-treatment based on a substrate with a more amenable TS content, sonication of a municipal DS with a solids concentration of 3% was studied based on the following hypothesis:

> ***Hypothesis #4:*** *"Ultrasonic co-treatment of digested sludge leads to economic enhancement of methane production and (volatile) solids removal, without negative effects on digestate dewaterability."*

For testing the hypothesis, DS was sonicated in a continuously operated two-part biogas test system, comprising a non-sonicated control digester and a sonicated test digester. To ensure transferability of the results to full-scale plants as much as possible, the test system operation was closely aligned to the conditions at a WWTP regarding HRTs and organic loading rate. Treatment effects were examined based on (i) specific biogas production, (ii) (volatile) solids removal, and (iii) digestate dewaterability, while treatment economics were estimated by balancing energy costs of the US reactor against savings due to additional methane and reduced residual sludge masses for disposal. The corresponding study is presented in Chapter 8.

Table 3.1: Dissertation structure summarizing research objectives, hypotheses, and corresponding publications.

Identifier	Chapter	Research objectives	Hypotheses	Publications
	4	**Research objective #1** Evaluation of the performance of a novel ultrasonic flatbed reactor for waste activated sludge pre-treatment at industrially feasible specific energy inputs.	**Hypothesis #1** "Ultrasonication of waste activated sludge at low specific energy inputs using a novel ultrasonic flatbed reactor leads to significant enhancement of the sludge´s biomethane potential so that the pre-treatment is energy-positive."	**Paper I** Lippert, T., Bandelin, J. Musch, A., Drewes, J.E., Koch, K. (2018), *Bioresource Technology*, 264, 298-305
	5	**Research objective #2** Evaluation of the impact of fluid dynamics and ultrasonic reactor design on the efficiency of sewage sludge disintegration.	**Hypothesis #2.1** "Sludge flow in ultrasound flatbed reactors is mostly laminar despite the impact of cavitation-induced micro-turbulences due to the high viscosity of the sludge."	**Paper II** Lippert, T., Bandelin, J., Schlederer, F., Drewes, J.E., Koch, K. (2019), *Ultrasonics Sonochemistry*, 55, 217-222
	6		**Hypothesis #2.2** "Ultrasonic reactor designs with maximum average cavitation noise levels provide both maximum treatment homogeneity and maximum disintegration efficiency."	**Paper III** Lippert, T., Bandelin, J., Schlederer, F., Drewes, J.E., Koch, K. (2020), *Ultrasonics Sonochemistry*, 68, 105223
	7	**Research objective #3** Full-scale assessment of the novel US reactor for waste activated sludge pre-treatment.	**Hypothesis #3** "Low energy input sonication of thickened waste activated sludge allows for significantly enhanced specific methane production, enhanced organic matter removal, lower sludge viscosity, and improved digestate dewaterability at a full-scale wastewater treatment plant."	**Paper IV** Lippert, T., Bandelin, J. Vogl, D., Alipour Tesieh, Z., Wild, T., Drewes, J.E., Koch, K. (2021), *ACS ES&T Engineering*, 1, 298-309
	8	**Research objective #4** Evaluation of the potential of ultrasonic co-treatment (i.e., the sonication of digested sludge during anaerobic digestion) as an alternative to conventional waste activated sludge pre-treatment.	**Hypothesis #4** "Ultrasonic co-treatment of digested sludge leads to economic enhancement of methane production and (volatile) solids removal, without negative effects on digestate dewaterability."	**Paper V** Lippert, T., Bandelin, J., Xu, Y., Chen Liu, Y., Hernández Robles, G., Drewes, J.E., Koch, K. (2020), *Renewable Energy*, 166, 56-65

CHAPTER 4

Energy-positive sewage sludge pre-treatment with a novel ultrasonic flatbed reactor at low energy input

This chapter has been previously published with editorial changes as follows: Lippert, T., Bandelin, J., Musch, A., Drewes, J.E., Koch, K. (2018). Energy-positive sewage sludge pre-treatment with a novel ultrasonic flatbed reactor at low energy input. Bioresource Technology 264, 298-305. DOI: 10.1016/j.biortech.2018.05.073

Author contributions: Thomas Lippert was responsible for the research plan, data analysis, and manuscript preparation. Jochen Bandelin planned and designed the ultrasound reactor. The experiments were conducted by Alexandra Musch and Thomas Lippert. Jörg E. Drewes and Konrad Koch supervised the study. The manuscript was reviewed by Jochen Bandelin, Jörg E. Drewes, and Konrad Koch.

Abstract

The performance of a novel ultrasonic flatbed reactor for sewage sludge pre-treatment was assessed for three different waste activated sludges. The study systematically investigated the impact of specific energy input (200 – 3,000 kJ/kg_{TS}) on the degree of disintegration (DD_{COD}, i.e., the ratio between ultrasonically and maximum chemically solubilized COD) and methane

production enhancement. Relationship between DD_{COD} and energy input was linear, for all sludges tested. Methane yields were significantly increased for both low (200 kJ/kg_{TS}) and high (2,000 – 3,000 kJ/kg_{TS}) energy inputs, while intermediate inputs (400 – 1,000 kJ/kg_{TS}) showed no significant improvement. High inputs additionally accelerated reaction kinetics, but were limited to similar gains as low inputs (max. 12%), despite the considerably higher DD_{COD} values. Energy balance was only positive for 200 kJ/kg_{TS}-treatments, with a maximum energy recovery of 122%. Results suggest that floc deagglomeration rather than cell lysis (DD_{COD}=1% – 5% at 200 kJ/kg_{TS}) is the key principle of energy-positive sludge sonication.

4.1 Introduction

During conventional wastewater treatment, large volumes of sewage sludge accrue as an inevitable byproduct of the mechanical and biological treatment steps. In the European Union, about 10 million tons of sludge (dry weight) is produced each year [2], whereas production is projected to increase to 13 million tons per year by 2020 [118, 119]. Also worldwide, sludge production is expected to rise, due to more stringent legislation and the increase of wastewater treatment in developing countries [19].

This sludge is a severe environmental concern due to its potential pathogen load, its biological activity, the affiliated odor nuisance, and the large volumes. However, the sludge also represents a valuable and energy-rich resource due to its biomethane potential (BMP), which allows to recover a substantial part of the energy expenses of the overall wastewater treatment process [8, 120].

Anaerobic digestion is the most commonly applied process in wastewater treatment to utilize this energy potential by converting the organic fraction of sludge into biogas while stabilizing and reducing the accumulated sludge volumes [14, 114]. However, anaerobic digestion is subjected to several impediments. While primary sludge, originating from primary sedimentation, is readily biodegradable, waste activated sludge (WAS) from the secondary biological treatment step, mostly comprising aerobic bacteria with intact cell walls, is characterized by a lower degradability [12, 36]. Hence, long hydraulic retention times in the digesters are necessary, which require large reactor volumes that are expensive to build and maintain. In spite of the long retention times, the degradation of the sewage sludges is still incomplete, which leaves relatively large volumes of digestate. The subsequent dewatering, drying, and disposing of these residue can account for up to 50% of the operating costs of wastewater treatment plants (WWTPs) [11].

The limited biodegradability of WAS gave rise to several pre-treatment methods including mechanical, thermal, chemical, and biological processes, from which ultrasonication was found to be among the most effective technologies [10, 19]. By inducing ultrasound (US) waves into the sludge, this pre-treatment method utilizes the phenomenon of acoustic cavitation. In successive compression and rarefaction cycles, gas bubbles are formed and expanded until they reach a critical size to finally collapse in cavitation events. These extremely violent bubble implosions generate powerful jets of liquid with local temperatures of 5,000 °C and pressures of 500 bars. The resulting hydromechanical shear forces deagglomerate sludge flocs, disrupt cell walls, and release easily degradable intercellular matter and active enzymes [12]. The process is subdivided into the two stages of sonodispergation and sonolysis [56]. While the former refers to a break-up of the floc agglomerates below a certain energy input threshold of approximately 1,000 kJ per kg of total solids (TS) [81], the latter connotes the lysis of cell walls. The rupture of the cells supports the hydrolysis step, which is commonly considered the rate-limiting step of anaerobic digestion of sewage sludge [121, 122].

In spite of the merits of the technology in terms of enhanced biogas production and reduced residual sludge volumes observed in various lab-scale studies [27, 70, 123], US technologies face several obstacles in full-scale applications. The most commonly applied sonotrode reactors, in which US is directly induced into the substrate by horn-shaped sonotrodes, is characterized by strong US intensity, but also by a fast erosion of the sonotrode tips and a considerable risk of clogging due to the interruption of the flow path [31, 44]. The economic viability of this reactor design is, thus, limited by high maintenance costs and frequent service cycles. Hence, tubular systems with US transducers mounted externally on the reactor walls could be an advantageous alternative. This design features an unobstructed pipe flow so that the clogging risk is minimized and exhibits a reduced risk of erosion, as US is transmitted over a much larger area (entire reactor wall) at lower oscillation amplitudes (6 – 8 μm wall oscillation vs. 60 – 80 μm sonotrode tip oscillation). Several researchers investigated such reactor configurations and, for instance, Appels et al. [29] employed a tube-shaped reactor for WAS treatment to increase methane yields by about 20% at a specific energy input as low as 327 kJ/kg_{TS}. Koch et al. [124] reported similar results when treating digested sludge (DS) with a TS of 2.4% using a tube reactor gaining up to 60% additional methane. However, when substrates with TS contents $> 3\%$ were sonicated, increases in gas production were no longer observed [29, 124]. While sludge characteristics other than TS might influence sonication amenability as well, efficiency of tubular designs seems limited by sound field attenuation in substrates with a high TS content, due to the reduced US intensity of the lower oscillation amplitudes [124].

Within the present study, a performance assessment of a novel US flatbed reactor design is presented that claims to combine the high performance of sonotrode-based systems with the low-maintenance operation of tubular reactors. The new flatbed design features a reaction chamber with an unobstructed rectangular cross-section, to reach a high ratio between oscillating surfaces and sonicated volume. To minimize critical losses due to sound field attenuation, the distance between the reactor's transducer plates was previously optimized for WAS treatment by the use of a hydrophone [125].

The objective of the present study was to assess the capability of the new reactor type to treat substrates with a high TS content at energy inputs low enough to be feasible in full-scale applications at WWTPs. For the first time, a range of seven different realistic energy inputs (200 – 3,000 kJ/kg_{TS}) was systematically investigated for a total of three different thickened WAS samples with TS contents ranging from 4.4% to 6.7%. The performance of the novel reactor was benchmarked against (i) chemical oxygen demand (COD) solubilization and degree of disintegration (DD_{COD}), (ii) increase in specific methane production (SMP) and specific methane yield (SMY), and (iii) the energy self-sufficiency of the treatment, expressed as the ratio of invested vs. recovered energy.

4.2 Materials and methods

4.2.1 Origin of WAS and DS

WAS and DS (used as inoculum) were collected at the Starnberg, Traunstein, and Munich East WWTPs (all located within the metropolitan area of Munich, Germany). In all WWTPs, sampling of thickened WAS was conducted after the belt filter, while DS was sampled from the recirculation loop of the anaerobic digester. Characteristics of the investigated sludge types and operational parameters of the digesters are summarized in Table 4.1.

4.2.2 Ultrasonication unit and experimental setup

A novel ultrasonic flatbed test reactor (BANDELIN electronic GmbH & Co. KG, Berlin, Germany) was applied for WAS treatment. The lab-scale reactor features a reaction chamber (length = 1,000 mm) with a rectangular cross-section (width = 80 mm, height = 40 mm) and six ultrasonic transducers mounted externally at the top and the bottom wall of the chamber (3 transducers each), as depicted in Figure 4.1. The system was operated at a frequency of 25 kHz with an electric power of 300 W. For the sonication experiments, the reactor was arranged in a recirculation loop setup, comprising the US reactor, an eccentric screw pump (Erich NETSCH GmbH & Co. Holding KG, Selb, Germany), and connecting tubes.

The total volume of the experimental system (i.e., the tallied volumes of reactor, pump, and tubes) was determined to be 5.2 ± 0.05 L. To ensure a complete filling up to the defined volume

Table 4.1: Average characteristics of investigated sludges types, operational parameters of anaerobic digesters and treatment capacities of the WWTPs.

	Starnberg WWTP		**Traunstein WWTP**		**Munich East WWTP**	
	WAS	**DS**	**WAS**	**DS**	**WAS**	**DS**
TS [% of FM]	6.7	2.8	4.4	2.1	5.3	2.5
VS [% of TS]	76.6	65.5	71.7	60.1	73.4	65.2
COD [mg/L]	69,550	31,704	42,815	16,341	53,779	27,710
Soluble COD [mg/L]	859	337	580	322	194	443
HRT [d]	25		30		37	
Digester temperature [°C]	37		38		38	
WWTP capacity [PE]	58,000		68,000		75,000	

TS = Total solids, FM = Fresh matter, VS = Volatile solids, COD = Chemical oxygen demand

HRT = Hydraulic retention time, PE = Population equivalents

without e.g., air bubble entrapments, WAS was injected into the system from a pressurized container (∼3 bar). After filling, the sludge was circulated through the system with an activated US reactor at a flow rate of 300 L/h. Due to the recirculation within the loop system, the sludge undergoes several reactors passages, thus simulating a treatment in series. Specific energy input was regulated by treatment duration and calculated according to:

$$SE = \frac{P \cdot t}{TS \cdot V \cdot \rho} \tag{4.1}$$

where SE is the specific energy [kJ/kg_{TS}], P is the electric power of the applied US system [kW], t is the treatment duration [s], TS is the total solids content [kg_{TS}/kg_{FM}], V is the tallied volume [L] and ρ is the density [kg_{FM}/L]. Density of the sludges was determined using a pycnometer and was found to be 1.00 ± 0.01 kg_{FM}/L for all WAS samples.

Specific energy inputs were chosen low enough to simulate treatment intensities feasible in full-scale WWTPs ranging from 200 kJ/kg_{TS} to 3,000 kJ/kg_{TS}, based on conditions at the investigated WWTPs and in accordance to [76]. After completion of the US treatments, the recirculation system was emptied and the content was mixed to yield samples for chemical analyses and BMP assays. Impact of thermal disintegration was deemed to be negligible, as even treatments at 3,000 kJ/kg_{TS} increased the sludge temperature by only about 2 K above the incubation temperature of 38 °C.

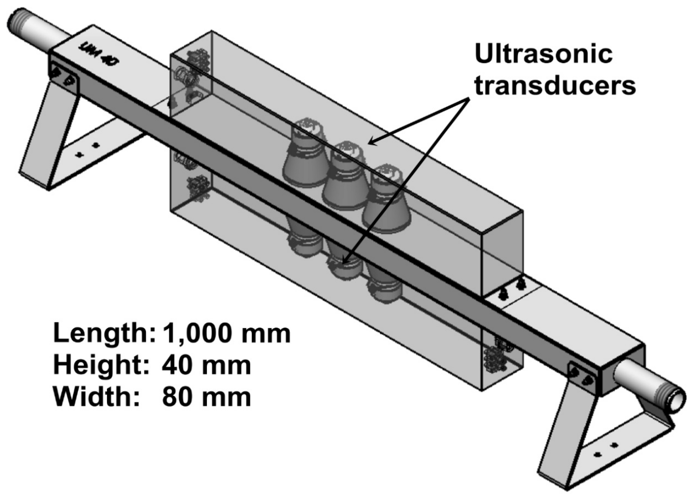

Figure 4.1: Schematic of the novel ultrasonic flatbed reactor.

4.2.3 Analytical methods

4.2.3.1 Sludge analysis

Determination of COD, soluble COD (sCOD), TS, and volatile solids (VS) was carried out according to standard methods [126]. The disintegration efficiency of US treatment was evaluated by analyzing sCOD and DD_{COD}, whereas the latter expresses the ratio between ultrasonically and maximum chemically solubilized COD:

$$DD_{COD} = \frac{sCOD_{US} - sCOD_0}{sCOD_{NaOH} - sCOD_0} \tag{4.2}$$

where $sCOD_{US}$ is the concentration of sCOD after sonication, $sCOD_0$ is the initial concentration of sCOD of the untreated sludge sample, and COD_{NaOH} is the sCOD concentration of the sludge sample after alkaline hydrolysis (all [mg/L]). For the chemical solubilization, 1M NaOH solution was mixed with WAS in a ratio of 2:1 at room temperature for 24 h as suggested by Bougrier et al. [81].

4.2.3.2 BMP tests

The BMP tests were conducted with two identical Automatic Methane Potential Test Systems (AMPTS II; Bioprocess Control Sweden AB), operated in parallel. A detailed description of the system and the test procedure can be found elsewhere [43]. Due to the presence of coarse particles, the inoculum was sieved through a 1 mm mesh prior to incubation. The ratio of inoculum to substrate (i.e., the ratio between DS and WAS) was chosen to be 2:1 based on VS content [127, 128] and digestion temperature was set to 38 ± 0.5 °C. Prior to incubation, the headspaces of all bottles were flushed with a synthetic gas mixture (65% N_2 and 35% CO_2) to establish anaerobic conditions with a CO_2 concentration similar to full-scale digester headspaces, as suggested by Koch et al. [129]. Each sample was tested in triplicate and all tests were carried out until the daily gas production during three consecutive days was < 1% of the total gas production [127]. The results of the BMP tests were validated according to the validation criteria of Holliger et al. [127], including that relative standard deviation within each triplicate should be below 5%. Endogenous gas production and fitness of all inocula were evaluated using blank and positive control samples (with microcrystalline cellulose as substrate), respectively.

4.2.3.3 Statistical testing

For statistical analysis, obtained data sets were compared using a Student's t test, whereas differences between two sets were denoted as significant for $p < 0.05$ and as highly significant for $p < 0.01$. The calculations were cross-checked with an online tool, available at https://www.graphpad.com/quickcalcs/ttest1.cfm.

4.3 Results and discussion

Three different thickened WAS samples with TS contents ranging from 4.4% to 6.7% were treated with a novel US flatbed reactor at seven different levels of specific energy input (200 – 3,000 kJ/kg_{TS}). The efficiency of sonication was evaluated with respect to (i) increase in sCOD and DD_{COD}, (ii) impact on SMP and gains in SMY, and (iii) energetic self-sufficiency. The objective of the study was to assess the performance of the novel US reactor in terms of SMP enhancement for substrates with high TS contents and to identify levels of specific energy input that allow for an energy-positive operation of the new reactor type at a full-scale WWTP.

4.3.1 Impact of specific energy input on COD solubilization

The relationship between specific energy input and COD solubilization for the three tested sludges is displayed in Figure 4.2. In all cases, the relation is close to positive linearity, which is in line with the results of previous studies [76, 82, 124]. Sonication resulted in a strong increase of sCOD already at very low specific energy inputs (e.g., 170% at 200 kJ/kg_{TS} for $WAS_{Traunstein}$), while reaching up to 1,400% at inputs of 3,000 kJ/kg_{TS} ($WAS_{Munich\ East}$). However, DD_{COD} remains relatively low to moderate for all sludges with values ranging from 1% to 22%, which

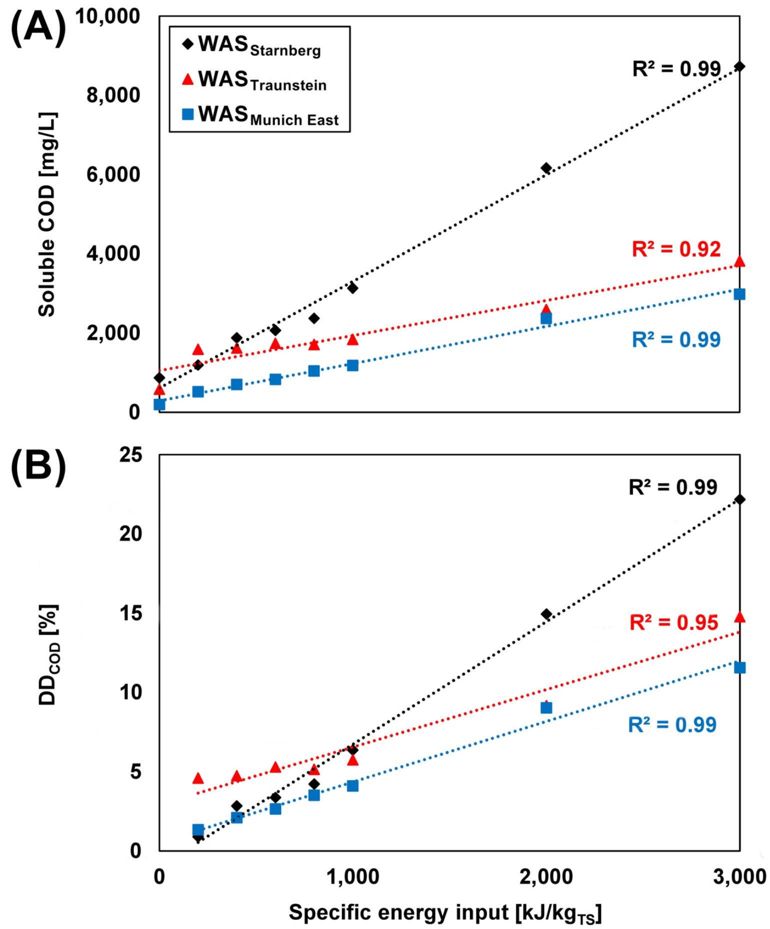

Figure 4.2: Solubilization of COD (A) and degree of disintegration DD_{COD} (B) vs. specific energy input.

is in the range of literature data referring to similar levels of specific energy input [81–83]. Interestingly, there seems to be no clear relationship between solubilization amenability and solids content. $WAS_{Starnberg}$, having the highest TS content (6.7%) and, thus, being possibly subjected to the highest sound wave attenuation, even showed the steepest increase of sCOD and DD_{COD} when increasing energy input.

When comparing the disintegration efficiency to a previous study with tube reactors, the flatbed reactor seems to clearly outperform the system with the circular cross-section. Koch et al. [124] reported that COD solubilization was substantially less pronounced, even for substrates with lower TS contents. For agricultural sludge (TS of 4.7%) and raw sludge (TS of 3.4%), maximum sCOD releases of 36% and 193% were observed, respectively, despite energy inputs as high as 10,000 kJ/kg_{TS} and 3,500 kJ/kg_{TS}. However, a direct comparison is tricky, as sludge characteristics other than TS content may have affected sonication amenability, too.

It should be noted that the experimental setup might slightly damp the impact of sound field attenuation due to the recirculation of the sludge. When considering a single floc that passes through the channel center (therefore potentially experiencing an attenuated sound field due to the maximum distance to the transducer plates), it will most likely travel along another streamline with a different distance to the transducers in the next cycle, possibly with a stronger sound field. Therefore, disintegrating effects are assumed to be distributed relatively homogenously over the treated sludge volumes. Still, the differences between a single stage treatment (as e.g., in full-scale applications) and the simulated serial treatment are considered to be levelled out when analyzing only the averaging sum parameters of sCOD and DD_{COD}. The experimental setup is therefore considered capable to deliver a reliable estimate on the reactor's solubilization efficiency even in full-scale installations.

4.3.2 Impact of specific energy input on SMY and production kinetics

The results of the conducted BMP assays show that US treatment was able to increase SMY values for all of the three tested sludges, at most tested energy inputs. The final SMYs for all tested samples and specific energy inputs, as well as the incremental increase of SMY relative to the untreated sample are depicted in Figure 4.3.

The maximum average increases in SMY were found to be 6.9% for $WAS_{Starnberg}$ and 11.7% for both $WAS_{Traunstein}$ and $WAS_{Munich\ East}$, as illustrated in Figure 4.3B. Interestingly, the maximum values for $WAS_{Starnberg}$ and $WAS_{Traunstein}$ were not observed at the highest (3,000 kJ/kg_{TS}), but at the lowest specific energy input (200 kJ/kg_{TS}). At the highest energy expenditure, improvements of only 2.0% and 7.5% were achieved, respectively. For $WAS_{Munich\ East}$ on the contrary, the 3,000 kJ/kg_{TS}-treatment led to the highest gain observed (11.7%). However, an only slightly inferior gain of 8.6% was realized at the comparably low energy input of 600 kJ/kg_{TS}. In three cases ($WAS_{Traunstein}$ treated with energy inputs of 600 kJ/kg_{TS} and 1,000 kJ/kg_{TS} and $WAS_{Munich\ East}$ treated with an energy input of 400 kJ/kg_{TS}), treatments did not improve methane production, and SMYs slightly dropped below the level of the untreated sample. The observed differences underline the necessity to test more than one sludge to derive generally valid conclusions.

In contrast to COD solubilization, there is no linear correlation between specific energy input and SMY. Instead, it seems that the strongest enhancements of SMYs were found either at the lower or the higher range of energy inputs tested. In addition, $WAS_{Starnberg}$, showing the highest affinity to COD solubilization (Figure 4.2), exhibits the lowest incremental increases in SMY. The results therefore challenge the suitability of the parameters sCOD release and DD_{COD} to predict SMYs, which is in line with observations from Kim et al. [87] and Koch et al. [124].

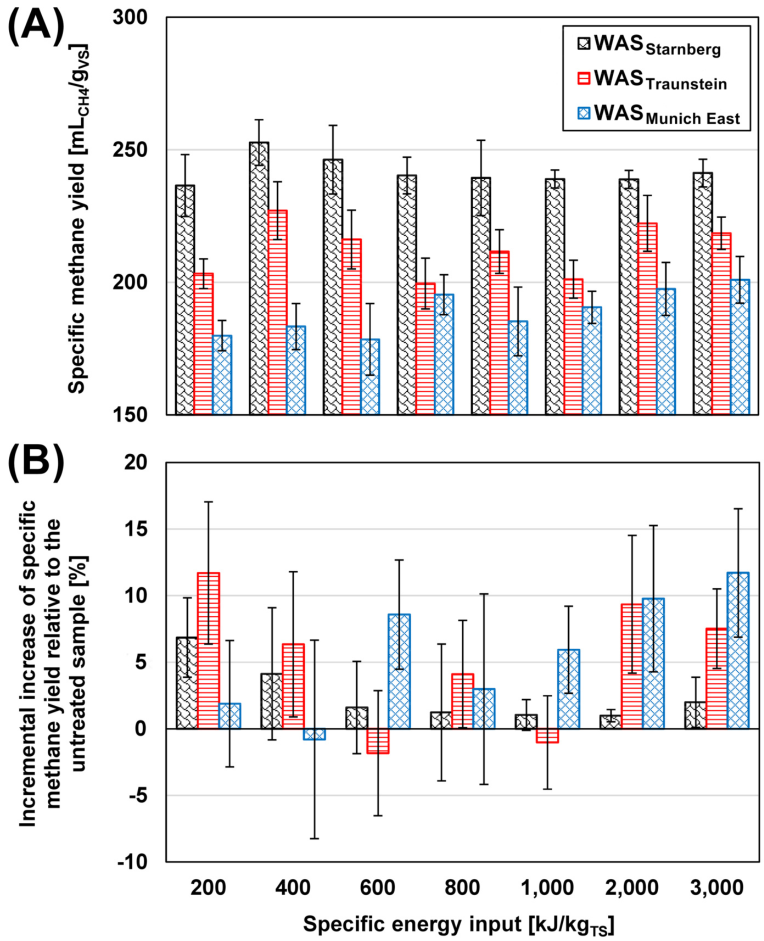

Figure 4.3: Average specific methane yield (A) and average incremental increase of specific methane yield relative to the untreated sample (B) vs. specific energy input. Error bars indicate standard deviation from the average of each triplicate. In five cases, relative standard deviations within triplicates were slightly above the suggested quality criterion of 5%: 5.9% for $\mathrm{WAS_{Starnberg}}$ (untreated), 5.1% for $\mathrm{WAS_{Starnberg}}$ (800 $\mathrm{kJ/kg_{TS}}$), 5.1% for $\mathrm{WAS_{Traunstein}}$ (400 $\mathrm{kJ/kg_{TS}}$), 7.5% for $\mathrm{WAS_{Munich\ East}}$ (400 $\mathrm{kJ/kg_{TS}}$) and 6.9% for $\mathrm{WAS_{Munich\ East}}$ (800 $\mathrm{kJ/kg_{TS}}$).

The interrelation between specific energy inputs and SMY values becomes more traceable when considering the temporal development of methane production. Figure 4.4 displays the cumulative SMP (left side) and the incremental increase in SMP relative to the untreated sample (right side) for all tested WAS samples and energy inputs. Two different patterns in methane production were identifiable, which were reproducible throughout all conducted experiments. For the high energy inputs (2,000 $\mathrm{kJ/kg_{TS}}$ and 3,000 $\mathrm{kJ/kg_{TS}}$), production kinetics were strongly enhanced within the first 72 h of incubation, leading to very pronounced incremental increases in SMP (e.g., up to a maximum of 25% more methane at t ∼20 h for $\mathrm{WAS_{Munich\ East}}$

at 3,000 kJ/kg$_{TS}$). However, after reaching this first peak, the relative increases in SMP diminished with advancing incubation time. In the case of WAS$_{Starnberg}$, this drop was so pronounced that the final incremental increases in SMY receded to 1 - 2% only. For both other sludges, relative increases dropped as well, but stabilized at elevated levels between 7.5% and 11.7%.

For the lower energy inputs (200 – 600 kJ/kg$_{TS}$) on the other hand, SMPs were more sustainably enhanced and relative increases either remained at a constantly elevated level (e.g., for WAS$_{Starnberg}$), or increased over the course of the incubation (WAS$_{Traunstein}$ and WAS$_{Munich\ East}$). Owing to this trend, low energy input treatments were able to keep up with or to even outperform the high energy input treatments in terms of SMY enhancement, as observed for WAS$_{Starnberg}$ and WAS$_{Traunstein}$.

The SMP curves of intermediate energy inputs (800 kJ/kg$_{TS}$ and 1,000 kJ/kg$_{TS}$) were closely aligned to the SMPs of the untreated sample in most cases and exhibited neither a boost in methanogenic activity in the beginning, nor a pronounced increase of SMP over time. The initially elevated reaction kinetics that were perceived for the high energy inputs might be explained by the comparably high release of sCOD (DD$_{COD}$ ranging from 9% to 22%), which serves as a readily available substrate for the anaerobic microorganisms. The subsequent drop of methanogenic activity is not surprising, but a logical consequence of elevated production kinetics when assuming a more or less equal final yield. However, this does not explain why incremental increases fall behind the increases generated by the low energy inputs. A possible explanation for the inferior performance could be re-flocculation, as a side effect of cell lysis. Released intercellular substances were reported to act as biopolymeric flocculants [4] and accordingly, several authors observed an augmentation of larger particles and a gradual increase of mean particle diameters above certain energy inputs [4, 81, 130]. As a consequence, floc surface area and biological accessibility can be reduced, which leads to decelerated reaction kinetics and a lower biogas yield from the particulate matter [131]. This compares well with the relatively flat SMP curves observed for the high energy input treatments after depletion of the initial sCOD. For the intermediate energy inputs (800 kJ/kg$_{TS}$ and 1,000 kJ/kg$_{TS}$), marking the onset of cell lysis, re-flocculation might have already taken place as well, thus negating potentially beneficial effects of floc deagglomeration.

The good performance of the low energy input treatments is surprising at first glance as various studies found that higher specific energy inputs lead to higher additional gains in methane yields [25, 77, 81, 95]. Furthermore, energy inputs below approximately 1,000 kJ/kg$_{TS}$ were reported not to facilitate cell lysis to a larger extent, but to be mostly restricted to sludge floc deagglomeration [25, 72, 81]). The latter is considered consistent

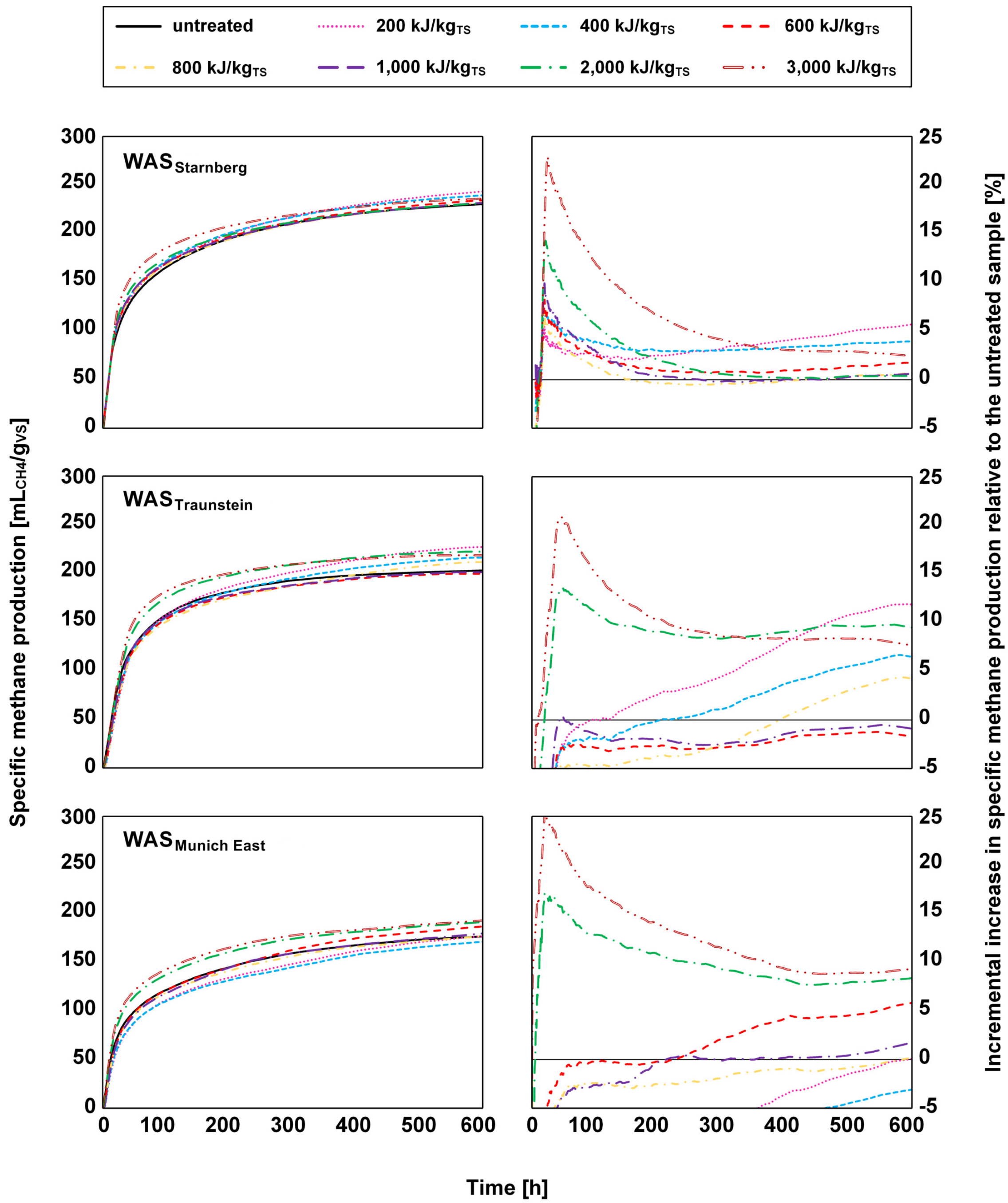

Figure 4.4: Average specific methane production (left) and average incremental increase in specific methane production relative to the untreated sample (right) at different specific energy inputs.

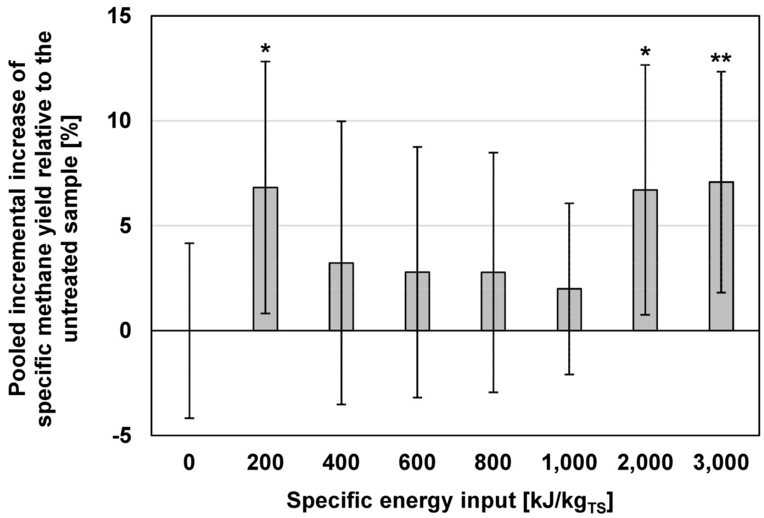

Figure 4.5: Pooled incremental increase of specific methane yield relative to the untreated sample vs. specific energy input. Error bars indicate standard deviation from the average of the nine values. Significant differences to the untreated sample are indicated with an asterisk at a 5% significance level and with two asterisks at a 1% significance level, according to the Student's *t* test.

with the observations in this study, as DD_{COD} values remain fairly low (1% – 5%) for the low energy input treatments. The observations suggests that floc deagglomeration rather than cell lysis was the key driver for the observed gains in SMY for the low energy input treatments. The hypothesis is supported by findings of Cella et al. [72], who stated that deagglomeration can enhance AD, even at minimal degrees of cell lysis. The positive effect of deagglomeration was also acknowledged by Tiehm et al. [25] and was reasoned by the detachment of single bacteria from their protective floc agglomerates, or in other words, by an increase in surface area.

The results regarding the potential of low energy inputs are in line with observations of other authors investigating the impact of specific energy input. Appels et al. [29] realized gains of 20% additional methane with an energy input of only 327 kJ/kg$_{TS}$, while inputs of 818 kJ/kg$_{TS}$ and 1,967 kJ/kg$_{TS}$ led to a lower or only slightly enhanced SMY, respectively. Apul and Sanin [70] found the highest increase in SMY after sonication for 15 min, but after 20 min, the final yield in methane was comparable to the untreated sample again.

In order to prove if more general conclusions can be drawn from the range of samples and energy inputs tested, normalized SMY values (Figure 4.3 right) of all three tested sludges were pooled into one data set, as illustrated in Figure 4.5. The eight data sets (one untreated sample, seven samples treated with different energy inputs) with nine values each were subsequently analyzed for significant differences by means of a Student's *t* test.

Results of the statistical analysis suggest that overall only the lowest energy input of 200 kJ/kg_{TS} and the two highest (2,000 kJ/kg_{TS} and 3,000 kJ/kg_{TS}) energy inputs were able to significantly increase SMYs ($p < 0.05$), whereas the 3,000 kJ/kg_{TS}-treatment even showed a highly significant difference to the untreated sample ($p < 0.01$). All other levels of energy input showed enhancements of SMY by trend, which were however not significant. Within these inputs, relative incremental increases in SMY decreased with increasing specific energy input, to reach a minimum at 1,000 kJ/kg_{TS}.

Among the different specific energy inputs, no significant difference could be discerned, except for the 3,000 kJ/kg_{TS}-treatment, which showed a significantly higher performance than the 1,000 kJ/kg_{TS}-treatment.

While the sample size might still be too small to derive general conclusions, the obtained results further indicate that there is a critical transition between the two working principles of sewage sludge sonication. Low energy inputs cause floc deagglomeration, which was shown to considerably enhance methane yields. At intermediate levels of energy input, positive effects of floc deagglomeration seem to be negated, possibly due to re-flocculating effects. At this stage, cell lysis is not pronounced enough to shift gas production to the solubilized phase, so that SMY enhancement is generally poor. At higher energy inputs and levels of DD_{COD}, methanogenic activity is boosted initially and SMY values can be significantly increased. However, possibly due to reflocculation, SMY enhancement can still drop to only intermediate levels as for $WAS_{Starnberg}$. Conclusions drawn from the statistical analysis should however not be generalized and individual testing is still highly recommended when a suitable energy input is to be determined for a given substrate. Further research should therefore address the relationship between sludge characteristics and sonication amenability in more detail.

Regardless of the impact of specific energy input, the general enhancement of methane production of WAS with TS contents as high as 6.7% using the novel flatbed reactor is considered a key finding of the present study. In previous studies, Koch et al. [124] and Appels et al. [29] investigated the performance of reactors conceptually similar to the employed flatbed reactor. Koch et al. [124] employed a tube reactor with an inner pipe diameter of 80 mm, whereas Appels et al. [29] used a rectangular reaction chamber similar to this study, but with a distance between transducer plates of only 16 mm. In both studies, SMYs could only be improved for substrates with low TS contents, i.e., for WAS with a TS content of 2.1% [29] and for DS with a content TS of 2.4% [124], respectively. For thicker WAS samples, no significant methane productivity enhancement was observed, neither for raw sludge (mixture of WAS and primary sludge with a TS of 3.4%) nor for agricultural sludge (TS of 4.7%).

In the case of the tube reactor, a comparably large diameter and a low ratio between oscillating surface area and sonicated volume due to the circular cross-section might have led to a high impact of sound field attenuation at elevated TS contents. In a recent study, Bandelin et al. [125] analyzed the cavitation intensity within US reactors by means of a hydrophone, thus measuring the sound emission of collapsing cavitation bubble as a surrogate parameter for US intensity. It has been shown that cavitation noise levels approached zero by increasing the distance between two transducer plates to 60 mm and 80 mm, respectively, when treating WAS. Correspondingly, cavitation noise levels were found to increase almost linearly when decreasing transducer plate distance. Results from Bandelin et al. [125] therefore insinuate that a large fraction of the sludge treated with the 80 mm tube reactor in Koch et al. [124] might have experienced no or only a weak treatment by ultrasonication. Furthermore, flow velocity and hence volume flow is largest in the tube center under laminar flow conditions, meaning that the major fraction of the substrate passes through the weakened sound field.

On the contrary, the performance of the 16 mm reaction chamber was probably not affected by sound wave attenuation to a larger extent, as indicated by Bandelin et al. [125]. However, reducing the distance between transducer plates comes at the price of a shorter treatment duration within the US reactor for a given flow rate, possibly leading to hydraulic retention times too short to allow for thorough sonication. The 40 mm design found to be most suitable by Bandelin et al. [125] and investigated in the present study might have effectively balanced treatment duration and cavitation intensity for the given sludge types.

4.3.3 Energy balance of ultrasonication

To estimate the economics of the novel reactor type, the energy invested during US treatment was balanced against the energy payback due to additional methane. For the calculations, an energy content of 10 kWh per m^3 of methane and an energy efficiency of 40% of the WWTPs' combined heat and power plant was assumed. Neither investment and maintenance costs for the ultrasonic system have been considered in this simplified assessment.

The energy payback of the novel reactor for the three sludge types and the seven energy inputs is illustrated in Figure 4.6, with 100% meaning that the amount of invested and recovered energy is equal. The diagram depicts that a positive energy balance could only be achieved for $WAS_{Traunstein}$, at a specific energy input of 200 kJ/kg_{TS}, with an energy recovery of 122%. For $WAS_{Starnberg}$ and $WAS_{Munich\ East}$, maximum energy paybacks were 89% and 27%, respectively, indicating an energetically unfavorable treatment. The considerate differences between energy input levels, but also between the three tested WAS samples further demonstrate that tests on several sludges and energy inputs are inevitable to derive generally valid estimates on treatment economics. For all treatment intensities above 200 kJ/kg_{TS}, more

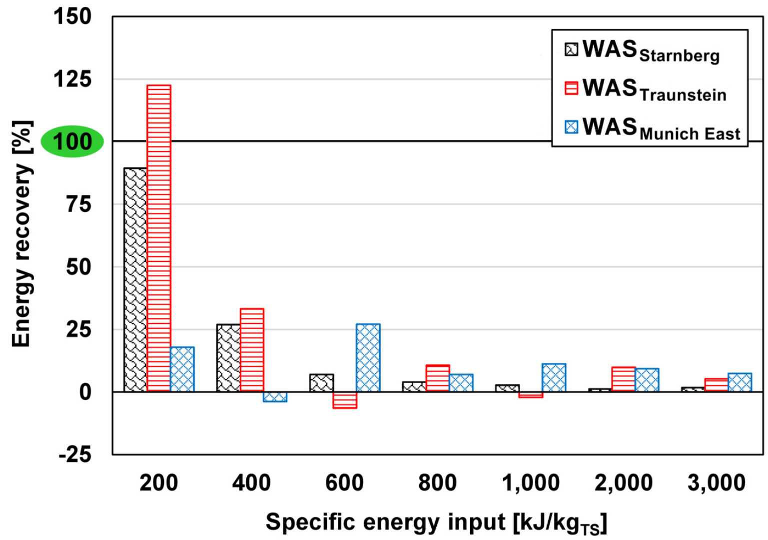

Figure 4.6: Energy recovery, i.e., ratio of electrically invested energy and regained energy by additional methane vs. specific energy input. 100% denote an energy- neutral operation.

energy was invested than additionally recovered, for all sludges tested. When considering that cell lysis was reported to be rather negligible below a specific energy input of 1,000 kJ/kg$_{TS}$, it is suggested that the effects of floc deagglomeration might be more relevant to full-scale applications than the effects of sonolysis requiring higher specific energy inputs. For the high energy treatments to be energy-neutral, theoretical gains in SMY of 53%, 106%, and 158% would have been required for specific energy inputs of 1,000 kJ/kg$_{TS}$, 2,000 kJ/kg$_{TS}$, and 3,000 kJ/kg$_{TS}$, respectively, as exemplified for WAS$_{\text{Munich East}}$.

However, it has to be considered that potential effects on (i) altered sludge dewaterability, (ii) reduced costs for sludge disposal due to enhanced VS removal, (iii) foam control in the digester, and (iv) a lower energy consumption for mixing due to improved sludge viscosity are not reflected in this economic estimation. Moreover, the observed enhancement of production kinetics at high specific energy inputs could be of great interest for WWTPs regardless of the negative energy balance, in case the construction of an additional digester could be avoided. A conclusive economic assessment is, hence, highly case-specific and requires a holistic assessment in a full-scale test.

4.3.4 Concluding remarks

Based on the obtained results, the novel flatbed reactor seems to be a promising follow-up design to existing tubular reactors, by combining the capability of treating highly viscous substrates with a low-maintenance operation. Particularly at low levels of specific energy input, the technology appeared promising with respect to an economically viable full-scale application with energy-neutral operation. Subsequent studies should therefore further elucidate the potential of very low energy inputs, targeting rather floc deagglomeration than cell lysis. However, as observed incremental increases were shown to be only moderate (max. 12%), further studies on the improvement of the reactor design are suggested. To develop tailored reactors for a given sludge type, the interplay between substrate characteristics, fluid dynamics, and sound wave propagation needs to be better understood in the future. For a better estimate of the performance of the novel reactor in full-scale installations, batch tests should be complemented with tests in continuous operation, and full-scale tests at selected WWTPs. Also, it should be evaluated if the expected improvement in erosion and clogging behavior holds true at full-scale operation.

The development of an efficient US reactor applicable in full-scale applications is seen as both a relevant and motivating research target, as the pre-treatment technology might be another important building block for unleashing the potential of the globally growing volume of sewage sludge towards energy-neutral wastewater treatment.

4.4 Conclusions

The investigated ultrasonic flatbed reactor demonstrated to have considerate potential for pre-treating WAS with high TS contents. Already at low energy inputs and DD_{COD} levels, ultrasonication was shown to significantly enhance methane yields, at promising energy balances. Performance of higher energy inputs seemed impaired, possibly due to re-flocculation at the onset of cell lysis, and entailed negative energy balances. Therefore, floc deagglomeration rather than sonolysis seems to be key principle of economic WAS sonication. Future research should validate the promising results in continuous and full-scale tests, also considering potential impacts on VS reduction, sludge dewaterability, digester foaming, and viscosity reduction.

4.5 Acknowledgments

The authors gratefully acknowledge the funding of this study by the Federal Ministry for Economic Affairs and Energy (BMWi), Grant 03ET1396B. The company BANDELIN electronic GmbH & Co. KG is kindly acknowledged for providing the flatbed reactor. Further thanks go to the staff members of the WWTPs of Starnberg, Traunstein, and Munich East and to Nicole Zollbrecht, Myriam Reif, Wolfgang Schröder, and Hubert Moosrainer for their excellent assistance.

4.6 Appendix - Supplementary material

The graphical abstract to this article can be found in Appendix B of this thesis.

CHAPTER 5

Impact of ultrasound-induced cavitation on the fluid dynamics of water and sewage sludge in ultrasonic flatbed reactors

This chapter has been previously published with editorial changes as follows: Lippert, T., Bandelin, J., Schlederer, F., Drewes, J.E., Koch, K. (2019). Impact of ultrasound-induced cavitation on the fluid dynamics of water and sewage sludge in ultrasonic flatbed reactors. Ultrasonics Sonochemistry 55, 217-222. DOI: 10.1016/j.ultsonch.2019.01.024

Author contributions: Thomas Lippert was responsible for the research plan, data analysis, and manuscript preparation. Jochen Bandelin assisted in preparing the sewage sludge substitutes. The experiments were conducted by Felizitas Schlederer and Thomas Lippert. Felizitas Schlederer furthermore supported the manuscript preparation by creating 3-D drawings of the ultrasound reactor. Jörg E. Drewes and Konrad Koch supervised the study. The manuscript was reviewed by Jochen Bandelin, Jörg E. Drewes, and Konrad Koch.

Abstract

The fluid dynamics of water, thickened waste activated sludge (WAS, total solids concentration 4.4%) and digested sludge (DS, total solids concentration 2.5%) within a lab-scale ultrasonic

flatbed reactor were experimentally investigated. For a visual observation of the opaque sludge flow, sewage sludges were approximated by transparent xanthan solutions with identical flow behavior. The visualization of the flow was realized by the use of an ultrasonic reactor with a transparent panel and dye streams injected into the flow. Without ultrasonic treatment, xanthan solutions showed distinct laminar flow behavior (generalized Reynolds numbers < 1), at a flow rate of 100 L/h. In water, dye streams remained coherent as well, but with slightly unsteady features (Reynolds number $\sim$350). Activation of the ultrasound reactor caused strong fluid dynamic disturbance in the water flow and dye streams were dissolved instantly, thus indicating turbulent mixing. For the xanthan solutions, however, mixing was considerably less pronounced. The dye streams in the DS substitute (0.5% xanthan solution) remained overall in laminar shape, but exhibited an eruption-like branching and an increase in diameter with advancing treatment duration. For the solution resembling WAS (2.0% xanthan solution), only weak dye stream disruption was observed, thus indicating that WAS flow in flatbed reactors is nearly laminar during ultrasonic treatment.

5.1 Introduction

In wastewater treatment, ultrasound (US) is sometimes applied as a mechanical disintegration method to enhance the digestibility of sewage sludge. By inducing ultrasonic waves into the sludge, acoustic bubbles are agitated to reach a critical size, upon which they collapse violently. These bubble implosions cause strong microturbulences with high shear forces in the sewage sludge, which deagglomerate sludge flocs and rupture cell walls. As a result of this shear-induced disintegration, both biogas yield and organic matter removal can be significantly enhanced [19, 123].

While there is consensus that such microturbulences and the resulting shear stresses are the predominant functional principle of ultrasonic disintegration of sewage sludge [4, 19], there are knowledge gaps to fill with respect to the impact of microturbulences on the macroscale flow regime in US reactors. The importance of the flow regime relates to the distribution of treatment duration and cavitation activity within flow-through US reactors. It was shown that cavitation noise levels decrease with advancing distance to the US transducers [125, 132] so that the treatment impact is considerably higher in the vicinity of the transducer surfaces than for instance in the channel center. A flow profile with laminar features would sharpen this treatment inhomogeneity, as laminae with the lowest cavitational activity would additionally exhibit the shortest treatment duration. A turbulent profile with transversal mixing, on the contrary, could homogenize the treatment impact.

The risk of laminar, undisturbed flow applies predominantly to tubular or flatbed US reactors, due to their entirely unobstructed flow path and the comparably low ultrasonic intensity [43, 133]. Hence, the aim of the present study was to evaluate whether the flow in US flatbed reactors exhibits turbulent features with transversal mixing under the impact of ultrasonic irradiation or not. For the first time, the fluid dynamics of water and two sewage sludges conventionally used for US treatment (i.e., waste activated sludge, WAS, and digested sludge, DS), were visually investigated within a novel US flatbed reactor. To allow for a visual observation of the flow field, the opaque sewage sludge was substituted by transparent xanthan solutions of identical non-Newtonian flow behavior. Dye streams injected into the transparent liquid and a transparent panel in the reaction chamber were used to visualize the fluid dynamics, for a qualitative determination of the flow regime in the US flatbed reactor.

5.2 Materials and methods

5.2.1 Approximation of sewage sludge by the use of aqueous xanthan gum solution

Due to the opacity of sewage sludge, a direct visual investigation of its fluid dynamics is not feasible. Thus, an aqueous xanthan gum solution was used as substitute. Just like sewage sludge, xanthan solution exhibits non-Newtonian, shear-thinning flow behavior, which renders the solution a suitable surrogate to simulate sludge flow as proven in previous studies [134, 135]. Due to its high solubility and its high level of transparency, a Cosphaderm X34 xanthan gum (Cosphatec GmbH, Hamburg, Germany) was utilized.

As references for the xanthan solutions, samples of thickened WAS and DS were used, as both sludge types were reported to be eligible substrates for US treatment [12, 43, 67]. The sampling of the two sludges was conducted at the Traunstein and Starnberg municipal wastewater treatment plants (WWTPs) (both located in the metropolitan area of Munich, Germany), whereas WAS was sampled after the belt filter and DS was taken from the recirculation loop of the anaerobic digester. Average total solids (TS) concentrations of WAS and DS amounted to 4.4% and 2.5% of fresh matter, respectively.

The mixing ratio of xanthan and water was iterated until the xanthan solutions exhibited the same relationship between shear rate and apparent viscosity as the sewage sludges. The determination of the shear-dependent viscosity was carried out by the use of a rotational viscometer (Viscotester VT 500, HAAKE Messtechnik GmbH & Co., Karlsruhe, Germany) at a constant temperature of 20 °C. For an accurate WAS and DS approximation, final mixing ratios of xanthan and water were determined to be 2.0:100 and 0.5:100, respectively.

The fit between the viscosity curves of the sludge samples and the two xanthan solutions was assessed through descriptive statistics. The statistical analysis included the evaluation of (i) the coefficient of determination (R^2), (ii) the slope of the regression line, and (iii) the coefficient of variation of the root mean square error (CV(RMSE)), according to [136].

5.2.2 Experimental setup

5.2.2.1 Ultrasonic reactor

For the sonication experiments, a US flatbed reactor (BANDELIN electronic GmbH & Co. KG, Berlin, Germany) was employed. The lab-scale reactor features a rectangular reaction chamber (length = 1,000 mm, width = height = 80 mm) with a total of six US transducers mounted at the top and the bottom of the chamber (three transducers each). The reactor reached an ultrasonic power of 300 W and was operated at a fixed frequency of 25 kHz, which was found to be within the most effective frequency range for sewage sludge sonication [4, 26]. A schematic representation of the reactor is displayed in Figure 5.1.

5.2.2.2 Fluid flow in the ultrasonic reactor

The flow rate of water and xanthan solutions through the lab-scale reactor was set to 100 L/h, to achieve a laminar flow field within the deactivated US reactor and to simulate specific energy inputs (defined as kJ/kg_{TS}) representative for full-scale installations. At the selected flow rate of 100 L/h, theoretical specific energy inputs of ~250 kJ/kg_{TS} and ~430 kJ/kg_{TS} were determined for the investigated WAS and DS samples, respectively, which is well in the range of energy inputs feasible in WWTPs [76, 133]. The equation for calculating the specific energy input is given elsewhere [133].

To ensure a steady flow without e.g., pump-related pulsing, fluid flow was driven by a constant overpressure in a feed tank (3 bar) that was connected to the reactor inlet, while the adjustment of the flow rate was conducted using a ball valve between inlet and feed tank. To assure a complete filling (i.e., a completely confined flow), the reactor was placed on an inclined plane so that the outlet was the highest point of the setup.

To characterize the initial flow regime in the reaction chamber without sonication, Reynolds (Re) and generalized Re (Re_{gen}) numbers were calculated for water and xanthan solutions, respectively, according to Metzner and Reed [137]:

$$Re = \frac{\rho \cdot \bar{u} \cdot D}{\eta} \tag{5.1}$$

$$Re_{gen} = \frac{\rho \cdot D^n \cdot \bar{u}^{2-n}}{K \cdot (\frac{3n+1}{4n})^n \cdot 8^{n-1}} \tag{5.2}$$

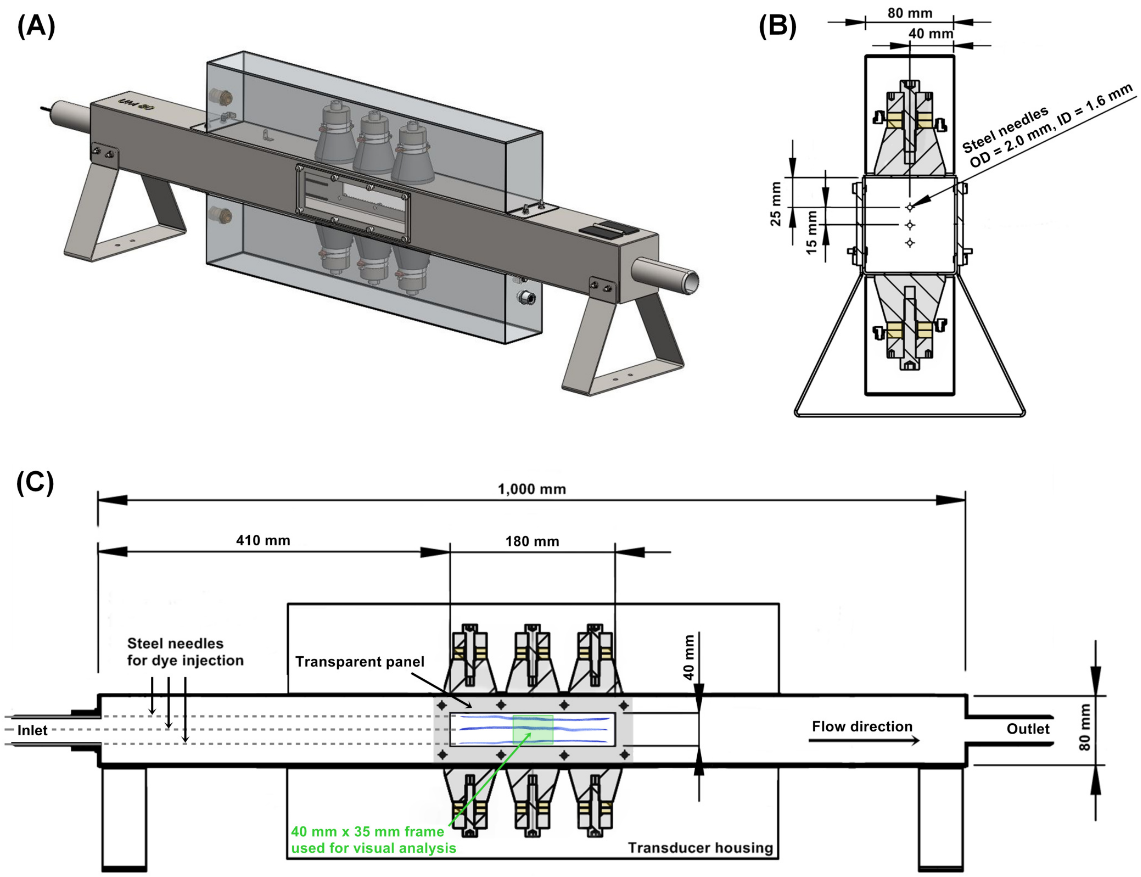

Figure 5.1: Schematic of the ultrasonic flatbed reactor setup, including a three-dimensional drawing of the reactor (A), a cross-sectional plot (B, where OD is outer diameter and ID is inner diameter), and a longitudinal section plot (C).

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]. By the use of a pycnometer, the densities of water and both aqueous xanthan solutions were determined to be ∼1,000 kg/m^3.

For the determination of K and n, the viscosity functions for both xanthan solutions were approximated by the Ostwald-de-Waele model, according to Ratkovich et al. [138]:

$$\tau = K \cdot \gamma^n \tag{5.3}$$

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 the use of the least squares method.

5.2.2.3 Visualization of fluid flow

To visualize the flow of water and xanthan solutions, dye streams were injected into the liquid flow by the use of medical infusion sets connected to three steel needles that were immersed into the reaction chamber. The inner and the outer diameter of the needles amounted to 1.6 mm and 2.0 mm, respectively, with their locations being disclosed in subfigures 5.1B and 5.1C.

Two transparent panels were centrally embedded into the reactor walls (width = 180 mm, height = 40 mm) to allow for an insight into the reaction chamber. A manufacturing of the panels over the entire height of the chamber was not possible due the constraints of a waterproof construction. The flow of the dye streams before and during US treatment was recorded in the center of the cavitational field (indicated in subfigure 5.1C) by the use of a Sony α 5000 digital camera (Sony Corporation, Tokyo, Japan).

5.3 Results and discussion

Fluid dynamics of water and two xanthan solutions (resembling surrogates for WAS and DS) within an ultrasonic flatbed reactor were visualized by the use of dye streams injected into the flow. The behavior of the dye streams before and during ultrasonic treatment was recorded through a transparent panel in the wall of the reaction chamber. The objective of the study was to qualitatively investigate, if cavitation-induced microturbulences provoke macro-scale mixing in initially nonturbulent flows of water and sewage sludge.

5.3.1 Fit between sewage sludge and xanthan solution

The agreement between sewage sludges and xanthan solutions regarding their viscosity – shear rate relationship is depicted in Figure 5.2. Descriptive statistics indicate that the fit between sewage sludges and the according xanthan solutions is generally good. For both pairs ($WAS_{Reference}$ - $WAS_{Xanthan}$ and $DS_{Reference}$ - $DS_{Xanthan}$), R^2 values, slopes of the regression lines and CV(RMSE) values were larger than 0.99, close to 1, and smaller than 10%, respectively. Hence, both xanthan solutions were found to be suitable surrogates for WAS and DS, thus confirming previous studies [134, 135]. Moreover, results suggest that the employed easy-to-use and highly transparent Cosphaderm X34 xanthan is equally applicable as conventional xanthan gum solutions.

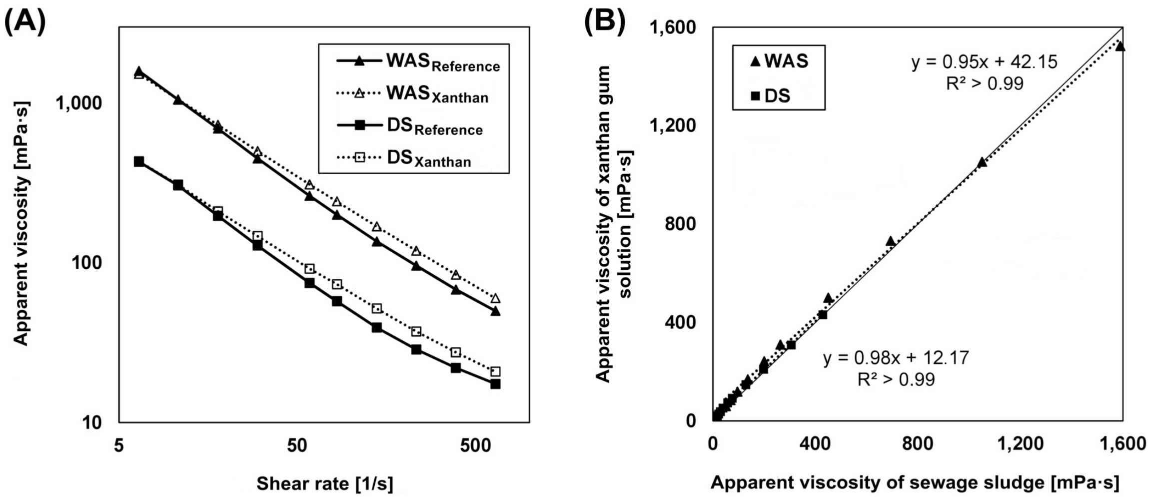

Figure 5.2: Relationship between shear rate and apparent viscosity for $WAS_{Reference}$, $WAS_{Xanthan}$, $DS_{Reference}$, and $DS_{Xanthan}$ in logarithmic scale (A), and linear regressions for the fit between sludges and xanthan solutions (B).

5.3.2 Flow regimes in the reaction chamber

5.3.2.1 Fluid dynamics without US

The observed flow patterns within the deactivated US reactor are depicted in Figure 5.3A, for all liquids tested. In the case of water, dye streams showed slightly unsteady features, despite a laminar Re number of ~350. Such flow disturbances might have arose from remaining entrance effects or irregularities on the wall surface. However, the flow was still considered steady enough to allow for a meaningful investigation of the effects of US treatment.

For the xanthan solutions on the contrary, dye streams were almost perfectly laminar at a flow rate of 100 L/h, which corresponds well to the very low Re_{gen} numbers of both xanthan solutions ($Re_{gen} \approx 0.14$ for $DS_{Xanthan}$ and $Re_{gen} \approx 0.03$ for $WAS_{Xanthan}$). The result is in good agreement with Eshtiaghi et al. [139], who found that enormous pipe flow rates of over 30 m^3/h were necessary to render the flow of sewage sludge transitional.

5.3.2.2 Fluid dynamics during ultrasonic treatment

The effects of US treatment on the flow are illustrated in Figure 5.3B, whereas the presented photographs display the conditions 6 s, 12 s, and 24 s after US reactor activation. After 24 s, a qualitative steady state of the flow pattern was reached, as the fluid retention time in the cavitational field (until the end of the frame used for visual investigation) amounted to approximately 22 s, based on an average flow velocity of ~0.5 cm/s.

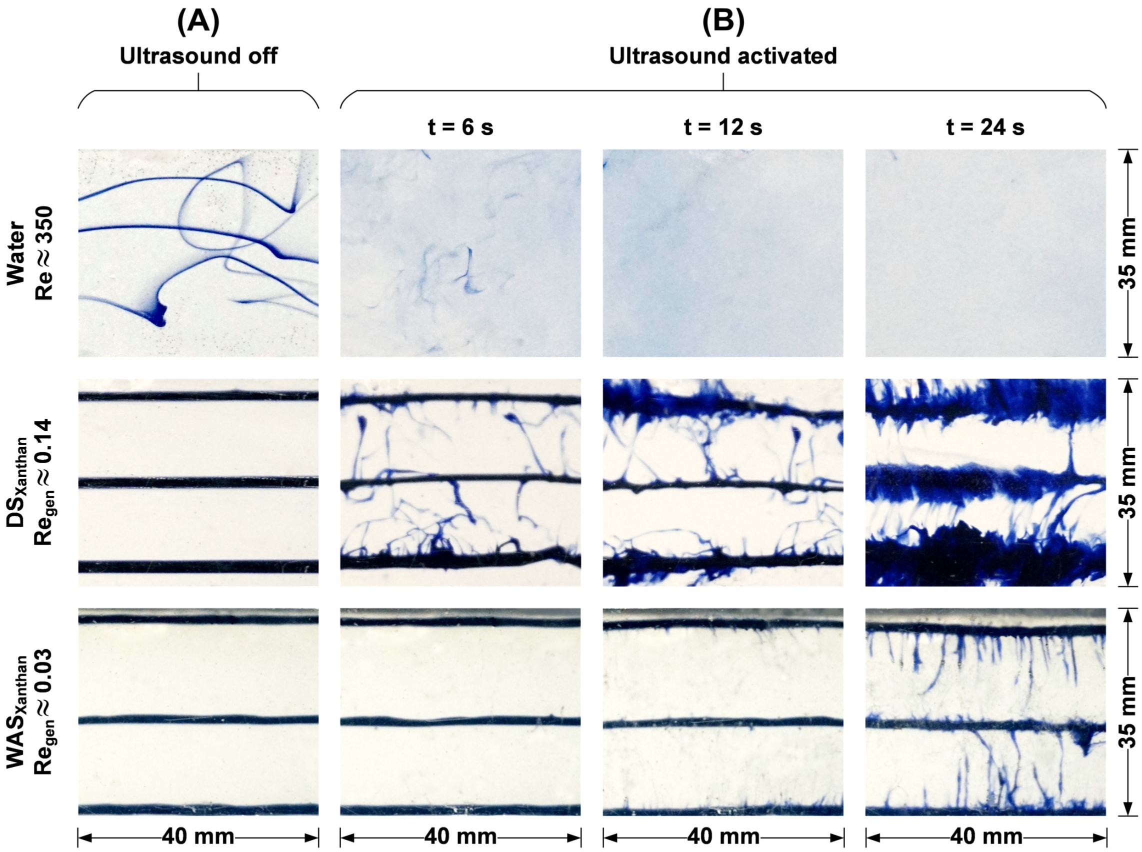

Figure 5.3: Photographs representing the flow conditions of water, $DS_{Xanthan}$ and $WAS_{Xanthan}$ before US treatment (A), and during ultrasonic irradiation at different times after treatment activation (B). The images represent the flow in the center of the cavitational field, as indicated in Figure 5.1C

For water, it can be seen that the dye streams are strongly disrupted already at t = 6 s, and completely mixed after 12 s and 24 s (entire fluid volume is colored in light blue). The results suggest that the activation of the US system created enough turbulence to completely mix the liquid. The observation agrees well with previous research, which similarly found strong hydrodynamic movement of water already at low to moderate ultrasonic irradiation [140–142]. Hereby, it should be noted that the pronounced mixing might not exclusively originate from imploding cavitation bubbles, but was potentially also amplified by the mechanical vibration of the ultrasonic reactor.

For the xanthan solutions, on the contrary, a completely different behavior was observed. Regardless of the impact of US, the initial shape of the dye streams remained clearly visible throughout the treatment for both $DS_{Xanthan}$ and $WAS_{Xanthan}$ and a complete mixing of the liquid, as observed for water, did not occur. However, eruptions of small dye streaks from the main stream were observed with advancing treatment time, while the intensity of these quickly occurring outbursts was considerably higher for $DS_{Xanthan}$ than for $WAS_{Xanthan}$. For $DS_{Xanthan}$, dye streams showed a considerate increase in diameter and a pronounced branching after a treatment duration of 24 s, while only slight streak formation was observed for $WAS_{Xanthan}$. It has to be noted that stronger flow instabilities might have occurred directly at the transducer plates, which was, however, not observable due to the limited height of the transparent panel. Based on the local character of the observed outbursts and a previous study verifying strong cavitational activity within the reaction chamber [125], it is hypothesized that mainly cavitation events were responsible for the observed fluid motion, despite the potential impact of the mechanical vibration of the reactor wall. A link to a video sequence displaying the flow behavior of the dye streams in water and both xanthan solutions before and during US irradiation can be found in Appendix C (Video S1).

Based on the visual observations, it seems therefore justified to attest a more or less laminar flow profile with only minor US-induced transversal mixing for the sewage sludge substitutes, especially for $WAS_{Xanthan}$. In this context, it has to be noted, that the investigated WAS sample (and hence its $WAS_{Xanthan}$ surrogate) still resembles a sludge with low to moderate viscosity, owing to its TS of only 4.4%. In practice, WAS samples can exhibit considerably higher TS contents of up to 7% [133], which would presumably entail an even more pronounced suppression of US-induced transversal mixing. Moreover, it should be considered that an increased sludge viscosity would not only damp the fluid motion that originates from the imploding cavitation bubbles, but would also decrease the intensity of cavitation events due to a more pronounced sound wave attenuation [125].

A potential strategy to enhance US-induced flow instabilities could be an increase of the flow rate due to the affiliated augmentation of flow inertia and the reduction of the viscosity of the shear-thinning sludge. However, calculations have shown that even a hundred-fold increase in flow rate (10 m^3/h instead of 100 L/h) still resulted in laminar Re_{gen} numbers of 53.0 and 13.5 for $DS_{Xanthan}$ and $WAS_{Xanthan}$, respectively. It remains therefore questionable, if the US treatment could have induced macro-scale turbulence in the still laminar flow regime. Moreover, specific energy inputs would drop to an almost insignificant level (< 5 kJ/kg_{TS}) due to the considerably shorter treatment duration, which would most likely negate any effect of the treatment. It is therefore hypothesized that an increase in flow rate is no practical strategy to enhance cavitation-induced mixing of viscous sludges in ultrasonic reactors. However, future studies could investigate in more detail how US irradiation can contribute to an earlier onset of the laminar/turbulent transition in viscous media flow.

5.3.3 Outlook regarding future investigations

By offering an insight into the flow regimes within US flatbed reactors, the presented findings are especially relevant for future modeling approaches. In previous studies, relatively complex turbulence models including e.g., moving boundary conditions and dynamic meshes were used to describe liquid motion in ultrasonic reactors [141–143]. However, the visual observation of the flow fields suggests that a laminar flow model can already provide meaningful estimates of the dynamics of viscous media flow. To further validate the admissibility of the laminar flow assumption, a particle image velocimetry (PIV) could be conducted in subsequent research. By coupling a PIV with a laminar flow simulation, the degree of compliance between the actual flow regime and the assumed laminar flow can be quantified using the Bray- Curtis similarity [144]. However, it is assumed that a PIV analysis, which is also based on a visual observation of the flow field, would lead to a very similar conclusion on the flow regime.

It has to be noted, however, that the presented findings only reflect on reactors with wall oscillation. Sonotrode reactors with considerably higher oscillation amplitudes (∼20 – 60 μm sonotrode oscillation vs. ∼6 – 8 μm wall oscillation [43]) and significantly smaller oscillating surfaces (sonotrode tips vs. reactor walls) might be able to cause a stronger disruption of the flow due to the much higher ultrasonic intensity (defined as W/m^2). Aside the higher intensity, the sonotrodes might additionally act as flow-disturbing baffles, thus further promoting an earlier onset of turbulent flow features. This considerate difference between the two reactor types might, thus, potentially limit the laminar flow assumption to tubular or flatbed US reactors. Subsequent research should therefore systematically investigate how ultrasonic intensity and reactor design affect flow conditions and the mixing of sludge, in order to achieve a more homogeneous exposure of the substrate towards sonication in spite of its high viscosity.

Nonetheless, results indicate that in ultrasonic flatbed reactors, the major fraction of the flow is only poorly mixed, when viscous media such as sewage sludge are treated. Especially for $WAS_{Xanthan}$, an inhomogeneous exposure to the cavitational field due to a nearly laminar flow profile seems to be a challenge. Future studies should therefore quantify the degree of treatment inhomogeneity and investigate, how such differences in exposition can be minimized, e.g., by an optimized reactor design.

5.4 Conclusions

This study investigated the flow of water and two sewage sludge substitutes (transparent xanthan solutions, resembling WAS and DS) within an ultrasonic flatbed reactor. Flow was visualized using dye streams injected into the bulk liquid and was observed through a transparent panel in the reactor wall. In water, ultrasonication led to a complete mixing of the liquid, while in sewage sludge, dye streams remained mostly laminar and were only slightly disrupted. Results suggest that cavitation-induced microturbulence does not lead to a significant mixing of viscous media within US flatbed reactors. Future research should however validate the qualitative results, e.g., by applying a PIV.

5.5 Acknowledgments

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5.6 Appendix - Supplementary material

The graphical abstract and supplementary material to this article can be found in Appendix C of this thesis.

CHAPTER 6

Effects of ultrasonic reactor design on sewage sludge disintegration

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Author contributions: Thomas Lippert was responsible for the research plan, data analysis, and manuscript preparation. Jochen Bandelin planned and designed the US reactor. The experiments were conducted by Felizitas Schlederer, Jochen Bandelin, and Thomas Lippert. Computational fluid dynamic simulations were conducted by Felizitas Schlederer and Thomas Lippert. Felizitas Schlederer furthermore supported the manuscript preparation by creating 3-D drawings of the ultrasound reactor. Jörg E. Drewes and Konrad Koch supervised the study. The manuscript was reviewed by Jochen Bandelin, Jörg E. Drewes, and Konrad Koch.

Abstract

The impact of ultrasound (US) reactor design on cavitation intensity distribution and disintegration efficiency was studied for sewage sludge pre-treatment, using a US flatbed reactor of variable reaction chamber height (RCH, 20 - 100 mm). Mapping of cavitation intensity and treatment effects was conducted using (i) hydrophone measurements, (ii) aluminum foil tests, and (iii) soluble chemical oxygen demand (COD) analyses. The overall disintegration efficiency was evaluated based on average COD solubilization. The impact of flow on treatment (in)homogeneity was additionally examined using computational fluid dynamics (CFD). Results of all measurement techniques suggest that small RCHs (20 mm, for instance) enable uniform and intense treatments, while large RCHs, which are subjected to strong sound wave attenuation, entail inhomogeneous treatments where large fractions of substrate are no longer exposed to notable cavitation activity. For instance, COD solubilization (relative to alkaline hydrolysis) in the channel center dropped from 6.4% to zero as RCH widened from 20 mm to 100 mm. Flow-through sonication further aggravates treatment inhomogeneity due to the high flow rates in the low-cavitation channel centers. The overall disintegration efficiency declined with increasing RCH, showing a drop in average COD solubilization by 73% from RCH = 20 mm to RCH = 100 mm. The drop correlated with average cavitation noise levels ($R^2 = 0.82$), indicating that hydrophone measurements may be a suitable tool for US reactor design optimization. Overall, results suggest that reactor geometry has a critical impact on both treatment (in)homogeneity and treatment efficiency and that equal specific energy inputs do not imply equal US treatments.

6.1 Introduction

In wastewater treatment, ultrasound (US) can be applied as a mechanical pre-treatment method for sewage sludge. Typically, the aim of such pre-treatments is to increase the digestibility of recalcitrant sludge types (such as waste activated sludge, WAS), by breaking up the sludge´s floc structure and by rupturing the microbial cell walls [19, 71]. Due to the hereby increased surface area and the release of intracellular sludge components, the performance of the subsequent anaerobic digestion step can be enhanced through increased methane yields, accelerated reaction kinetics, and improved organic matter removal [35, 91, 97, 145].

In the case of US pre-treatments, sludge disintegration is facilitated by transient acoustic cavitation, i.e., the formation, growth, and collapse of acoustic micro-bubbles within the sludge matrix [47]. The extreme violence of the bubble implosions (local temperatures of 4,000 - 15,000 K [54], pressures of up to 1.3 GPa [55]) inflicts high shear forces upon the surrounding liquid, eventually leading to a mechanical disintegration of the sludge [19]. Thus, strong cavitational activity is the key pre-requisite for successful sludge disintegration.

The strength of cavitational activity in ultrasonic reactors is controlled by several factors, including US frequency, US intensity, and reactor geometry. While the impact of both frequency and intensity is already well-researched in the field of sewage sludge sonication (see [4, 77, 146–148]), literature addressing reactor geometry is still scarce. At the same time, design aspects seem especially relevant for sludge pre-treatments. First, the high total solids (TS) content of sludge causes strong sound wave attenuation, which leads to a sharp decrease in cavitational activity with advancing distance to the US transducers [114, 125]. Second, the high viscosity of the sludge was shown to suppress cavitation-induced mixing, which means that a non-uniform distribution of cavitation intensity necessarily leads to an inhomogeneous exposition of the sludge towards the treatment [149]. Third, ultrasonication of sewage sludge is commonly conducted using flow-through reactors (especially in full-scale applications at wastewater treatment plants, WWTPs) [12] so that treatment conditions also depend on the fluid dynamics in the reactor. For instance, flow rates might be highest in the center of the reaction chamber, where the cavitation field is most attenuated. Therefore, the flow condition might further aggravate treatment inhomogeneity in unfavorable reactor designs.

According to these challenging boundary conditions, previous studies addressing US reactor geometry could confirm its critical impact on sludge disintegration. For instance, Nickel and Neis [78] observed that a full-scale US reactor had more than twice the disintegration efficiency than a lab-scale batch system, despite the same energy inputs provided by the same ultrasonic probe. A similar finding was made by Zielewicz [56] who contrasted three lab-scale US reactors (ultrasonic bath, sonotrode tip, and flat emitter) regarding their efficiency in causing cell lysis and sludge floc de-agglomeration. Despite constant energy inputs, sonication efficiencies varied significantly among the tested setups. However, a clear relationship between geometry and performance could not be established in either study, due to rather non-systematic changes in reactor design [78] or due to structural differences among the reactor systems [56].

To gain a more mechanistic understanding of how reactor design affects sonication performance, cavitation noise (i.e., the sound frequencies emitted by imploding cavitation bubbles) was measured in three identical US flatbed reactors that were only differing in reaction chamber height (RCH, 40 mm, 60 mm, and 80 m) in our previous study [125]. Results demonstrated that cavitation noise in the channel center gradually decreased with increasing RCH, indicating that small chamber spacings offer the most intense cavitation fields. However, as the relationship between primary effects as cavitation noise and secondary treatment effects (such as sludge solubilization) is yet to be established, the study could not conclude with full certainty that the small reactor gap would also lead to higher sludge disintegration.

Therefore, the objective of the present study was to systematically investigate how reactor geometry affects sewage sludge disintegration, by considering both primary and secondary treatment effects. For this purpose, a size-adjustable US flatbed reactor similar to the reactors of our previous study [125] was used, which allowed testing five different RCHs ranging from

20 mm to 100 mm. All sonications were conducted at the same specific energy input using thickened WAS as a representative test medium. The spatial distribution of cavitation intensity within the different reaction chambers was determined using hydrophone measurements, while the distribution of secondary effects was mapped using aluminum foil tests and soluble COD analyses. The overall disintegration efficiency was assessed based on the average COD solubilization of each reactor design. To additionally account for flow-through sonication, the flow of the non-Newtonian WAS through the different chamber geometries was simulated using computational fluid dynamics (CFD). Based on the CFD simulations, the distribution of mass flow rates and hydraulic residence times (i.e., exposure times) and their effect on the overall treatment (in)homogeneity could be estimated. Hence, for the first time, the effects of US reactor design were investigated with respect to (i) cavitation intensity distribution, (ii) treatment effect distribution, (iii) resulting treatment (in)homogeneity, and (iv) overall disintegration efficiency.

6.2 Materials and methods

6.2.1 Source and characteristics of the investigated WAS sample

Thickened WAS was collected at the municipal Freising WWTP (located 35 km north of Munich, Germany) after the belt thickener. Hydrophone measurements and flow simulations were conducted using one batch of sludge (sample #1), while aluminum foil tests and soluble COD analyses were performed using another batch (sample #2). Due to the same source and the verifiably small difference, all conducted experiments were deemed directly comparable to each other. The sludge was analyzed according to standard methods [126], and the average characteristics of both samples are displayed in Table 6.1.

Table 6.1: Average characteristics of the investigated WAS sample.

Parameter	**WAS (sample #1)**	**WAS (sample #2)**
TS [% of FM]	4.6	4.4
VS [% of TS]	73.3	68.5
Density [kg/L]	1.0	1.0
COD [mg/L]	n.d.	58,000
Soluble COD [mg/L]	n.d.	480

WAS = Waste activated sludge, TS = Total solids, FM = Fresh matter,
VS = Volatile solids, COD = Chemical oxygen demand

For modeling the fluid flow of the non-Newtonian WAS, the relationship between shear rate and apparent viscosity was determined for sludge sample #1 using a rotational viscometer (Viscotester VT 500, HAAKE Messtechnik GmbH u. Co., Karlsruhe, Germany). Measurements were carried out in triplicate at a controlled temperature of 20 °C.

6.2.2 US test reactor setup

For the sonication experiments, a customized size-adjustable ultrasonic flatbed reactor was used (BANDELIN electronic GmbH & Co. KG, Berlin, Germany). The system features a box-shaped reaction chamber, formed by two opposite, identical ultrasonic plate reactors (each powered by ten ultrasonic transducers), and a surrounding steel case. The case was adjustable so that the distance between the plate reactors (defined as RCH) was variable between 20 mm and 100 mm. The width and the length of the reaction chamber amounted to 160 mm and 450 mm, respectively, according to the circumference of the plate reactors. Sonication tests were performed at five different RCHs (20 mm, 40 mm, 60 mm, 80 mm, and 100 mm) at a constant electric power of 1 kW (i.e., 500 W for each plate reactor). All US transducers were connected in parallel and powered by a single ultrasonic generator with a constant frequency of 25 kHz, which has been identified to be within the most efficient frequency range for sewage sludge disintegration [4, 26]. The diameter of the transducers amounted to 60 mm, and the distance between the transducers was 4 mm, both in the horizontal and the vertical direction. A schematic representation of the ultrasonic reactor setup for RCH = 100 mm is displayed in Figure 6.1.

6.2.3 Sonication tests

All treatments were carried out in batch mode at a constant specific energy input of approximately 200 kJ per kg_{TS}, which was calculated according to Eq. 6.1:

$$SE = P \cdot \frac{t}{V} \cdot \frac{1}{\rho \cdot TS} \tag{6.1}$$

where SE is the specific energy [kJ/kg_{TS}], P is the electric power of the reactor system [kW], t is the treatment time [s], V is the volume of the sonicated sludge [L], ρ is the density of the sludge [kg_{FM}/L], and TS is the total solids concentration of sludge [kg_{TS}/kg_{FM}]. The comparably low specific energy input of 200 kJ/kg_{TS} was selected to mimic treatments feasible in full-scale installations [76, 133].

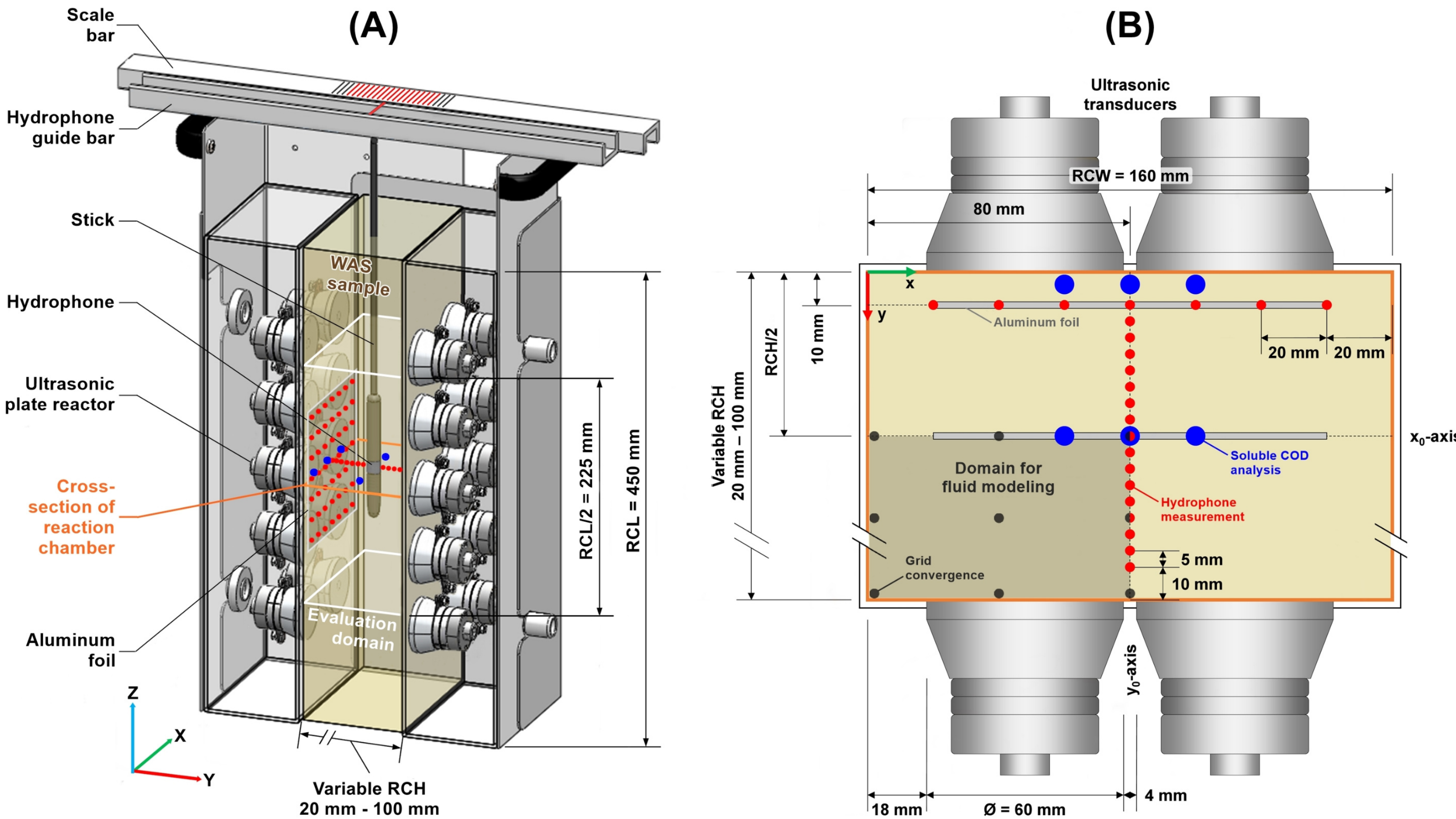

Figure 6.1: 3-D schematic of the employed ultrasonic reactor system, including the two ultrasonic plate reactors and the steel case (subfigure A) and 2-D plot of the cross-section in the center of the reaction chamber (subfigure B), with WAS = waste activated sludge, RCH = reaction chamber height, RCL = reaction chamber length, RCW = reaction chamber width, and COD = chemical oxygen demand. Measurement locations for cavitation noise and soluble COD are indicated by red and blue dots, respectively, while grey dots indicate probe points for the grid convergence study of the fluid flow simulation. The positions of the aluminum foils are disclosed in subfigure (B). The domain used for the evaluation of cavitation noise levels and fluid flow is highlighted in subfigure (A). For better visibility, the front part of the steel case and the frame holding the aluminum foil are not shown.

Table 6.2: Conditions of the sonication tests depending on RCH.

RCH [mm]	Reaction chamber volume [L]	Treatment time [s]	SE input [kJ/kg_{TS}]
20	1.44	13	~ 200
40	2.88	26	~ 200
60	4.32	39	~ 200
80	5.76	52	~ 200
100	7.20	65	~ 200

RCH = Reaction chamber height, SE = Specific energy

For maintaining a constant energy input with a changing reaction chamber volume, the sonication time was adapted to the respective reactor volume, i.e., the quotient of t/V was kept constant. The resulting treatment conditions for all RCHs are displayed in Table 6.2.

6.2.4 Cavitation field analysis

Mapping of the cavitation field was conducted using a radially sensitive hydrophone with a maximum diameter of 15 mm (TC 4034-1, RESON A/S, Slangerup, Denmark) according to the IEC TS 63001:2019 norm for cavitation noise measurement [150], and similar to the studies of Grönroos et al. [77], Petkošek et al. [151], and Bandelin et al. [125]. The hydrophone recorded both the sound pressure of the ultrasonic waves (frequency range $f_0 = 25 \pm 2.5$ kHz) and the sound pressure of the cavitation bubble implosions (frequency range of 2.15 - 2.35$\cdot f_0$). The differentiation between the two ranges, as well as the quantification of the cavitation noise level (CNL), were conducted using the evaluation software KaviMeter (Elma Schmidtbauer GmbH, Singen, Germany). The software calculated CNL in decibel [dB], defined as 20 times the decadic logarithm of the ratio of the recorded cavitation noise figure to a reference value of 1 W/m^2.

To perform the CNL measurements, the sensor head of the hydrophone was submerged into the sludge-filled reaction chamber. For precise positioning of the sensor, the hydrophone was firmly fixed to a guide bar via a steel stick, which was movable along an adjacent scale bar. For resolving the cavitation field in all spatial directions, measurements were conducted both in a plane parallel to the transducer plates (in x-z-directions) and perpendicular to the transducer plates (in y-direction). The measurement in the x-z-plane comprised recordings at 49 locations covering an area of 120 mm by 120 mm and was conducted 10 mm apart to one of the two identical transducer plates at an RCH of 100 mm. For the measurements of CNL in y-direction, the hydrophone was moved across the central y_0-axis, whereas recordings were conducted every 5 mm. All measurement locations are indicated by red dots in Figure 6.1.

As the transducer arrays covered only about 70% of the reaction chamber length (RCL), the measured cavitation field was not applicable for the full geometry. Thus, to be on the safe side, the conducted CNL recordings were seen as representative for the middle half of the reaction chamber. The according domain (termed evaluation domain) with a length of RCL/2 = 225 mm is highlighted as a white box in Figure 6.1A.

The potential impact of sonication on the sludge characteristics (and hereby on sound wave attenuation) was minimized by renewing the sludge after every change of the experimental setup (i.e., every change of RCH) from the same well-mixed batch. To avoid potentially harmful direct contact between the hydrophone and the transducer surfaces, measurements were started and ended 10 mm away from the walls.

6.2.5 Approximation of CNL measurements by polynomials

The CNL values obtained on the y-axis were subsequently approximated by polynomials, using the software Origin Pro 2020 (OriginLab Corporation, Northampton, MA, USA). Due to the symmetry of the setup, CNL values were expected to be more or less symmetric to the central x_0-axis. To yield such axially symmetric polynomials from potentially slightly non-symmetric measurement data, obtained CNL values were mirrored at the x_0-axis and merged. As no CNL values were recorded within 10 mm to the walls, the obtained polynomials were extrapolated until the transducer walls by a constant linear extrapolation of the average of the outmost measurement values.

6.2.6 Analysis of the secondary effects of cavitation

The secondary effects of cavitation were examined using aluminum foil tests and soluble COD analyses, at a constant specific energy input of approximately 200 kJ/kg_{TS}.

6.2.6.1 Aluminum foil tests

The distribution of treatment effects regarding mechanical damage was investigated using aluminum foil tests according to the IEC TR 60886:1987 norm [152]. For precise positioning, the employed sheets of aluminum foil (120 mm by 120 mm, thickness 30 μm) were firstly tautened on a steel frame and then immersed into the sludge-filled reaction chamber. The frame was placed both in the center of the reaction chamber (y = 0 mm) and close to the transducer surface (10 mm apart, see Figure 6.1B), for three different RCHs (20 mm, 60 mm, and 100 mm). In the case of the 20 mm design, this resembles the same location. The different positions were tested one after another, to avoid potential shielding effects of the frame and the foil. To ensure the same conditions for each test, the sludge was renewed after every sonication. Upon completion of the treatments, the frame with the aluminum foil was carefully taken out of the reaction chamber, rinsed, and photographed for visual inspection.

6.2.6.2 Analysis of COD solubilization

The sludge disintegration efficiency was assessed by means of the degree of disintegration (DD_{COD}), defined as the ratio between ultrasonically and chemically facilitated sludge disintegration:

$$DD_{COD} = \frac{sCOD_{US} - sCOD_0}{sCOD_{NaOH} - sCOD_0} \tag{6.2}$$

where $sCOD_{US}$ is the concentration of sCOD after sonication, $sCOD_0$ is the sCOD concentration of the untreated sample, and $sCOD_{NaOH}$ is the sCOD concentration after alkaline hydrolysis (all in [mg/L]). To achieve full alkaline hydrolysis, a mixture of 100 mL sludge and 200 mL 1 M NaOH solution was stirred for 24 h at room temperature, according to Bougrier et al. [81].

For the sCOD analysis, sludge samples were filtered through a cellulose nitrate filter with a pore size of 0.45 μm using a pressurized filter holder. As only a small amount of filtrate was needed, previous centrifugation was not necessary. Subsequently, the filtrate was analyzed using Hach-Lange cuvette tests (LCK 314, 515, and 614) and a spectrophotometer (DR 2800, Hach Company, Loveland, CO, USA).

Sampling for the DD_{COD} analysis was conducted in triplicate both at the center of the reaction chamber and directly at the transducer plates (see Figure 6.1B), using a 10 mL syringe connected to a hollow needle with a length of 20 cm. In addition, the average DD_{COD} was determined as a measure for the overall disintegration efficiency. For obtaining a representative sample, the entire reactor content was collected in a canister after each sonication and thoroughly mixed.

6.2.7 Modeling fluid flow within the US reactor

6.2.7.1 Selection of flow rate

In order to achieve the same average exposure times within the evaluation domain as in the batch sonications, the flow rate was set to 200 L/h, according to Eq.6.3.

$$Q = \frac{A}{\bar{t}_e} \cdot \frac{RCL}{2} \tag{6.3}$$

where A is the cross-section of the reaction chamber [m^2], $\bar{t_e}$ is the average exposure time [s], and RCL/2 is the length of the evaluation domain [= 0.225 m].

6.2.7.2 Modeling the viscosity of WAS

To account for the non-Newtonian flow behavior of the WAS sample in the CFD model, the previously obtained relationship between viscosity and shear rate was approximated using the Ostwald-de-Waele model, due to its simplicity and its goodness fit [35, 138]:

$$\mu = K \cdot \dot{\gamma}^{n-1} \tag{6.4}$$

where μ is the apparent viscosity [Pa·s], K is the consistency index [Pa·s^n], $\dot{\gamma}$ is the shear rate [1/s], and n is the power-law index [-]. Consistency index K and power-law index n were fitted to the experimental data using the least-squares method to obtain values of 14.96 Pa·s^n for K and 0.17 for n.

6.2.7.3 Fluid domain modeling

To ensure unmitigated compatibility between the results of the CNL analysis and the results of the flow simulations, the domain used for fluid modeling was also limited to the evaluation domain of the reaction chamber. In order to assess if a fully developed flow profile undisturbed by entrance or exit effects prevails in the domain, entrance lengths l_e were calculated for all RCHs. Due to the high viscosity of the sludge and the comparably low flow velocities, obtained entrance lengths were rather short (maximum l_e = 78 mm for RCH = 100 mm) so that a fully developed flow profile could be safely assumed for the centrally located evaluation domain. The equation for calculating l_e is given elsewhere [153].

As fully developed flow profiles remain constant in flow direction, flow was modeled using a periodic domain. Moreover, owing to the twofold symmetry of the reaction chamber, only a quarter of the cross-section was required for CFD modeling (see Figure 6.1B). The length of the domain (in the periodic z-direction) was arbitrarily selected to be 1 mm. The resulting 3-D domain and the applied boundary conditions can be found in Appendix D, Figure D.2.

6.2.7.4 Computational grid and grid convergence assessment

The five different domains were discretized using hexahedron mesh elements with quadratic cross-sections and 25 layers of ultrathin hexahedron elements at the no-slip boundaries to resolve the steep velocity gradients in wall vicinity. A detail of the finally applied mesh is given in Appendix D, Figure D.2. For the most efficient use of the computational resources, the length of the cuboid mesh elements in z-direction was set to 1 mm, equal to the length of the periodic domain.

To assess the convergence of the computational grid, i.e., the independence of the results from the mesh resolution, all simulations were conducted on three different meshes of increasing accuracy, with hexahedron side lengths in the x-y plane of 1,000 μm, 500 μm, and 250 μm, resembling a constant grid refinement ratio of 2. Grid convergence was assessed according to the grid convergence index (GCI), as proposed by Roache [154]. GCI calculations were based on the velocity magnitudes at nine probe points (indicated as grey dots in Figure 6.1B), with locations both in the bulk flow and near to the no-slip walls (0.1 mm distant to the walls).

Calculated GCI values for the finest mesh resolution were found to be below 1%, at all probe points, and for all tested domain sizes, which is considered very good grid convergence, especially for singular values. The probe point in the left lower corner poses the only exception, with GCI values ranging from 5.8 - 11.2% due to its high exposition to boundary effects. However, even such a range of GCI values is still considered sufficient for CFD simulations [155]. A further decrease in cell size or the addition of more surface mesh refinements did, therefore, not result in an improved result accuracy. Final meshes comprised 66,170 to 232,090 elements, depending on RCH.

6.2.7.5 Flow model

Fluid flow through the periodic domain was computed according to the incompressible, laminar, steady-state Navier-Stokes equations using COMSOL Multiphysics 5.4a (COMSOL, Inc., Burlington, MA, USA):

$$\rho(\boldsymbol{u} \cdot \nabla)\boldsymbol{u} + \nabla p = \nabla \cdot (\mu \nabla \boldsymbol{u}) \tag{6.5}$$

$$\nabla \cdot \boldsymbol{u} = 0 \tag{6.6}$$

where $\boldsymbol{u} = (u_x, u_y, u_z)$ is the velocity vector [m/s], and p is the pressure [Pa]. The flow through the periodic domain was driven by a pressure difference between the inlet and outlet faces. This difference was iterated using an additional constraint until the actual flow rate Q_{in} was equal to the targeted flow rate Q_{set} (50 L/h), according to Horstmeyer et al. [156]. The reduction from 200 L/h to 50 L/h stems from the smaller cross-section of the symmetric fluid domain, only being a quarter of the full cross-section.

$$\int_{A_{Inlet}} u_z dA = Q_{in} = Q_{set} \tag{6.7}$$

The assumption of the incompressibility of the WAS sample was verified beforehand using a pressurized acrylic glass cylinder. Exposing the sludge to compressed air at a gauge pressure of up to 4 bar did not cause a notable compression of the fluid. Considering typical operating pressures of around 1 - 2 bar in WAS pipes at WWTPs [125], the assumption of an incompressible fluid was hence deemed acceptable.

To check for the validity of the laminar flow assumption, generalized Reynolds ($\mathrm{Re}_{\mathrm{gen}}$) and critical $\mathrm{Re}_{\mathrm{gen}}$ numbers (including consistency index K and power-law index n) were calculated for the non-Newtonian WAS flow according to Metzner and Reed [137]:

$$Re_{gen} = \frac{\rho \cdot D^n \cdot \bar{u}^{2-n}}{K \cdot (\frac{3n+1}{4n})^n \cdot 8^{n-1}} \tag{6.8}$$

$$Re_{gen,crit} = \frac{6464n}{(3n+1)^2} \cdot (2+n)^{\frac{2+n}{1+n}} \tag{6.9}$$

where $\bar{u}$ is the average flow velocity [m/s], and D is the hydraulic diameter [m]. With resulting $\mathrm{Re}_{\mathrm{gen}}$ values between 0.01 and 0.11 and a resulting $\mathrm{Re}_{\mathrm{gen,crit}}$ number of 2,030, the flow was found to be clearly within the laminar region at the given flow rate of 200 L/h, for all RCHs tested.

In terms of a potential impact of cavitation on the fluid dynamics, it was previously found that cavitation-induced micro-turbulences are not powerful enough to notably disrupt the laminarity of viscous WAS flow in US flatbed reactors [149]. This observation is deemed fully applicable to the present study due to (i) a very similar flatbed reactor geometry, (ii) a similar range of $\mathrm{Re}_{\mathrm{gen}}$ numbers, and (iii) a sludge viscosity approximately twice as high as compared to the one reported in [149]. Hence, it was assumed that already a laminar flow model could provide meaningful estimates on the fluid dynamics of WAS flows in US flatbed reactors. Besides, it was visually observed during the experiments that the sludge largely remained at rest despite the sonication.

6.2.8 Statistical analysis

To assess the agreement between the polynomial approximations and the CNL measurements, a descriptive statistical analysis was conducted, including the evaluation of (i) the coefficient of determination (R^2), (ii) the regression line slope (m), and (iii) the coefficient of variation of the root mean square error (CV(RMSE)) according to Dandikas et al. [136].

Differences between measurement data were interpreted based on a one-way ANOVA (analysis of variance) with a post-hoc Tukey HSD (honestly significant difference) test with p values < 0.05 indicating significant differences and p values < 0.01 indicating highly significant differences. The significance tests were performed using the software Origin Pro 2020.

6.3 Results and discussion

6.3.1 Spatial distribution of cavitation noise within the different reaction chambers

6.3.1.1 Independence of CNL from the x- and z-position

The independence of CNL from the location at the transducer plate was verified by 49 triplicate measurements across a plane 10 mm distant to one of the two identical plate reactors. Both the measurement locations and the obtained CNLs are disclosed in Figure 6.2. CNLs remained at a similar level throughout the measurement plane, indicating that the dense placement of transducers allowed for more or less uniform irradiation. For assessing the observed similarity using statistical tools, measurement values were firstly grouped in x- and z-rows (as exemplified in Figure 6.2A), and then compared using a one-way ANOVA and a post-hoc Tukey HSD test. The pooling of the data in rows was conducted to avoid a comparison of all 49 measurement locations among each other, which would have led to an extensive number of $\sum_{k=1}^{49-1} k = 1,176$ significance tests.

The statistical analysis confirms that CNL is independent from z within the measurement plane, as no significant difference could be discerned between the seven analyzed x-rows ($p > 0.78$). Also, no significant difference was found for the z-rows from x = -40 mm to x = 40 mm. However, at x = -60 mm and x = 60 mm (i.e., at the outmost measurement locations close to the edges of the reaction chamber), a highly significant decline in CNLs was observable ($p < 0.01$). On average, this drop amounted to approximately 20% compared to the overall mean value. Such a reduction might be related to the central placement of the transducer array or the potentially higher resistance to transducer plate oscillation at the edge of the plate reactor. However, except for this drop, it was shown that the reactor is capable of generating a relatively homogeneous cavitation field throughout its transducer plate surface, which agrees with previous studies reporting a uniform cavitation field in multi-transducer US reactors [106, 112]. Therefore, it seemed fair to assume that CNLs recorded in the center of the measurement plane are representative for most of the transducer plate surface. For the subsequent hydrophone measurements in the y-direction, CNLs were, thus, only considered at x = z = 0.

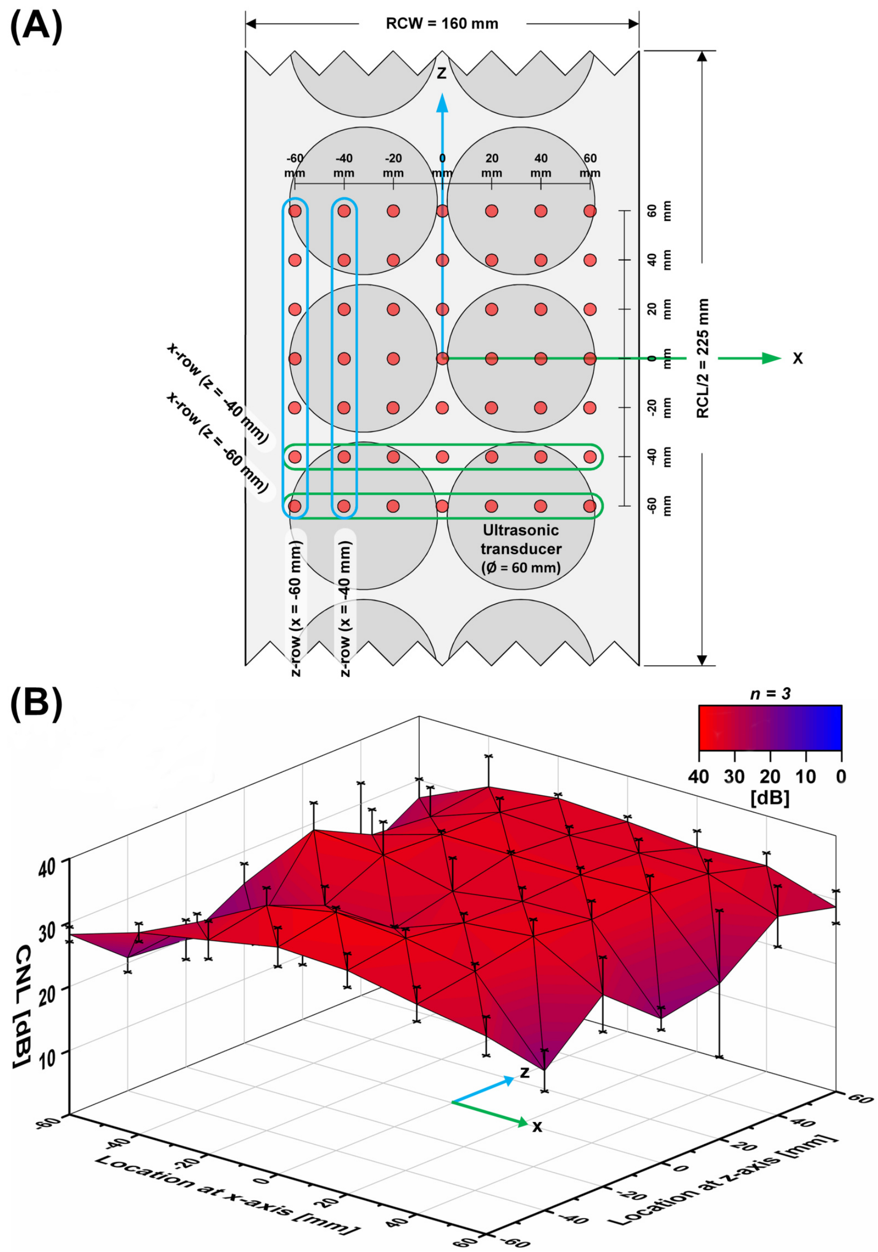

Figure 6.2: 2-D front view of the transducer plate with hydrophone measurement locations indicated by red dots (subfigure A), with RCW = reaction chamber width and RCL = reaction chamber length. Hydrophone measurements were conducted in triplicate at a distance of 10 mm to the wall, and at a constant reaction chamber height of 100 mm. Resulting average cavitation noise levels (CNLs) are depicted in subfigure (B), with error bars indicating the standard deviation. For the statistical analysis, CNL recordings were grouped in x- and z-rows, as exemplified in subfigure (A).

6.3.1.2 Dependence of CNL on the y-position

The evolution of CNLs along the y_0-axes for all RCHs is presented in Figure 6.3, whereas colored markers denote the average measurement values. The strongest cavitation noise was recorded in the 20 mm gap (CNL = 39 dB), possibly due to a concentration and reflection of the sound waves in the thin reaction chamber [125]. At RCH = 40 mm, CNLs were slightly lower (35 - 36 dB), but also remained relatively constant, indicating only minor sound wave attenuation.

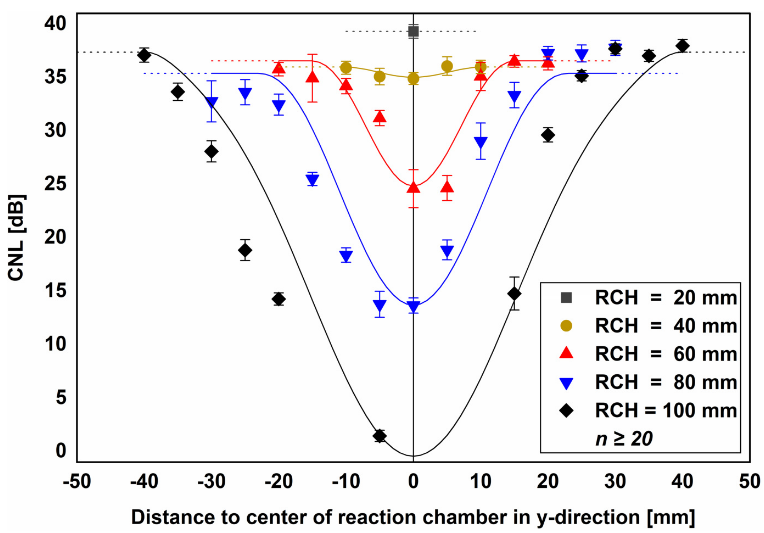

Figure 6.3: Distribution of cavitation noise levels (CNLs) along the y_0-axes for reaction chamber heights (RCHs) from 20 - 100 mm. Markers denote average values of $n \geq 20$ measurements, while error bars indicate the standard deviation. Solid lines represent polynomial approximations of the measurement values based on the mirrored measurement data, whereas dotted lines represent constant linear extrapolations of the outmost measurement values.

For all other RCHs, on the contrary, sound wave attenuation seemed to play a critical role, and cavitation noise strongly decreased with advancing distance from the transducer plates. For instance, CNLs declined by 34% and 63% from transducer plate vicinity to the center of the reactor for RCHs of 60 mm and 80 mm, respectively, and even dropped below the detection limit at several points in the 100 mm gap. The observations are in line with previous studies, similarly reporting a sharp decline of CNL with advancing distance to the US transducers [77, 125].

Such pronounced drops must be regarded as critical since CNLs below 26 dB indicate already an undershooting of the cavitation threshold (according to the pre-standard DIN SPEC 40170 [157] and the manufacturer of the hydrophone). While this threshold refers to water so that a direct comparison is difficult, it nevertheless suggests a substantial attenuation of cavitational activity in the larger reaction chambers.

In order to derive continuous functions from the discrete CNL measurements for the inclusion in the CFD model, polynomials were fitted to the experimental data, as indicated by solid lines in Figure 6.3. The statistical analysis indicated a very good fit between the polynomial functions and the symmetrized measurement data, with R^2 values > 0.95, regression line slopes $m > 0.93$, and CV(RMSE) values $< 3\%$. The fit between non-symmetrized data and polynomials was naturally not equally good (R^2 between 0.57 and 0.86, m between 0.56 and 0.85, CV(RMSE) between 0.91% and 17.9%) due to the measurement values´ slight deviation from axial symmetry. However, by accounting for both sides, the symmetric polynomials were accepted as good model approximations for the measured CNL values.

Possible reasons for the slight deviation from axial symmetry could be a slightly different performance between the two plate reactors, a slightly varying sensitivity of the radially sensitive hydrophone (possibly due to its fixation), or small, treatment-related changes in sludge temperature and viscosity during the measurement. At the same time, a certain deviation from axial symmetry was already expected due to the highly dynamic nature of cavitation fields [106].

6.3.2 Analysis of the secondary effects of sonication

In contrast to cavitation noise (which reaches its ultimate value immediately after switching on the US reactor), secondary effects depend on both cavitational activity and exposure time. Thus, secondary treatment conditions at a given location could not be characterized by CNL alone but required the consideration of the varying exposure times. This characterization was conducted by coupling the two factors using an auxiliary exposition index (termed *exposure to acoustic cavitation*, EAC), defined as the product of CNL and exposure time t_e (in [dB·s]):

$$EAC = CNL \cdot t_e \tag{6.10}$$

By this definition, the proposed index is closely aligned to well-established expressions of ultrasonic energy such as specific energy input or ultrasonic dose, which similarly multiply treatment intensity (as electric power in kW) by treatment duration (in s) [12].

6.3.2.1 Aluminum foil tests

Photographs of the sonicated sheets of aluminum foil are displayed in Figure 6.4. Overall, it can be seen that sonication caused a highly visible erosion of the foil, which indicates a strong, mechanically effective cavitation field. As the erosion patterns are relatively homogeneous within the individual sheets, the previously observed uniformity of cavitation noise in x- and z-directions is substantiated by the aluminum foil test.

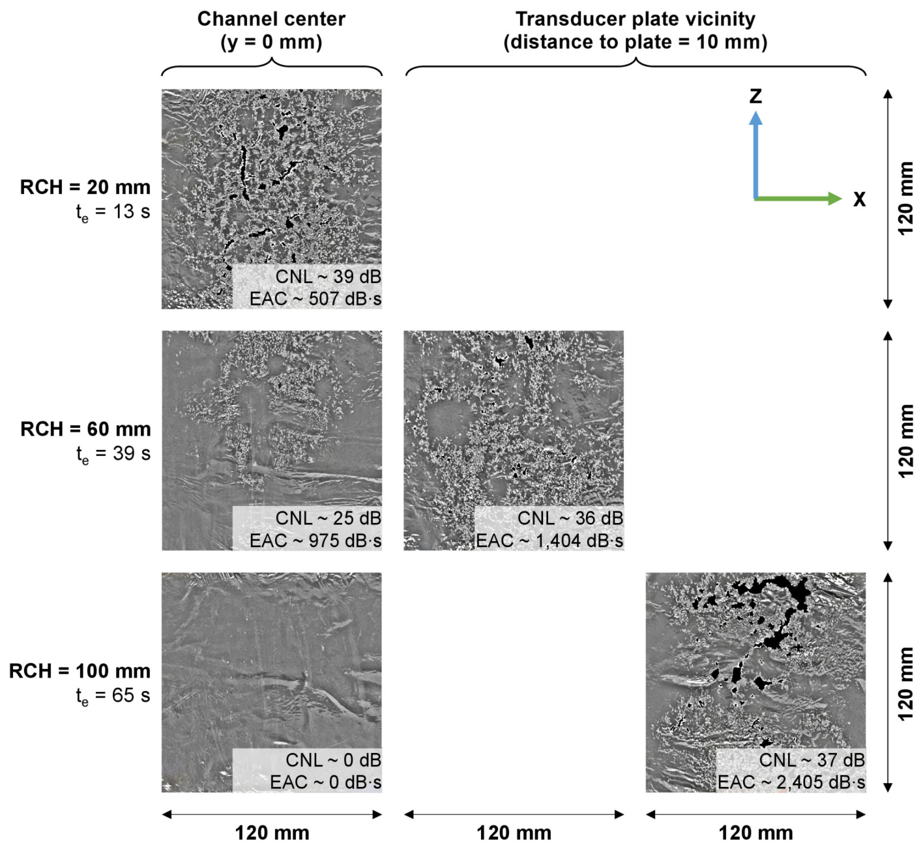

Figure 6.4: Photographs of sonicated aluminum foils at a constant energy input of ~ 200 kJ/kg_{TS} at different locations within the reactor (centrally positioned and positioned 10 mm apart from the transducer plate) for three different reaction chamber heights (RCHs). For RCH = 20 mm, the two locations are identical. Cavitation noise levels (CNLs), exposure times (t_e) and exposition indices (EAC = CNL·t_e) are shown next to the photographs.

Generally, the sheets in the center show less damage than sheets placed close to the transducer plates, which relates well to the previous CNL measurements. For an RCH of 60 mm, the difference between the two locations is already visible, while it is most distinct for an RCH of 100 mm, where a practically unaffected sheet in the center (CNL = 0 dB, EAC = 0 dB·s) is contrasted by a severely perforated sheet in transducer plate vicinity (CNL = 37 dB, EAC = 2,405 dB·s).

Erosion damage in the middle of the reactor decreases with increasing RCH, which is again in good agreement with the hydrophone measurements. Yet, a direct comparison is difficult, due to the different exposure times. Nonetheless, the extreme difference between the strongly perforated sheet at RCH = 20 mm (t_e = 13 s) and the intact sheet at RCH = 100 mm (t_e = 65 s) clearly demonstrates that a lack of cavitation intensity cannot be compensated for by extended exposure time.

The examination of the impact of exposure time can be done best by considering the sheets near the transducers (including the sheet from the 20 mm gap which is also 10 mm apart from the plate), as CNLs are relatively similar (36 - 39 dB), while only exposure times differ. Comparing the 60 mm and 100 mm gaps reveal that the foil sonicated in the larger reaction chamber shows a stronger erosion pattern, indicating that an increase in exposure time (from 39 s to 65 s) increases the degree of mechanical damage. Based on this observation, a coupling of both parameters (as was done for EAC) seems worthwhile (1,404 dB·s at RCH = 60 mm vs. 2,405 dB·s at RCH = 100 mm). On the other hand, the erosion of the sheet placed in the 20 mm gap (39 dB, 13 s) is more severe than the erosion observed at the outer sheet of the 60 mm reactor (36 dB, 39 s). Thus, in this case, the slightly higher CNL appeared to be more influential than the triplication of exposure time. Similar findings were made in previous studies, reporting that intense treatments with short exposure time are more efficient than less powerful treatments with a long exposure time (at constant energy inputs) [77, 97, 146]. This insight already challenges the newly defined index of EAC, which considers exposure time as equally important as CNL. Consequently, the finding also questions the validity of well-established expression of ultrasonic energy (such as specific energy input or ultrasonic dose), which share the same simplification.

6.3.2.2 DD_{COD} analysis

DD_{COD} values for samples taken both at the channel center and directly at the transducer plates are displayed in Figure 6.5. The DD_{COD} at the center significantly decreased with increasing RCH, and the values dropped from 6.4% at RCH = 20 mm to an even slightly negative value at RCH = 100 mm (at $p < 0.01$).

On the other hand, DD_{COD} values remained relatively constant in transducer plate vicinity. This is surprising at first glance, given the increased exposure times at the larger RCHs, and the relatively constant CNLs in transducer surface vicinity (~ 34 - 39 dB). However, as previously observed for the aluminum foil test and based on [77, 97, 146], cavitation intensity seems to have a more significant impact on mechanical disintegration than exposure time. At the same time, it should also be considered that strong cavitational activity promotes local mixing even in sludge [149] so that the extracted samples most likely resemble a mixture from within a certain radius around the sampling location. A sharp peak as that of a spatially precisely

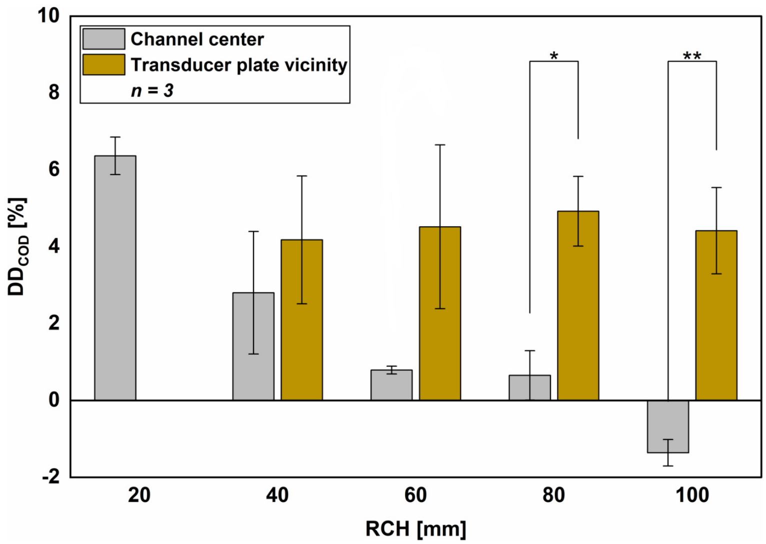

Figure 6.5: Degree of disintegration (DD_{COD}) for samples taken at the channel center and in transducer plate vicinity for all tested reaction chamber heights (RCHs) at a constant energy input of ~ 200 kJ/kg_{TS}. Error bars denote the standard deviation. Significant differences between inner and outer measurement locations are shown using asterisks (* = significant at $p < 0.05$, ** = highly significant at $p < 0.01$).

fixated measure is therefore not to be expected.

Despite the potential mixing on a local scale, significant differences in DD_{COD} were observed between the inner and the outer measurement locations. Generally, the larger the RCH, the more distinct this difference became and for RCHs of 80 mm, and 100 mm, COD solubilization in the channel center was significantly lower than in transducer plate vicinity (at $p < 0.02$ and $p < 0.01$, respectively). Thus, the result agrees with both CNL measurements and aluminum foil tests and confirms that sludge is not turbulently mixed during sonication on a larger scale [149]. The comparably low COD solubilization (maximum $DD_{COD} = 6.4\%$) is due to the selected low specific energy input of only 200 kJ/kg_{TS}. The result is in line with a previous study reporting a DD_{COD} between 0.9% and 4.6% for the same energy input [133].

In order to check for potential correlations between CNL and DD_{COD} and between EAC and DD_{COD}, linear regression analyses were performed. The correlation between CNL and DD_{COD} was relatively strong ($R^2 = 0.83$, Figure 6.6), whereas the additional consideration of exposure time using the proposed index of EAC showed no linear correlation ($R^2 = 0.29$, see Figure D.3 in Appendix D). While keeping in mind that the DD_{COD} values from transducer plate vicinity might resemble mixtures to some extent, the result again confirms the prime role of cavitation intensity.

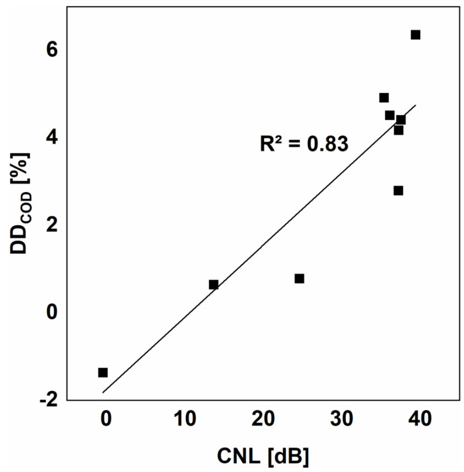

Figure 6.6: Linear regressions between cavitation noise level (CNL) and degree of disintegration (DD_{COD}).

6.3.3 Comparison of batch and flow-through sonication and estimation of treatment (in)homogeneity

Using CFD simulations, the impact of flow on treatment (in)homogeneity was estimated and a comparison of batch and flow-through sonication regarding exposure times is depicted in Figure 6.7A. The exposure times for flow-through sonication were calculated as the reciprocal of the simulated velocity vectors u_z times the length of the evaluation domain ($t_e = 1/u_z \cdot 225$ mm), which was feasible due to the laminar flow model.

The exposure time distribution featured relatively broad peaks, suggesting that the high viscosity of the sludge imposed a plug-flow. Despite this relatively uniform flow profile, exposure times for sludge particles in the channel center were still considerably shorter than for particles traveling in wall vicinity. Thus, the flow adds a certain inhomogeneity in exposure time as compared to batch sonication. A more detailed depiction of the flow including velocity profiles can be seen in Appendix D, Figure D.4.

For a coupling of the modeled exposure time patterns with the recorded cavitation intensity (according to the proposed index of EAC), the 1-D CNL polynomials needed to be extrapolated over the 2-D cross-section. Such constant extrapolation was deemed admissible, as CNL was shown to be relatively independent of x- and z-positions (see Figure 6.2). The extrapolation was moreover deemed feasible for both batch and flow-through sonication, as low flow velocities (such as the ones tested) were shown to have only negligible impact on CNL [158].

The extrapolated CNLs, as well as the coupling of both factors using the index of EAC are displayed in Figures 6.7B and 6.7C, respectively. The EAC plots highlight that for batch

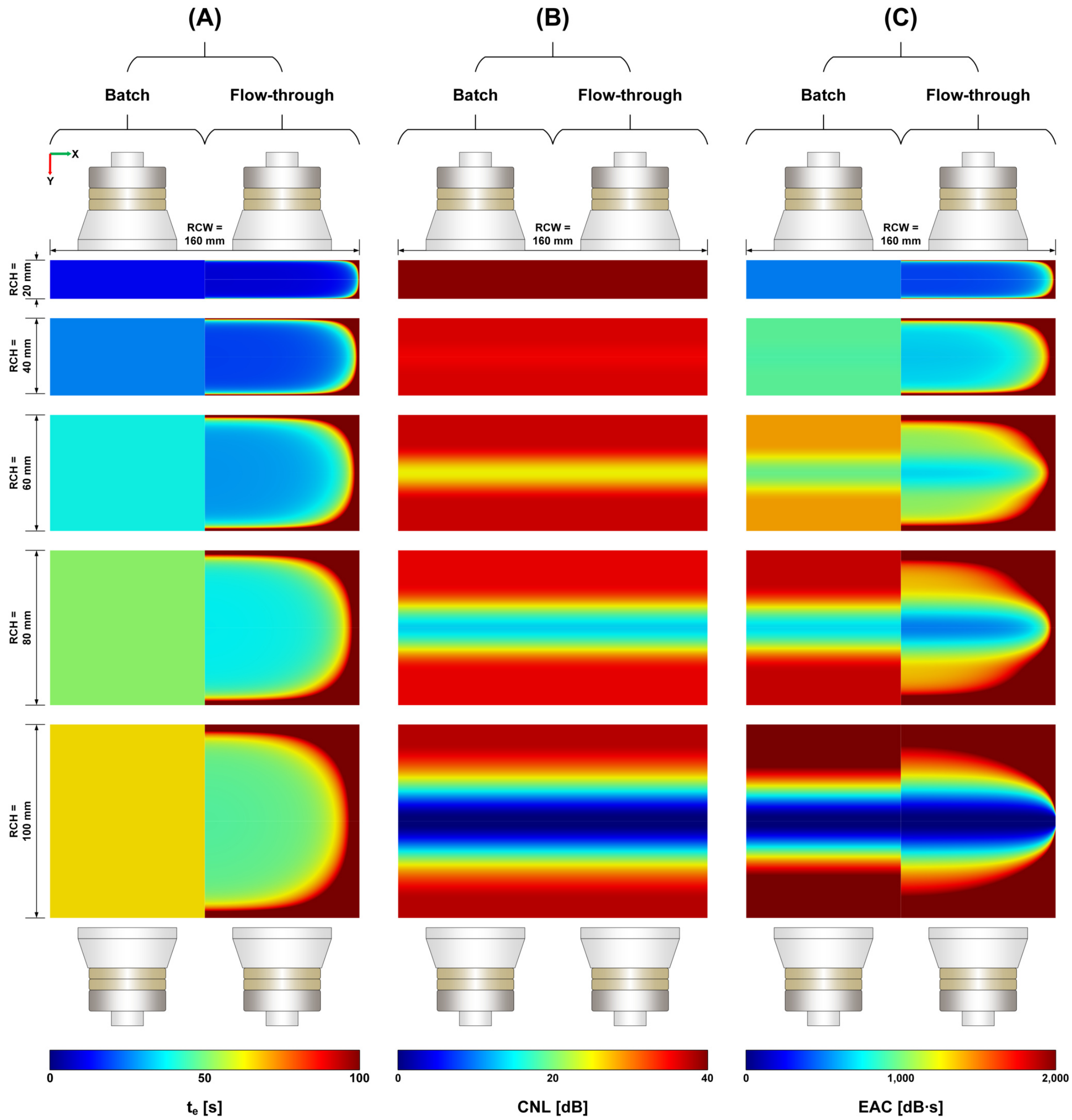

Figure 6.7: Distribution of exposure times (t_e, subfigure A), cavitation noise levels (CNLs, subfigure B), and exposition indices (EAC = CNL·t_e, subfigure C) for all reaction chamber heights (RCHs), with RCW = reaction chamber width. The left sides of the plots resemble batch sonications, while the right sides resemble flow-through sonications. Computed exposure times t_e refer to the residence time within the evaluation domain at a flow rate of 200 L/h. Plots of CNL resemble a constant extrapolation of the polynomial approximations in x-direction. To ensure good visual comparability between the different plots, the scales of both t_e and EAC were cut at 100 s and 2,000 dB·s, respectively.

sonication, treatment (in)homogeneity was only governed by the distribution of cavitation intensity, while for flow-through sonication, the non-uniformity of CNL was further aggravated by the drop of exposure times towards the channel center. EAC values are highest in wall vicinity due to the elevated CNLs and the prolonged exposure times and lowest in the center of the channel, for the opposite reasons. The EAC plots for RCHs of 20 mm and 40 mm moreover insinuate that even treatments with a constant CNL may not lead to an entirely homogeneous exposition during flow-through sonication, due to the varying exposure times.

Exposition (in)homogeneity was further illustrated using boxplots for both batch and flow-through sonication in Figure 6.8. The plots demonstrate that the range of both CNL and EAC values keeps increasing with RCH and that flow-through sonication imposes a larger disparity of treatment intensities than batch sonication. For instance, the interquartile range for the batch treatment at RCH = 20 mm was zero, as the entire sample was uniformly exposed to CNL and EAC values of approximately 39 dB and 510 dB·s, respectively. Also, for flow-through sonication, a high level of uniformity was preserved thanks to the non-attenuated CNLs, and the spread of EAC values remained fairly low (∼100 dB·s).

At an intermediate RCH of 60 mm, on the other hand, a large portion of the substrate was already subjected to a severely weakened cavitation field, and 32% and 39% of the substrate remained below a CNL of 26 dB (cavitation threshold in water) for batch and flow-through sonication, respectively. At the largest spacing of 100 mm, the fractions of poorly exposed sludge increased further to 47% for batch treatment and 57% for flow-through sonication. When aiming for a controllable treatment, however, such overly inhomogeneous exposition seems highly unfavorable, as different treatment intensities were shown to cause very different effects (for instance, de-agglomeration of flocs at low intensities, and cell lysis at high intensities [56, 81]).

At the same time, the interpretation of both Figures 6.7 and 6.8 needs to be conducted in the light of the applied model approximations. The complete omission of cavitation-induced mixing certainly overestimates treatment inhomogeneity, as the minor, nonetheless, present cavitation-induced mixing will contribute to a slight homogenization of the treatment [149]. Also, the constant extrapolation of CNLs in x- and z-direction might be an oversimplification. However, a qualitative exploitation of the results seems fully justified due to the clear tendencies among the different RCHs and between batch and flow-through sonication.

6.3.4 Impact of RCH on overall disintegration efficiency

For determining the overall disintegration efficiency, the DD_{COD} of the mixed reactor content was analyzed after each sonication, and the obtained results are shown in Figure 6.9A.

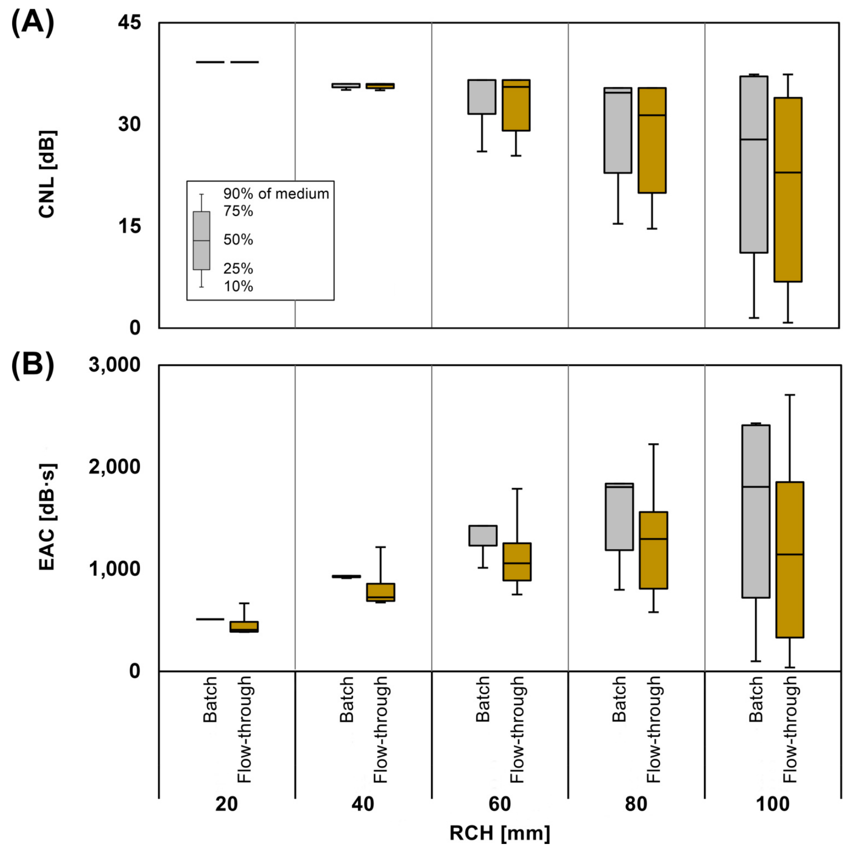

Figure 6.8: Distribution of cavitation noise levels (CNLs, subfigure A) and exposition indices ($EAC = CNL{\cdot}t_e$, subfigure B) over the treated sludge for batch and flow-through sonication, for all reaction chamber heights (RCHs). The lower and upper ends of the boxes denote the 25th, and 75th percentiles, respectively, while the band in the box depicts the 50th percentile. The lower and upper whiskers denote the 10th, and the 90th percentiles, respectively.

DD_{COD} values showed a constant decline with increasing RCHs, thus demonstrating that average disintegration efficiency was dependent on reactor design. From RCH = 20 mm to RCH = 100 mm, the drop in DD_{COD} was even significant ($p < 0.05$) and amounted to almost 73%. Considering that specific energy inputs (and thus, electricity consumption and treatment costs per unit volume of sludge) were equal for all RCHs, the distinct difference in DD_{COD} demonstrates that reactor geometry has a considerable impact on treatment economics.

When revisiting the equation of specific energy input (see Eq. 6.1) in search for an explanation for the difference, it becomes highly visible that the electric power P resembles only a surrogate parameter. Ultimately, the targeted disintegration is not facilitated by electric power, but by cavitational activity. While electric power is independent of the reactor geometry, the previous investigations demonstrated that cavitation activity is not. At the same time, all

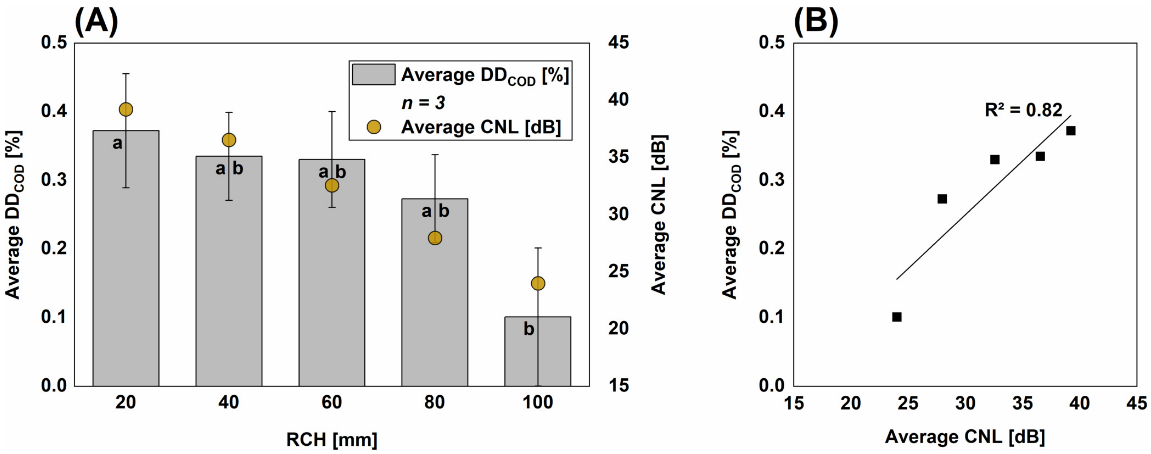

Figure 6.9: Average degree of disintegration (DD_{COD}) and average cavitation noise level (CNL) for all reaction chamber heights (RCHs) tested (subfigure A) and linear regression analysis of both parameters (subfigure B). Error bars denote the standard deviation, and letters a and b indicate significance levels. Data sets without a common letter are significantly different at $p < 0.05$.

other variables in the equation remain unaffected by a variation in reaction chamber spacing. Both the density and the TS concentration of the sludge are, of course, independent of reactor design. Also, the quotient of t/V needs to remain constant for a meaningful comparison of different reactor designs (as in the present study). Thus, it seems that considering cavitational activity instead of electric power offers a good possibility to explain the difference in average DD_{COD} values.

Therefore, average CNLs of each RCH were plotted against the average DD_{COD} (Figure 6.9A), while a linear regression analysis between the two variables is displayed in Figure 6.9B. The regression indicates a relatively good fit ($R^2 = 0.82$), which suggests that cavitation noise could be a useful parameter for evaluating the disintegration performance of ultrasonic reactor designs for sewage sludge pre-treatment. It needs to be stated, however, that the outcome of only one experiment with 5 data points is assuredly not enough to draw definite conclusions or to establish $CNL_{average}$ as an index to characterize US pre-treatments. However, as to date no expression of ultrasonic energy is able to relate reactor design and disintegration efficiency, a further investigation of the observed correlation seems worthwhile.

6.3.5 Implications on full-scale applications

Concerning full-scale applicability, the finding that small RCHs are most suitable for WAS pre-treatment may conflict with the operational requirements at WWTPs. To avoid high pressure losses and blockages, WAS pipes typically have relatively large diameters. Thus, an RCH as small as 20 mm seems not well suited for an application in a WWTP, despite its disintegration performance. It should also be noted that the WAS sample investigated had a comparatively low TS concentration (4.4% - 4.6%), while the solid fraction of thickened WAS samples could easily reach values of over 6% [36, 105, 133]. At such TS ranges, however, the conflict between operational stability and sonication efficiency might even be sharper.

Thus, sewage sludges with lower TS content (for instance, digested sludge with typical solids contents of 3% [159]) may resemble a more suitable substrate for ultrasonic flatbed reactors. A possible application could be co- or inter-treatment, where digested sludge is either sonicated in the recirculation line used for digester mixing and heating or when passing from a first to a second digester (in two-stage systems) [66]. Such suggestion is substantiated by previous studies reporting that digested sludge was more amenable to sonication in flatbed reactors than WAS [43, 145].

The observed increase in treatment inhomogeneity due to flow-through operation is deemed difficult to avoid in full-scale applications when flow-through reactors are to be applied. A potential solution could be treatment in series with flow-disturbing baffles between the reactors. However, the advantage of such additional mixing needs to be balanced against a potential clogging risk. Promoting cavitation-induced mixing by increasing flow velocities, on the other hand, seems impractical, as massive flow rates would be required to shift WAS flow to a transitional range [139]. At such flow rates, however, specific energy inputs would most likely drop below effective levels [149]. Thus, treatment of less mixing-resistant substrates such as digested sludge might again be a targeting strategy to reduce exposition inhomogeneity.

The considerable differences among the tested reaction chambers demonstrated that conventionally applied expressions of ultrasonic energy, such as specific energy input, offer only an approximate characterization of US treatments. For a more effect-based description of the treatment, the assessment of cavitation noise and the index of EAC were proposed. No linear correlation was observed between EAC and DD_{COD} values, indicating that cavitation intensity is more influential than exposure time concerning secondary effects of cavitation. Accordingly, the correlation between CNL and DD_{COD} was more distinct, and especially the observed link between average CNLs and average DD_{COD} values should get further attention. Assuming the correlation can be reproduced in subsequent research, hydrophone measurements could be a promising, easy-to-use, and comparably cheap tool to systematically optimize reactor designs for a wide variety of substrates.

6.4 Conclusion

The impact of US reactor design on treatment (in)homogeneity and overall disintegration efficiency was systematically investigated using a flatbed reactor of variable RCH (20 - 100 mm). The effects of sonication were examined using three independent measurement techniques, including hydrophone measurements, aluminum foil tests, and soluble COD analyses. The results of all measurement techniques suggest that treatments can either result in an intense and uniform exposition of the sludge towards sonication (at small RCHs), or in an inhomogeneous exposition where a substantial part of the substrate is not exposed to cavitation anymore (at large RCHs). Results of the flow simulations furthermore suggest that US treatments in flow-through reactors are more inhomogeneous than sonications in batch mode due to the overlapping effect of the laminar flow regime. The overall disintegration efficiency declined significantly with increasing RCHs (average DD_{COD} dropped by up to 73%), which was shown to correlate well with receding average CNLs in larger chamber spacings. Therefore, hydrophone measurements might be a cheap and easy-to-use tool to optimize US reactor designs for sewage sludge pre-treatment. The presented findings suggest that equal specific energy inputs do not imply equal treatments and that reactor geometry might have a defining impact on the efficiency and the economics of US pre-treatments.

6.5 Acknowledgments

This work was supported by the German Federal Ministry for Economic Affairs and Energy (BMWi), Grant 03ET1396B. The company BANDELIN electronic GmbH & Co. KG is kindly acknowledged for providing the size-adjustable US reactor. Special thanks go to Ramona Schütt, Myriam Reif, and Nadine Stellwag for the excellent assistance in the lab and to Leonie Daxl for the artwork in the graphical abstract. Further thanks go to the staff members of the Freising WWTP for their outstanding support.

6.6 Appendix - Supplementary material

The graphical abstract and supplementary material to this article can be found in Appendix D of this thesis.

CHAPTER 7

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

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Author contributions: Thomas Lippert and Jochen Bandelin were responsible for the research plan and the manuscript preparation. Thomas Lippert conducted the data analysis. Jochen Bandelin developed the full-scale ultrasound reactor and was responsible for the implementation of the reactor system and its automation at the Starnberg wastewater treatment plant. Sampling and laboratory analyses were performed by Dominik Vogl, Zahra Alipour Tesieh, and Thomas Lippert. Thomas Wild coordinated the full-scale experiment on-site and was responsible for synchronizing the feeding regimes of the two full-scale digesters and for frequently checking the operation of the ultrasound reactor. Jörg E. Drewes and Konrad Koch supervised the study and reviewed the manuscript.

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 rather typical operating conditions was selected. The effects of the treatment (conducted at a specific energy input of 200 kJ per kg of total solids) were monitored regarding improvements in sludge viscosity, methane production, biosolids removal, and 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 yield (+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 negligible, as the reduced pumping costs were less than 1‰ of the US reactor's electricity costs. Additional methane yields enabled for partial cost recovery (roughly equivalent to the energy costs), while investment costs of the US equipment could not be regained. Yet, the incorporation of improved methane production in the economic assessment remains somewhat speculative, given the only insignificant increase. Results imply that ultrasonic WAS pre-treatment is uneconomical for typical, well-performing WWTPs.

7.1 Introduction

Ultrasound (US) can be utilized in wastewater treatment as a mechanical pre-treatment method for sewage sludge disintegration [12, 60]. Hereby, the phenomenon of transient acoustic cavitation is utilized, which is defined as the successive formation, growth, and implosion of acoustic micro-bubbles in the sludge [47]. Through the severe violence of the bubble implosions (creating hot spots with temperatures of 4,000 - 15,000 K [54] and pressures of up to 1.3 GPa [55]), 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) [56, 89, 160]. In consequence, the initial and often rate-limiting hydrolysis step of anaerobic sludge digestion can be accelerated, leading to faster reaction kinetics, enhanced methane production, and increased organic matter degradation [4, 10, 19]. Further reported merits are reduced sludge viscosity and improved digestate dewaterability [33, 161]. Besides enhancing anaerobic digestion performance through pre-treatment, US can also be applied for microbial community regulation (for instance, for enhancing ammonia-oxidizing bacteria while selectively eliminating nitrite-oxidizing bacteria [162, 163]) or as post-treatment for improved digestate dewaterability [164, 165].

Given the various benefits and applications, a large body of research was devoted to advance ultrasonic pre-treatment technologies further [4, 10, 12, 19, 162–165]. However, most hitherto studies were conducted under laboratory conditions, which can strongly differ from the conditions at full-scale wastewater treatment plants (WWTPs). For instance, specific energy inputs feasible at full-scale WWTPs are usually quite low and rarely exceed

1,000 kJ per kg of total solids (TS) due to the large sludge volumes to be treated and accordingly short treatment times in the US reactors (such as seconds or minutes at best) [45, 84, 85]. In lab-scale studies, on the other hand, specific energy inputs typically range from 1,000 - 16,000 kJ/kg_{TS} [10, 19], or can even reach values of 27,000 kJ/kg_{TS} [36] or 108,000 kJ/kg_{TS} [80]. As treatment effects typically improve with increasing energy input [12, 166], the transferability of lab-scale studies to full-scale applications is, hence, limited. A further difference relates to the 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, as most full-scale digesters are designed as continuous systems [11], the transferability of lab-scale BMP tests to full-scale digesters is again limited [93, 104]. Last but not least, the 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 testing at full-scale WWTPs.

Despite the research need for full-scale trials, only a few studies are available to date. One of the best documented studies was performed by Xie et al. [84], who pre-treated raw sludge (RS) with an average TS content of 1.6% (comprising primary sludge, PS, and waste activated sludge, WAS) using a 20 kHz radial horn reactor at the Ulu Pandan Water Reclamation Plant in Singapore. Despite a low energy input of below 400 kJ/kg_{TS} (calculated based on the published data) and a comparably long hydraulic retention time (HRT) of 30 d of the sludge in the digester (potentially canceling out kinetic effects of the treatment [10]), an increase in methane yield by 13% - 58% relative to a control digester was observed. Similarly promising results were obtained by Hogan et al. [111], who reported an increase in biogas yield by up to 50% and a slight, but still significant improvement of digestate dewaterability (TS of filter cake increased by 1.6 percentage points on average), at a full-scale test at the Orange County Sanitation District, USA. Another full-scale trial was performed by Neis [45] at the Bamberg WWTP in Germany, where two sonotrode reactors (5 kW each) were installed for side-stream WAS pre-treatment (80% treated at an energy input of 148 kJ/kg_{TS}) to cope with a shortened HRT of only 18 d. During the full-scale trial conducted from 2004 to 2011, biogas production and volatile solids (VS) degradation increased from about 1.5 million m^3/a to 2.2 million m^3/a, and from 34% to 50%, respectively, thus enabling stable digestion despite the shortened HRT. However, as no control digester was used, it was difficult to ascertain that the positive development was solely related to the US pre-treatment.

Contrasting such positive outcomes, a report from the Swedish Water and Wastewater Association stated that ultrasonic WAS pre-treatment at the Gässlösa WWTP in Sweden had only a negligible effect on biogas production or organic matter degradation [107]. A second full-scale trial at the Swedish Ernemar WWTP did not result in noticeable anaerobic digestion enhancement, either, as repeated malfunctions of the US reactor prohibited reliable test conditions [107]. Operational challenges were also experienced by Oon et al. [46], who stated that the radial horn reactor employed in the full-scale test at the Ulu Pandan Water Reclamation Plant (Singapore) exhibited considerable clogging susceptibility, which necessitated manual cleaning cycles every second day. Besides, the lifetime of the radial horns was rather short due to pronounced cavitation erosion and amounted to less than 2,000 h (or 83 d in 24/7 operation). Neis [45] confirmed the high erosion susceptibility and reported that the sonotrodes used at the full-scale test at the Bamberg WWTP (Germany) required annual replacement.

In addition to the somewhat miscellaneous (operational) experiences, 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 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 of currently available information on full-scale sonication tests is difficult, which has also been observed for other pre-treatment methods such as thermal hydrolysis [167, 168].

Thus, to obtain a reliable assessment whether US pre-treatment enhances 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 a control and one used as a 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 well-performing WWTP with rather typical operating conditions was selected. For assessing treatment economics, the investment and electricity costs of the US reactor were balanced against potential benefits through decreased sludge viscosity, additional methane production, improved biosolids degradation, or better digestate dewaterability.

7.2 Materials and methods

7.2.1 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 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 [45, 84]). The goal of selecting such typical, or even challenging plant was to obtain results that are also transferable to other WWTPs (according to the so-called New-York-principle: *"If I can make it there, I´ll make it anywhere"*). Further selection criteria were (i) the availability of two digesters operated in parallel for a setup comprising a control and a test digester, (ii) a high degree of controllability of the plant (especially regarding feeding regimes), and (iii) dedicated 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 7.1.

The comparison demonstrates that the Starnberg WWTP features rather representative characteristics regarding design capacity, digester volume, and feed sludge characteristics (TS of WAS = 6.3% vs. a median value of 6.7%, TS of PS = 3.8% vs. a median value of 3.9%). Especially typical WAS characteristics were deemed important for the full-scale test, as the solids content of WAS directly affects sonication performance [14, 125]. A larger deviation, on the other hand, was observed for the HRT, which amounted to 42 d on average as compared to 32 d for the median WWTP from the field survey. It should be noted that the HRT at the Starnberg WWTP exhibited a certain deviation (ranging from 39 d to 45 d), due to the slightly fluctuating sludge production (see Table 7.1). Such increased HRT of over 40 d represents a somewhat challenging boundary condition, 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. Regarding the share of WAS to RS, the Starnberg WWTP also shows a certain deviation from the other plants surveyed (= 33% vs. a median value of 43%, based on VS_{fed}). Yet, as current sludge minimization measures are designed to significantly reduce WAS production [169, 170], a comparably small share of WAS to RS may be a likely scenario for future WWTPs. Moreover, the survey demonstrated that the share of WAS to RS is generally well below 50% (except for one plant), thus emphasizing that the condition at the Starnberg WWTP is not categorically different to the other plants. Full information on the operational characteristics of the 17 plants examined is given in Appendix E (Table E.1).

Table 7.1: Average operating characteristics of the Starnberg wastewater treatment plant (WWTP) and median values of the survey among 17 WWTPs in southern Germany.

Parameter	Starnberg WWTP	Median of the 17 WWTPs
Design capacity [PE]	100,000	100,000
Daily PS production [m^3]	89 ± 19.1	108
Daily WAS production [m^3]	31 ± 8.0	59
TS of PS [% of FM]	3.8 ± 0.6	3.9
VS of PS [% of TS]	86 ± 3.1	80
TS of WAS [% of FM]	6.3 ± 0.4	6.7
VS of WAS [% of TS]	74 ± 1.3	73
Digester volume [m^3]	2 · 2,500	5,000
Digester temperature [°C]	37 ± 0.7	38
Average HRT in the digester [d]	42 ± 2.5	32
Share of WAS relative to RS [% of VS_{fed}]	33 ± 4.0	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.

7.2.2 Digester operation and experimental schedule

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 (conducted 24 h/d) was alternated every two hours, i.e., after the control digester was fed for 2 h, feeding changed to the test digester for the next 2 h and so on. Feeding with thickened WAS was conducted for 8 h/d, while the control and the test digester were fed for 2 times 2 h each, respectively.

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 d) without US pre-treatment. After completing the start-up, the experiment was started, and the US reactors were activated for 4 h per day (during WAS feeding of the test digester). The test phase of the experiment was conducted for seven consecutive HRTs (or 292 d), thus fully complying with the suggestion of the German technical guideline 4630 to monitor changes to continuous digesters for at least three to five HRTs [128].

7.2.3 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, donut-shaped 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 sound wave attenuation and cavitationally inactive zones), according to previous reactor design optimizations for sewage sludge treatment [125, 171]. 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 of reactor volume. A 3-D schematic of the novel reactor system is depicted in Figure 7.1A.

For the full-scale trial, a total of three double-tube reactors with a tallied 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 Figures 7.1B and 7.1C, respectively.

The reactors 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 (Figure 7.1C). 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. The resulting specific energy input amounted to approximately 200 kJ/kg_{TS}, which is similar to previous full-scale trials [45, 84] and has been chosen in accordance to previous lab-scale studies, revealing that low energy inputs (for instance, 200 kJ/kg_{TS}) lead to the same methane yield enhancement as high energy inputs (for instance, 3,000 kJ/kg_{TS}) [29, 133, 145]. At intermediate inputs (for instance, 400 kJ/kg_{TS} - 1,000 kJ/kg_{TS}), on the other hand, a so-called

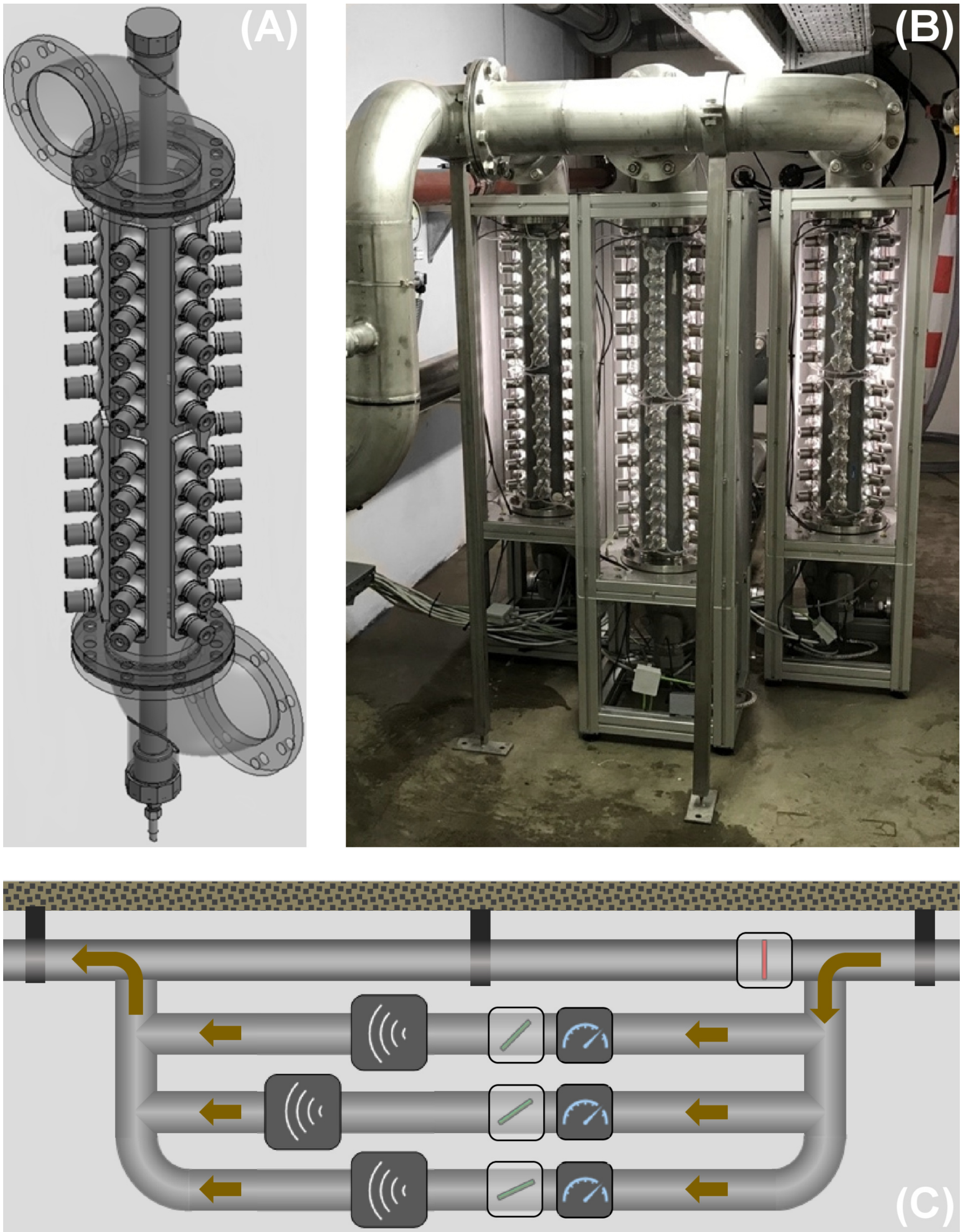

Figure 7.1: 3-D schematic of the employed double-tube reactor (A), photograph of the whole reactor setup (B), and top view assembly plan of the reactor installation (C). The closed valve in the main waste activated sludge (WAS) pipe is indicated by a red bar, while the partially opened valves in the inlet pipes to the ultrasound reactors are indicated by green bars. WAS flow is indicated by brown arrows.

'performance gap' was detected, which did not allow for significantly enhanced methane yields [133, 145]. Thus, to take advantage of the high performance of low energy inputs and to avoid uneconomic intermediate ranges, sonication was only performed at an energy input of 200 kJ/kg_{TS}.

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 d. To nonetheless ensure safe reactor operation and 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 m^3/h or the temperature rose above 70°C at any point in the setup, US reactors were automatically deactivated.

7.2.4 Monitoring of treatment effects and digester performance

7.2.4.1 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 the ESDU 91025 technical report for predicting frictional pressure loss [172]:

$$\Delta h = \left(\frac{4LK}{D} \left[\frac{8Q}{\pi D^3} \frac{3n+1}{n} \right]^n \right) \cdot \frac{1}{10} \tag{7.1}$$

where Δh is the head loss [m], L is the length of the WAS pipe [= 45 m], *K* is the consistency index [$Pa \cdot s^n$], D is the inner pipe diameter [= 0.15 m], Q is the flow rate [= 0.001 m^3/s, or 3.75 m^3/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 using the least squares method according to Eshtiaghi et al. [173] and Lippert et al. [149]. Calculations of frictional head loss were cross-checked using an online calculator available at the CheCalc website for chemical engineering calculations ("https://checalc.com").

The pump energy savings due to the reduced head loss were calculated according to Eq. 7.2:

$$E_s = tQ\rho g\Delta h\frac{1}{\eta} \tag{7.2}$$

where E_s is the saved shaft energy [kWh], t is the operating time [h], ρ is the sludge density [= 1,000 kg/m^3], g is the acceleration of gravity [= 9.81 m/s^2], Δh is the head loss [m], and η is the assumed pump efficiency [= 0.6].

7.2.4.2 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 15 minutes.

7.2.4.3 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 [126].

7.2.4.4 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 [174].

7.2.5 Statistical analysis

According to the decision diagram for statistical tests in anaerobic digestion experiments [175], 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 [175].

7.3 Results and discussion

7.3.1 Operational aspects

Despite the omission of cleaning and backwash, no clogging events were encountered throughout the entire experimental phase of 374 d. 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 should moreover be noted that the equal distribution of sludge flow over the three parallel pipes was somewhat challenging, despite the automated electronic valves. To maintain equal flow rates, a frequent adaptation of the valve position was necessary (every few minutes), which would presumably lead to wear of the valves over a longer operating time. It was furthermore shown by Bandelin et al. [158] that increased flow velocity in serially arranged 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.

7.3.2 Effects of the pre-treatment on sludge viscosity

The apparent viscosity of WAS before and after the pre-treatment is displayed in Figure 7.2 for a selected shear rate of 644 1/s, which 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) amounted to almost 8% and was observed in HRT #6. 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 $p < 0.01$). The same outcome was observed for the other recorded shear rates (data not shown). A discussion on the economic implications of the decreased viscosity is presented in Section 7.3.6.

The achieved reduction in viscosity is in line with previous studies [36, 161], which similarly reported reduced WAS viscosity after US pre-treatment. Yet, the obtained viscosity reduction of around 6% is considerably lower than typically reported decreases in lab-scale studies. For instance, Ruiz-Hernando et al. [36] achieved a reduction in viscosity of over

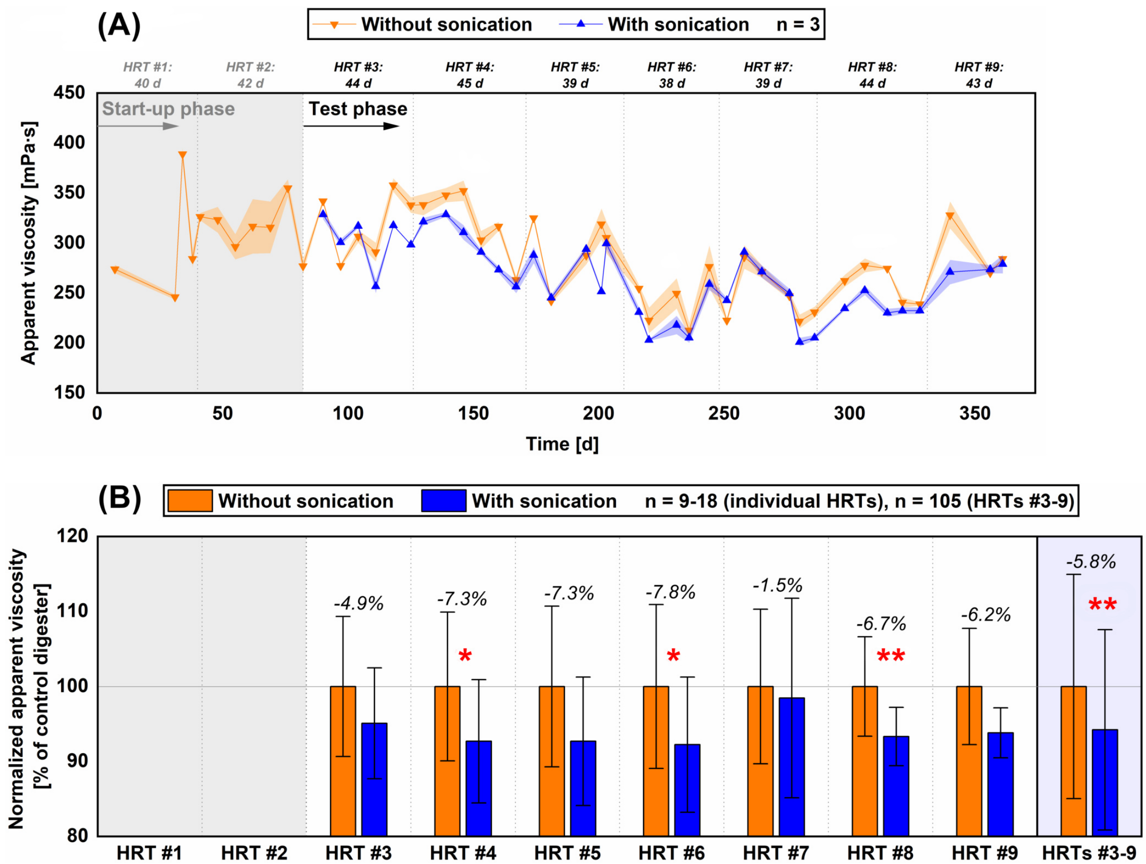

Figure 7.2: Development of apparent viscosity of waste activated sludge with and without sonication at a shear rate of 644 1/s (A) and normalized apparent viscosity relative to the control digester (B). Significant and highly significant differences between two data sets are denoted with one, or two asterisks, respectively. Error bands and error bars denote the standard deviation of the mean.

70%, albeit at 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 enormous 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 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 Figure E.2 in Appendix E). 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

89 m^3 of PS and 31 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.

7.3.3 Effects of the pre-treatment on methane production

The development of the methane production of both digesters is displayed in Figure 7.3. 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 conditions, the test phase could already be initiated after a start-up phase of only two HRTs.

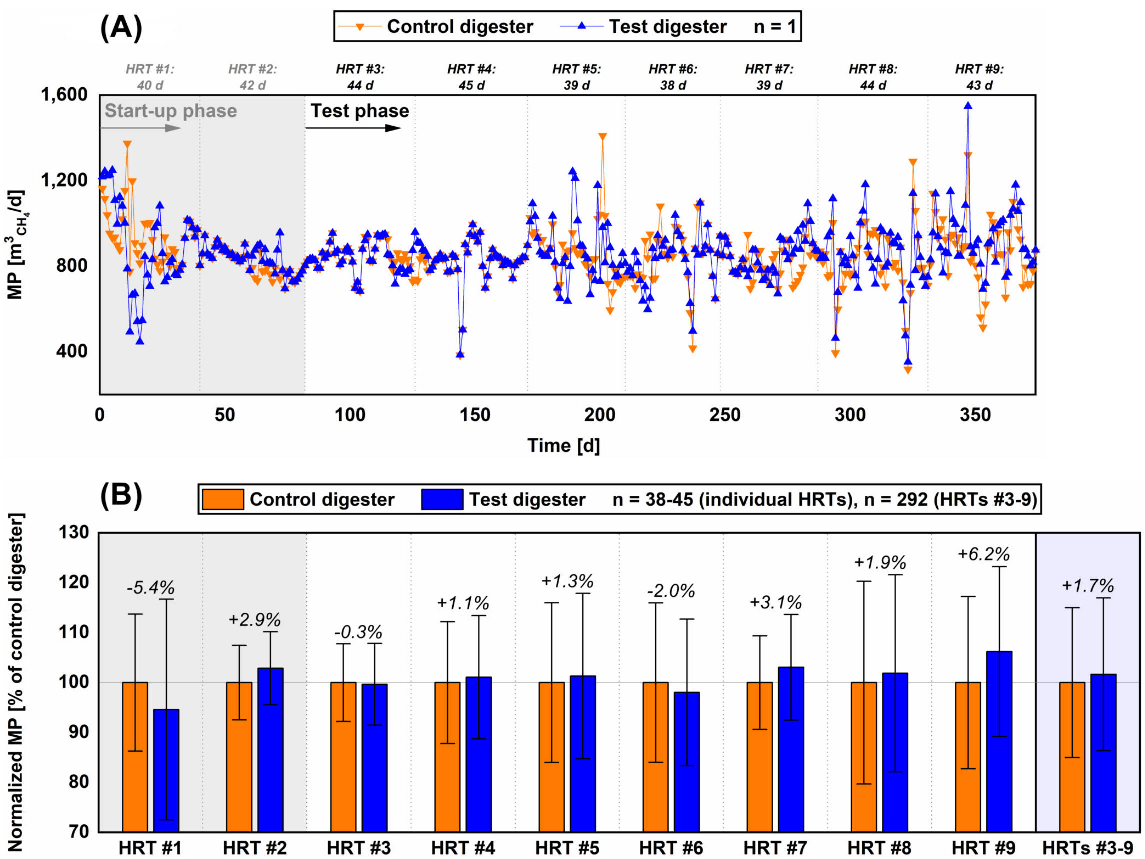

Figure 7.3: Development of daily methane production (MP) of the control and the test digester (A) and normalized MP relative to the control digester (B). Error bars denote the standard deviation of the mean.

The test phase was conducted for a total of seven HRTs (or 292 d). 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 in HRT #9, with an HRT-averaged percentage increase of 6.2% relative to the control digester (at $p < 0.1$).

The finding of 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 [45, 84, 111].

A possible reason for the only moderate enhancement could be the comparably low ultrasonic density of the novel double-tube reactor of 222 W/L, which clearly falls below reported US densities of full-scale sonotrode (345 W/L) and radial horn reactors (1,714 W/L) (Neis, 2015; Xie et al., 2007). Especially in combination with the high TS content of the treated WAS (= 6.3%), the disintegration performance of the double-tube reactor might have been critically compromised. Such hypothesis is substantiated by a recent study by Bandelin et al. [145], who compared the performance of tube and sonotrode reactors for sludge treatment 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). A similar finding was made by Nguyen et al. [42], who reported that laboratory sonotrodes had higher disintegration efficiency than ultrasonic baths at total suspended solids (TSS) contents above 0.9%, while the opposite was observed at TSS concentrations below this mark. Generally, Show et al. [14] demonstrated that disintegration efficiency declines above TS contents of 3%, thus further confirming that the solids content of the present WAS sample (= 6.3%) was far higher than optimal TS concentrations for US treatment. Such performance decline due to elevated solids contents seems a general issue for WAS pre-treatment, as thickened WAS usually exhibits TS concentrations well above 5% (see Table 7.1, Ruiz-Hernando et al. [36], and Suhartini et al. [105]).

Another factor for the only moderate enhancement might be the long HRT of over 40 d at the Starnberg WWTP, which may have rendered kinetic effects of the treatment difficult to detect [10]. On the other hand, Barber [109] reported an increase in biogas production by 45% following US pre-treatment despite an extremely long HRT of 69 d 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 d [45, 84, 111].

Finally, the relatively low share of WAS to RS (= 33%, 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 yield increase of over 16% from WAS only would have been necessary to realize the observed maximum total increase of 6.2%.

7.3.4 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 (Figure 7.4). As for methane production, the solids contents of the two digestates were highly similar during the start-up phase (deviation of less than 1% averaged over HRT #1 and HRT #2), again indicating comparable digestion performance.

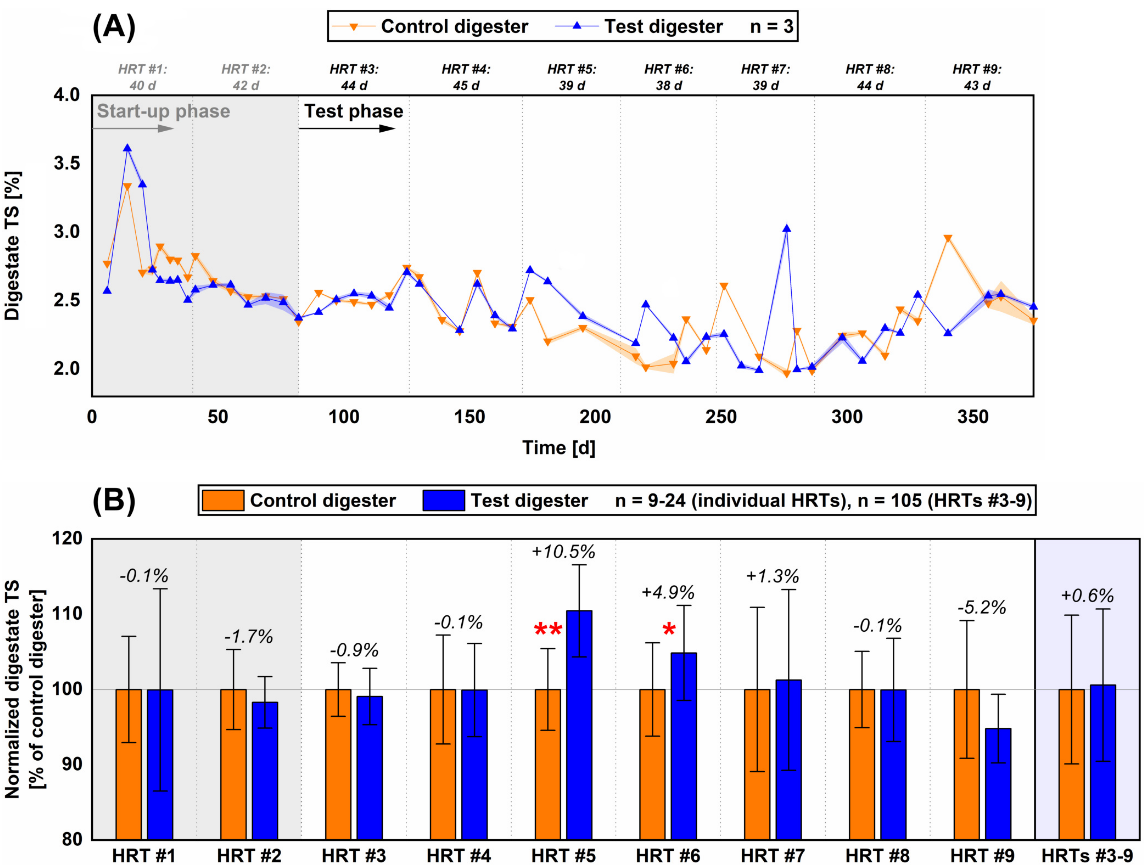

Figure 7.4: Development of the total solids (TS) concentration of the digestate from the control and the test digester (A) and normalized digestate TS relative to the control digester (B). Significant and highly significant differences between two data sets are denoted with one, or two asterisks, respectively. Yet, the significant increase of the test digester´s TS content is a result of a temporal feeding malfunction at the beginning of HRT #5, and not an effect of the ultrasound pre-treatment. Error bands and error bars denote the standard deviation of the mean.

Similar to 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, this increase was not due to the US pre-treatment but most likely due to a short-term feeding malfunction, in which all of the accruing sludge was fed to the test digester for five consecutive working days at the beginning of HRT #5. The erroneous feeding was balanced in the subsequent week, by solely charging the control digester for five days. Yet, a similarly increased TS content of the control digester was not observed, possibly due to its previous starvation. The effect of the two irregular feeding events can also be detected for methane production in Figure 7.3, with the most pronounced peaks at day 189 (test digester) and day 201 (control digester). After digester feeding was balanced, feeding regimes were set back to normal.

Overall, results suggest that savings due to reduced amounts of residual biosolids for disposal are not to be expected. The finding is again in contradiction to the majority of the previous full-scale trials [45, 84, 111], while it confirms the results of the Swedish study at the Gässlösa WWTP [107]. 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.

7.3.5 Effects of the pre-treatment on the digestate dewaterability

CST values for both digesters are depicted in Figure 7.5. 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. [111] could, therefore, not be realized. Given previous findings that US pre-treatments can also deteriorate sludge dewaterability [102, 176], 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 [177].

7.3.6 Economic aspects

The economics of the full-scale pre-treatment were assessed by comparing realized benefits (including savings due to reduced pump energy demand through lower WAS viscosity and increased electricity production due to enhanced methane yields) and treatment costs (including electricity and investment costs). Hereby, investment costs were considered as

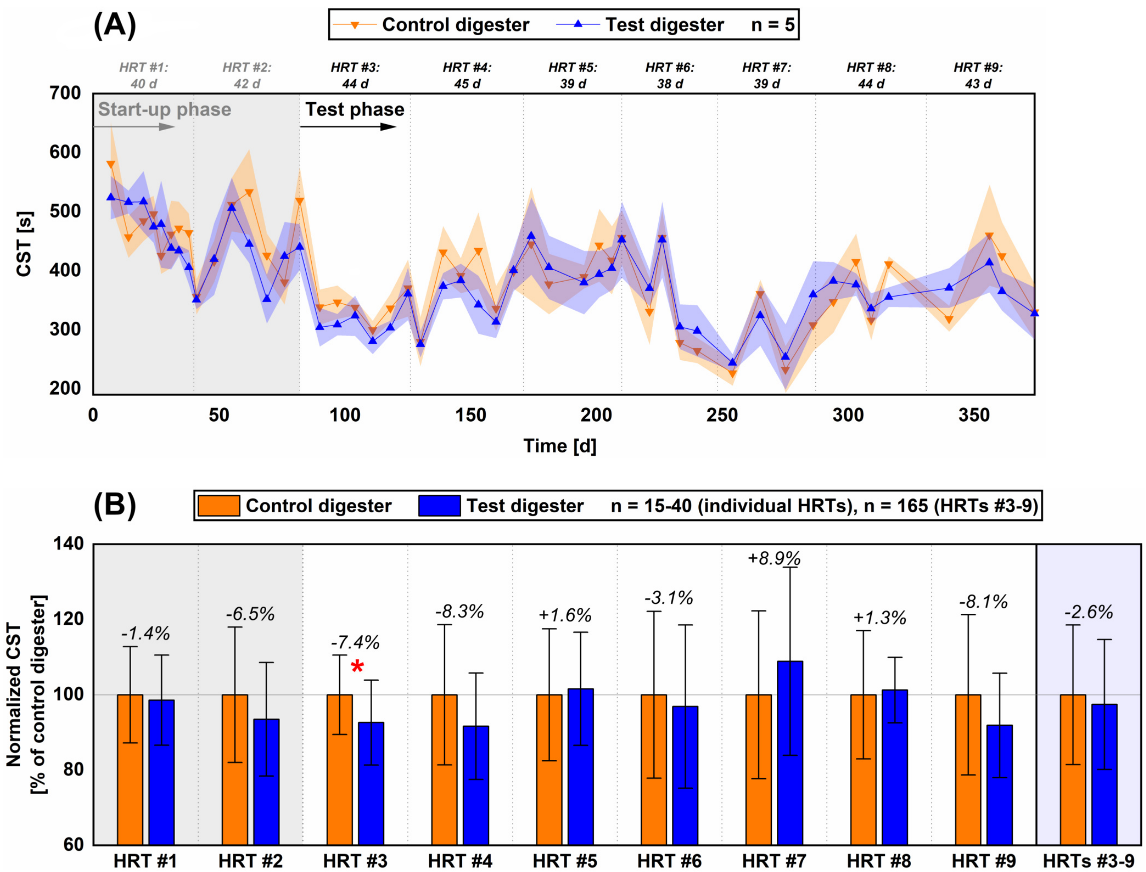

Figure 7.5: Development of the capillary suction time (CST) of the digestate from the control and the test digester (A) and normalized CST relative to the control digester (B). Significant differences between two data sets are denoted with an asterisk. Error bands and error bars denote the standard deviation of the mean.

depreciation costs (i.e., investment costs divided by the expected lifetime of the reactor), assuming three different depreciation periods of 1, 3, and 6 years. Furthermore, it should be noted that the economic assessment assumed pre-treatment of the full amount of the daily produced WAS, as an economic assessment of the conducted test operation (where only one digester receives sonicated sludge) would consider the full investment costs but only half the treatment effect. In order to extrapolate from test conditions to normal operating conditions (where both digesters receive sonicated sludge), electricity costs of the US reactor, savings due to reduced pump energy demand, and savings through additional methane yield were doubled. For calculating the savings through additional methane, an energy content 10 kWh per m^3 of CH_4, an electrical efficiency of the combined heat and power plant of 40%, and an electricity price of 21 Cent/kWh was assumed. The resulting cost comparison is depicted in Figure 7.6.

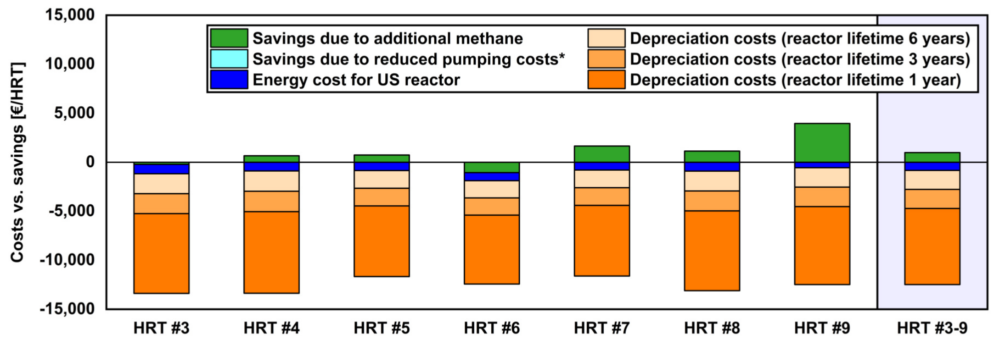

Figure 7.6: Development of the costs and savings of the full-scale ultrasound (US) pre-treatment, with HRT = Hydraulic retention time. *The cost reduction due to reduced pumping energy demand (< 1 € /HRT) is too little to be discernible in the graph.

Overall, obtained results suggest that the conducted full-scale treatment was uneconomical. When considering the average costs and benefits obtained from HRT #3 to HRT #9, treatment costs were roughly 13, 5, and 3 times higher than the achieved benefits, for depreciation periods of 1, 3, and 6 years, respectively. Hereby, it should be noted that the depreciation costs posed a much greater financial burden than electricity costs; even when assuming a long reactor lifetime of 6 years, average depreciation costs were still more than twice as high as electricity costs.

Given the only moderate enhancement of anaerobic digestion performance, the result of unfavorable treatment economics is not surprising. Only WAS viscosity was significantly reduced through the treatment (by -5.8% on average), which entailed a slightly lower head loss in the WAS conveying pipe (= 0.42 m on average). However, as the reduction only led to average pump energy savings and corresponding cost reductions of 3.2 kWh and 0.68 € per HRT, respectively, the improvement was negligible. Given this really minor saving of less than 1 € /HRT, reduced pumping costs are not even visible in (Figure 7.6). Moreover, it should be noted that the saved average head loss was less than the additional head loss caused by the US reactor (= 0.70 m on average) so that the full-scale reactor even caused a small net increase in head loss (= 0.28 m on average), which was naturally also irrelevant concerning treatment economics.

Electricity production from additional methane, on the other hand, allowed for noteworthy cost recovery, and at least electricity costs were regained by the increased methane yields. On average, cost savings due to additional methane (= 980 € /HRT) were almost 20% higher than the electricity demand of the US reactor (= 830 € /HRT), while in HRT #9, cost recovery through surplus methane was over seven times higher than the electricity costs of the US reactor (4,000 € /HRT vs. 550 € /HRT). In fact, during HRT #9, the gain in methane yield even sufficed to recover the depreciation costs, when assuming a depreciation period of 6 years. This promising cost recovery was, however, also favored by strongly reduced electricity consumption of the reactor in HRT #9, as the power supply was reduced to less than 8 kW due to the onset of wear of the US transducers. Given this development, it seems highly questionable if an assumed reactor lifetime of 6 years is a meaningful estimate, however. Besides, it should be noted that the incorporation of additional methane yield in the economic assessment is speculative, given the only insignificant increase.

7.3.7 Recommendations for future (full-scale) testing

The obtained results suggest that US pre-treatment does not lead to significant (or economically attractive) improvements of anaerobic digestion performance at well-performing WWTPs with 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 larger share of WAS 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 in case the pre-treatment can prevent the construction of an additional digester. Compared to the construction costs of a digester (tyically amounting to millions of € [20]), the investment and operating costs of a US reactor seem verifiably small so that even a treatment with a negative energy balance could be economical.

Another possibility to increase the performance of ultrasonic disintegration could be a shift from pre-treatment 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 increases than WAS pre-treatment [43, 65, 67, 145], which could be explained by the significantly lower TS and the hereby improved formation of cavitation bubbles [125]. 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 full-scale WWTPs. 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 - 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 treatment.

7.4 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. Besides, the additional gains were not able to recover the investment costs of the reactor, thus rendering the treatment overall uneconomic. Regarding operational stability, no clogging events were experienced despite an operation of 374 d 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.

7.5 Acknowledgments

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7.6 Appendix - Supplementary material

The graphical abstract and supplementary material to this article can be found in Appendix E of this thesis.

CHAPTER 8

From pre-treatment to co-treatment - How successful is ultrasonication of digested sewage sludge in continuously operated anaerobic digesters?

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Author contributions: Thomas Lippert was responsible for the research plan, data analysis, and manuscript preparation. Jochen Bandelin, Konrad Koch, and Thomas Lippert planned, realized, and automated the experimental setup. Sampling, laboratory analyses, and reactor maintenance were conducted by Yunqi Xu, Yu Chen Liu, Gabriel Hernández Robles, and Thomas Lippert. Jörg E. Drewes and Konrad Koch supervised the study. The manuscript was reviewed by Jochen Bandelin, Jörg E. Drewes, and Konrad Koch.

Abstract

The present study assessed the performance of ultrasonic co-treatment, i.e., the treatment of digested sewage sludge in continuously operated anaerobic digesters. Experiments were

carried out using a non-sonicated control digester and a sonicated test digester. The test digester received side-stream sonication (∼10% of reactor volume per day) at a specific energy input of 2,000 kJ/kg_{TS}. Treatment effects were monitored based on (i) specific methane production, (ii) (volatile) solids removal, and (iii) digestate dewaterability. Results revealed that co-treatment significantly enhanced average methane production (+6%), volatile solids removal (+9%), and solids reduction in the digestate (-5%). However, due to the only moderate enhancement and the relatively high energy input, the average cost recovery (i.e., the ratio between electricity costs and benefits due to additional methane and improved solids removal) was only 6% - 9%, depending on the assumed disposal costs. Moreover, as sonication led to impaired digestate dewaterability (average increase in normalized capillary suction time by 14%), the cost recovery due to reduced residual sludge might be negated again by lower dewatering efficiency. Overall, co-treatment seemed not economical under the conditions investigated. To render co-treatments economically feasible, further research, especially exploring the potential of low energy input sonication, is required.

8.1 Introduction

Ultrasound (US) can be applied in wastewater treatment for mechanical sewage sludge disintegration [12]. The disintegrating effect of US treatments is based on transient acoustic cavitation, which is defined as the consecutive formation, growth, and implosion of acoustic micro-bubbles [47]. Due to the extreme violence of the bubble implosions, strong shear forces are released into the sludge, causing the de-agglomeration of sludge flocs and the lysis of bacterial cell walls [56]. Commonly reported effects of sludge disintegration include increased biogas production, enhanced organic matter removal, and changes in sludge dewaterability [29, 33, 91, 102].

On the one hand, ultrasonication of sewage sludge is a well-investigated research topic, and numerous studies confirm its positive effects on anaerobic sludge digestion [4, 12, 19]. On the other hand, most studies considered US a pre-treatment only, commonly applied to waste activated sludge (WAS) [67, 178]. Yet, sonication of WAS creates several (operational) challenges. First, the relatively high total solids (TS) concentration of thickened WAS (typically 4% - 6% of fresh matter [133, 179]) causes severe sound wave attenuation with potentially detrimental effects on sonication efficiency [114, 171]. Optimum TS levels for WAS pre-treatment were found to be as low as 2.3% - 3.2% [14]. Second, pre-treatments can be somewhat non-selective, as the raw, undigested sludge contains not only recalcitrant, but also relatively easily degradable compounds. This particularly applies when mixtures of WAS and primary sludge (PS) are pre-treated [65, 66]. Third, the thickening of WAS (commonly conducted using a belt filter press) is sometimes undertaken during the working hours of the wastewater treatment plant (WWTP) staff only, which also limits the operating times of US reactors to 8 - 9 hours per day. In consequence, the efficiency of ultrasonic WAS pre-treatment is hampered by daily stand-by periods of up to 16 hours.

Based on the above arguments, sonication of digested sludge (DS) seems like a promising alternative. With a solids content typically ranging from 2.5% to 3.0% [159], DS exhibits TS concentrations within the optimum range for US treatments (2.3% - 3.2%, [14]). Moreover, as DS has already undergone anaerobic digestion, US treatments can be efficiently concentrated on the remaining recalcitrant sludge substances [65]. Furthermore, stand-by periods can be entirely avoided, as recirculation loops (being an ideal location for the US reactor installation) are typically operated 24 hours per day for digester mixing, heating, and raw sludge feeding. By being conducted not before, but during anaerobic digestion, such treatments are referred to as *co-treatments* [26]. A similar treatment scheme is termed *inter-treatment*, where DS is sonicated when passing from a first to a second digester [66, 180].

Despite the above-listed advantages, available literature on co- and inter-treatments is still scarce. Yet, the results of existing studies seem to confirm that DS sonication is, in fact, a promising alternative to conventional WAS pre-treatment. Koch et al. [43] compared the amenability of three different sludge types (raw sludge, agricultural sludge, and DS) towards US treatment and found that DS was by far the most suitable substrate, showing a significant increase in biochemical methane potential (BMP) of over 50% at a specific energy input of 2,670 kJ/kg_{TS}. In contrast, the methane production enhancement for the other two substrates remained insignificant. Equally promising results were achieved by Bandelin et al. [145], who realized gains in BMP of over 60% at an energy input of 3,500 kJ/kg_{TS}. Garoma and Pappaterra [65] even reached increases of over 130% for a similar municipal DS sample, albeit at an energy input of almost 60,000 kJ/kg_{TS}.

Nonetheless, the above results require critical interpretation, as they were obtained in batch digestion tests, which tend to overestimate (pre-)treatment effects. For instance, Janke et al. [93] found that methane production of pre-treated sugar cane filter cake was enhanced by over 20% in batch tests, while no enhancement was observed in a continuous test. A similar result was obtained by Moeller et al. [94] who reported increases in BMP of about 15% after mechanical disintegration of triticale grains, while the enhancement nearly vanished in continuous digestion tests. Promising results of batch tests are, therefore, not directly transferable to continuous (or semi-continuous) full-scale digesters and require validation in continuous digestion tests [104].

To date, only one study by Azman et al. [64] addressed DS sonication in continuously operated digesters. In their study, manure digestate was co-treated in a continuous pilot-scale digester at specific energy inputs of 1,500 kJ/kg$_{TS}$ and 3,000 kJ/kg$_{TS}$. Despite energy inputs similar to the previous batch tests, only moderate increases in methane production could be achieved (maximum gain of 18%), while in two out of three test phases, methane production even declined due to the treatment [64]. Thus, the results confirm that an enhancement of methane production is more challenging to sustain in continuous systems. Yet, it should be noted that the manure digestate exhibited a TS concentration of over 7% so that the co-treatment was subjected to the same sound wave attenuation as WAS pre-treatments. One of the key advantages of DS sonication (i.e., a comparably low TS content) remained, therefore, unexploited.

To further investigate the potential of co-treatments based on a substrate with a more suitable TS concentration, sonication of municipal DS with a solids content of approximately 3% has been studied. For the first time, the effects of co-treatment were holistically evaluated based on (i) methane production, (ii) (volatile) solids degradation, and (iii) digestate dewaterability in a continuously operated biogas test system. To obtain results transferable to full-scale applications, the operating conditions of the lab-scale test system were closely aligned to the conditions of a WWTP, especially regarding organic loading rate (OLR) and hydraulic retention time (HRT). For investigating whether co-treatments could help to shorten digestion time while keeping the same digestion performance, the experiments were carried out at two different HRTs of 20 and 30 days. Comparability to the results of previous studies on co-treatments was ensured by selecting a specific energy input of approximately 2,000 kJ/kg$_{TS}$. For a preliminary estimation of treatment economics, the electricity costs of the US reactor were balanced against potential benefits due to additional methane production and reduced residual sludge masses for disposal.

8.2 Materials and methods

8.2.1 Origin of inoculum, feed sludge, and sampling procedure

Sewage sludges were sampled at the municipal Freising WWTP, located 30 kilometers north of Munich, Germany. The plant has a design capacity of 110,000 population equivalents and operates three anaerobic digesters for the stabilization of PS and WAS. The digesters are operated at a temperature of 38°C, an HRT of 30 days, and an OLR of 1.1 - 1.5 kg volatile solids (VS) per day and cubic meter of digester volume.

For the initial inoculation of the lab-scale digesters, a one-time sampling of DS from the effluent line of the digesters was conducted. Before the inoculation, the sludge was sieved through a 1 mm mesh to prevent pre-mature wear of the laboratory pumps due to coarse material. Sampling of thickened WAS (used as feed sludge) was conducted once a week at a sampling tap after the belt thickener. Both TS and VS concentrations of the sludges were determined according to standard methods [126] and are presented in Table 8.1.

Table 8.1: Characteristics of the sampled sludge types.

	Inoculum (sieved)	**WAS** Mean	Min.	Max.	Standard deviation
TS [% of FM]	2.89 ± 0.01	4.41	2.98	5.09	0.47
VS [% of TS]	51.7 ± 0.31	71.1	67.0	74.6	1.83

TS = Total solids, FM = Fresh matter, VS = Volatile solids, WAS = Waste activated sludge

8.2.2 Laboratory digester system

The employed lab-scale digester system (BTP 2 Control, Umwelt- und Ingenieurtechnik GmbH, Dresden, Germany) comprised two identical anaerobic digester setups, one of which was used as a control digester (non-sonicated) and one as a test digester (sonicated). A schematic of the digester system is presented in Figure 8.1.

8.2.2.1 Digestion tanks and recirculation loop

Both setups contained a cylindrical digestion tank with a nominal volume of 32 L. The tanks were continuously stirred at 40 revolutions per minute using propeller agitators and tempered to 38°C by the use of heating jackets.

Each digester setup was furthermore equipped with an external recirculation loop, comprising a US flatbed reactor (BANDELIN electronic GmbH & Co. KG, Berlin, Germany), an eccentric screw pump (KB-20-S, Pumpenfabrik Wangen GmbH, Wangen, Germany), and connecting tubes. The US reactor in the loop of the control digester was permanently de-activated and only used to ensure identical flow conditions and equal volumes in the loops (3.2±0.05 L each). Tallied volumes of digestion tank and recirculation loop amounted to 35.2 L. Recirculation of the sludge was conducted using the eccentric screw pumps, which were activated for the first ten minutes of every hour at a flow rate of approximately 100 L/h. To preserve mesophilic conditions also in the external recirculation loops, the tubes and the US reactors were thermally insulated and equipped with a pipe heating.

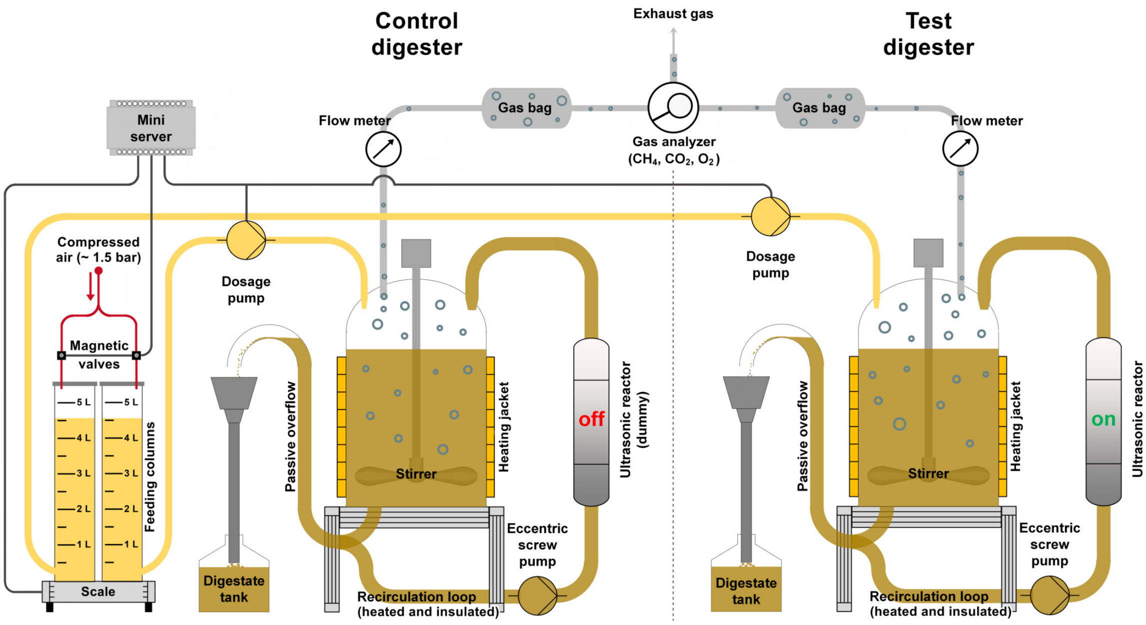

Figure 8.1: Schematic of the laboratory digester system, including feeding system, anaerobic digesters (control and test digester), recirculation loops, ultrasonic reactors, and gas analysis system.

8.2.2.2 Feeding

After inoculating both digesters with 30 L of sieved DS, digesters were fed semi-continuously with identical amounts of WAS four times a day (i.e., every six hours). During start-up and test phase #1, 1 kg of WAS was fed to each digester per day (i.e., 250 g per feeding), leading to a statistical HRT of 30 days. During test phase #2, the daily feeding was increased to 1.5 kg (i.e., 375 g per feeding), causing a reduction of HRT to 20 days. The resulting average OLRs amounted to approximately 1.1 $kg_{VS}/(m^3{\cdot}d)$ during start-up and test phase #1, and to approximately 1.6 $kg_{VS}/(m^3{\cdot}d)$ during test phase #2, similar to the OLRs at the Freising WWTP.

The feeding of the digesters was automated using the house automation technology Loxone, comprising a miniserver and automation software (Loxone Electronics GmbH, Kollerschlag, Austria). Further parts of the automated feeding system were an electronic scale (WPT-60-C2-K, Radwag Waagen GmbH, Hilden, Germany), two 5 L acrylic glass feeding columns (one for each digester), a compressed air system, and peristaltic dosage pumps. Feeding columns were placed on the scale, filled with WAS, and connected to the compressed air system. Using magnetic valves, the system could impose an overpressure of approximately 1.5 bar on the feeding columns, to aid the peristaltic dosage pumps in conveying the highly viscous sludge. For enabling the interaction of the scale, the magnetic valves, and the dosage pumps, all components were interconnected through the miniserver.

Feeding events were initiated by imposing pressure on one of the two feeding columns. Due to the pressure, a small amount of sludge was forced into the connecting tubes, causing a weight reduction on the scale. To avoid reduced feeding precision due to this effect, the system was given a 10-minute stabilization period to reach a constant weight again. After the stabilization, the current weight on the scale $m_{current}$ was read off by the miniserver, and the target weight m_{target} was calculated by subtracting the feeding amount (for instance, 250 g). Subsequently, the miniserver activated the peristaltic dosage pump, until the weight on the scale reached m_{target}. After the feeding cycle was completed for one digester, the overpressure was released from the feeding column, and the feeding procedure was repeated for the other digester with an offset of 30 minutes. The precision of the feeding system was monitored by automatic logging of the weights on the scale, and by visually checking the filling levels of the graduated feeding columns.

The surplus volume of sludge that was added during feeding events was released from the digesters using passive overflows and discarded. For ensuring identical filling levels in both digesters, the height of the overflows was adjusted using a spirit level. Clogging of the overflows (potentially leading to uneven filling levels and increased maintenance) was prevented by attaching the recirculation loops to the lowest point of the passive overflows.

8.2.2.3 Gas analysis system

The volume of the produced biogas was measured using drum-type gas flow meters (TG-05, Dr.-Ing. RITTER Apparatebau GmbH & Co. KG, Bochum, Germany) and subsequently normalized to standard conditions (dry gas, temperature of 0°C, pressure of 101,325 Pa). For the humidity correction, the temperature-dependent water vapor pressure was determined according to Justesen et al. [181] assuming water vapor saturation in the produced biogas.

After passing through the gas flow meters, the biogas was temporarily stored in 10 L sample bags. Every four hours, the stored gas was conveyed to a gas analyzer (SSM-6000, Pronova Analysetechnik GmbH & Co. KG, Germany) and automatically analyzed for CH_4, CO_2, and O_2 concentrations. Based on the gas composition analysis, methane production was calculated by multiplying the biogas norm volume with the methane concentration.

8.2.3 US reactor and ultrasonic treatment

Ultrasonication of DS was conducted using a lab-scale US flatbed reactor with six piezoelectric transducers, operated at a constant frequency of 25 kHz, and a nominal power of 50 W each (total power of the reactor = 300 W). The lab-scale reactor features a box-shaped reaction chamber (height = 30 mm, width = 80 mm, length = 200 mm) with a resulting active cavitation field volume of approximately 0.5 L. A detailed depiction of the reactor layout can be found in [149].

Sonication was performed six times per day (i.e., every 4 hours) for 100 seconds each time. The treatments were conducted during the off-times of the recirculation pumps to achieve constant energy inputs despite potential variations in flow rates due to the wear of the pumps. Owing to a reaction chamber volume of 0.5 L and a number of six treatments per day, about 10% of the digester content was sonicated daily. The resulting specific energy input ranged from approximately 1,800 kJ/kg_{TS} to 2,300 kJ/kg_{TS}, depending on the slightly varying TS content of the DS. The equation for calculating the specific energy input is given elsewhere [133].

8.2.4 Digestate analysis

Digestate was collected twice a week (Mondays and Thursdays) from sampling taps in the recirculation loops before the US reactors. The sampling was conducted during the operation of the eccentric screw pumps to ensure the collection of a representative sample.

The collected digestate was subsequently analyzed for TS, VS, and capillary suction time (CST). TS and VS were determined in triplicate, while the CST analysis was performed in quintuplicate using a CST test apparatus (CST 101/A, Axchem Deutschland GmbH, Erzhausen, Germany) and Axchem CST paper. A detailed description of both the test apparatus and the test procedure can be found in [174]. The volatile solids removal (VSR) during anaerobic digestion was calculated according to Koch [182].

8.2.5 Steady-state analysis

According to the VDI 4630 guideline for the fermentation of organic materials [128], all test phases were conducted for at least three HRTs, including the start-up phase. To test whether the digesters achieved steady performance after this time (so that a shift to the next test phase was admissible), a steady-state analysis was conducted according to the following criteria:

- Measurement values are independent of time for at least one HRT, i.e., the slope of a linear trend line through the measurement values shows no significant deviation from zero [175].
- The relative standard deviation (RSD) of measurement values obtained during one HRT with time-independent performance remains below 15% [70].
- Steady-state conditions are tested for all measurement variables investigated (methane production, (volatile) solids removal, and sludge dewaterability).

For assessing whether the slopes of the trend lines showed a significant deviation from zero, an analysis of variance (ANOVA) was performed using the software Origin Pro 2020 (OriginLab Corporation, Northampton, MA, USA), with significant deviations indicated by p values < 0.05. The selection of an RSD limit of 15% was based on previously observed result variability in continuous digestion tests (for instance, RSDs of 12% - 20% for biogas production [70]) and on the results observed in this study. A lower RSD limit (such as 5% for triplicate measurements in batch digestion tests [127]) seems too strict for continuous tests, especially when feed sludge characteristics are varying (as in the present study, see Table 8.1). The criterion that all measurement variables should be included in the steady-state analysis was based on the assumption that different variables require different adaptation times to reach steady performance.

It should be noted that the defined steady-state criteria were regarded as recommendations, not strict rules, as full compliance with all criteria at all times was difficult to achieve due to the dynamic nature of the biological system investigated.

8.2.6 Assessment of the differences between control and test digester

Due to the chosen setup (one control digester vs. one test digester), differences between the obtained data sets were assessed using a Student's t-test. Hereby, a data set was defined as the volume of all measurements obtained during one individual HRT. Differences were denoted as significant for $p < 0.05$ and as highly significant for $p < 0.01$. Compliance with the requirements of t-tests (i.e., normal distribution of data and homogeneity of variance) was checked beforehand using the Shapiro-Wilk and the F-test, respectively. The additional t-test requirement of independent measurement values was met through the selected experimental setup [175].

8.3 Results and discussion

The effects of ultrasonic co-treatment on the performance of anaerobic digestion were investigated with respect to (i) methane production, (ii) (volatile) solids degradation, and (iii) digestate dewaterability. For a preliminary estimation of treatment economics, the electricity cost for the US reactor were balanced against the potential benefits due to additional methane production and reduced residual sludge masses for disposal.

8.3.1 Effect of the co-treatment on the specific methane production (SMP)

Before examining the effect of the co-treatment on methane production, the performance of both digesters during the start-up phase was compared. The SMPs and the percentage difference between both digesters are displayed in Figures 8.2A and 8.2B, respectively.

According to the steady-state analysis, both digesters showed steady performance for most of the start-up phase, as confirmed by the insignificant slopes of the linear trend lines ($p_{\mathrm{min}} > 0.08$, for trend lines plotted over the four HRTs of the start-up phase) and by SMP fluctuations below the previously defined RSD limit of 15% (except for the third HRT with RSDs of up to 21%). Yet, a significant offset between both digesters was detected, which amounted to almost 11% in the first HRT and still 6% in the fourth HRT, on average. While a certain deviation between control and test digesters is common [175, 183], a significant offset as in the present case is critical as it impedes the exact determination of SMP enhancement due to the lack of a reliable reference value. To ensure a conservative assessment, the digester with the lower SMP was selected as test digester. Therefore, SMP enhancement was necessarily underestimated.

The detected offset demonstrates that even a carefully synchronized system can produce somewhat deviating data, which emphasizes the importance of both a control digester and a start-up phase. Without such scientific controls, the observed differences of 6% to 11% may have already been interpreted as treatment effect. The offset moreover suggests that also for continuous tests, the use of duplicates or even triplicates would significantly increase result reliability. However, given the comparably high costs of continuous digestion systems, a shift to duplicate or triplicate testing is also a question of available resources.

Upon activation of the US reactor, the SMP of the test digester increased both instantly and significantly, and in the first HRT of test phase #1, the mean SMP of the test digester was 13% higher than the mean SMP of the control digester. Due to the offset-related underestimation, the effective enhancement might have been even larger. However, after this initial boost, SMP declined again and even dropped to slightly negative percentage increases in the fourth and fifth HRT of test phase #1. In the second test phase, however, the enhancement slowly increased again, while the gains remained in approximately the same range as in test phase #1. Overall, the realized average improvements were rather small and amounted to 5.3% in test phase #1 and to 7.6% in test phase #2. The largest individual enhancement (averaged over one HRT) was obtained in test phase #2 and amounted to approximately 16%. The relatively similar enhancement in both test phases furthermore indicates that the reduction of HRT in test phase #2 did not notably change treatment performance, which is in line with Braguglia et al. [90], who found that sonication performance remained relatively

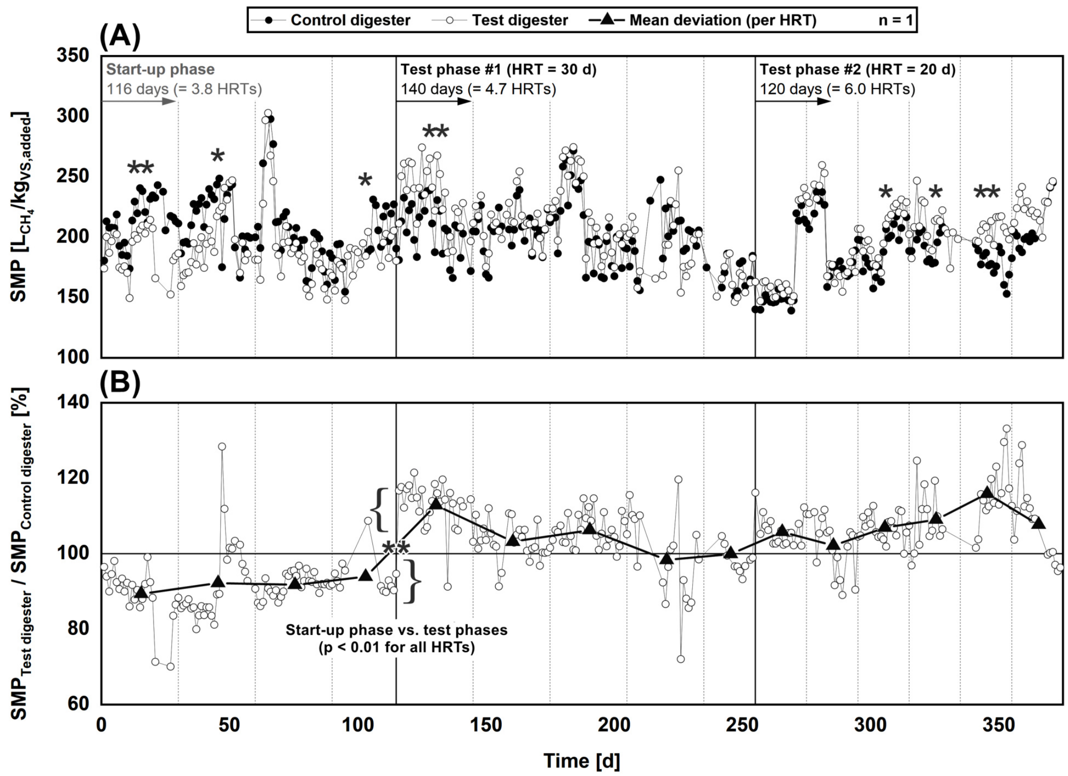

Figure 8.2: Development of daily specific methane production (SMP) of the control and the test digester (A) and percentage difference between both digesters (B). Significant and highly significant differences between the two data sets are denoted with one or two asterisks, respectively, for each hydraulic retention time (HRT).

unaffected when changing HRT from 20 to 10 days.

Although the setup somewhat underestimates treatment effects and hereby contributes to the only moderate results, it should be noted that even a doubling of SMP enhancement (which would be an overestimation) would still resemble much lower increases as the ones reported in previous batch tests (for instance, more than 50% at a comparable energy input [43]). Thus, the results challenge the initial hypothesis that a lower TS content would substantially increase the performance of co-treatments in continuous systems. In fact, the obtained SMP improvements were in a very similar range as the ones reported by Azman et al. [64] (maximum of 18%), despite a considerably lower TS content (3% vs. 7%).

When comparing the SMPs of control and test digester directly, SMP enhancement was only significant for four out of ten HRTs (see Figure 8.2A). However, such direct comparison is again biased by the offset between the two digesters. Thus, significance testing was additionally conducted based on percentage differences, i.e., percentage differences observed in the start-up phase were compared to the percentage differences in the test phases. Following this approach, SMP enhancement was found to be highly significant (at $p < 0.01$) for all HRTs of both test phases. Hence, it was concluded that co-treatments are able to significantly improve methane production, albeit to a much lesser extent as indicated in batch tests.

A likely reason for the comparably low enhancement is the previously acknowledged tendency of batch tests to overrate pre-treatment effects [93, 94, 175]. One explanation for such tendency could be long-term effects caused by a release of refractory or inhibiting compounds (as hypothesized by Azman et al. [64]), which cannot be detected within the short time span of a batch tests [104]. In the particular case of co-treatments, a damage of methanogens (or other vital anaerobic microorganisms) during sonication could be another explanation. Especially filamentous archaea (such as *Methanosaeta* [123]) might be critically affected by the treatment (just like filamentous bacteria in WAS pre-treatment [69]) so that a considerable part of the available organic material may not be used for methane production, but for microbial (re)growth and maintenance [184].

8.3.2 Effect of the co-treatment on (volatile) solids removal

The VSRs of both digesters and the resulting digestate TS concentrations are presented in Figures 8.3A and 8.3C, whereas their respective percentage differences are depicted in Figures 8.3B and 8.3D. In contrast to the methane production, deviations between control and test digester were mostly insignificant during the start-up phase for both parameters, indicating a highly comparable performance of the two digesters despite the previously observed offset in SMP. Steady-state criteria were met for both digesters during most of the start-up phase, and for the last HRTs of both test phases.

The results obtained in both test phases demonstrate that US co-treatment was able to significantly enhance VSR, with average and maximum improvements of 9% and 14%, respectively. The enhanced VSR naturally translated to lower digestate TS concentrations, showing an average reduction by approximately 5%.

The reduction of HRT in test phase #2 led to an expected declining trend in VSR and an increase in digestate TS concentrations for both digesters. Nevertheless, the co-treated digester still showed superior (volatile) solids removal compared to the control digester. At the same time, the treatment was not able to keep VSRs at the same level as observed for the non-sonicated control digester in the first test phase. The initial claim that co-treatments could preserve the same digestion performance at shortened HRTs (or increased OLRs) is, therefore, only partially met.

It is interesting to note that the highest VSR enhancement was found for the two HRTs

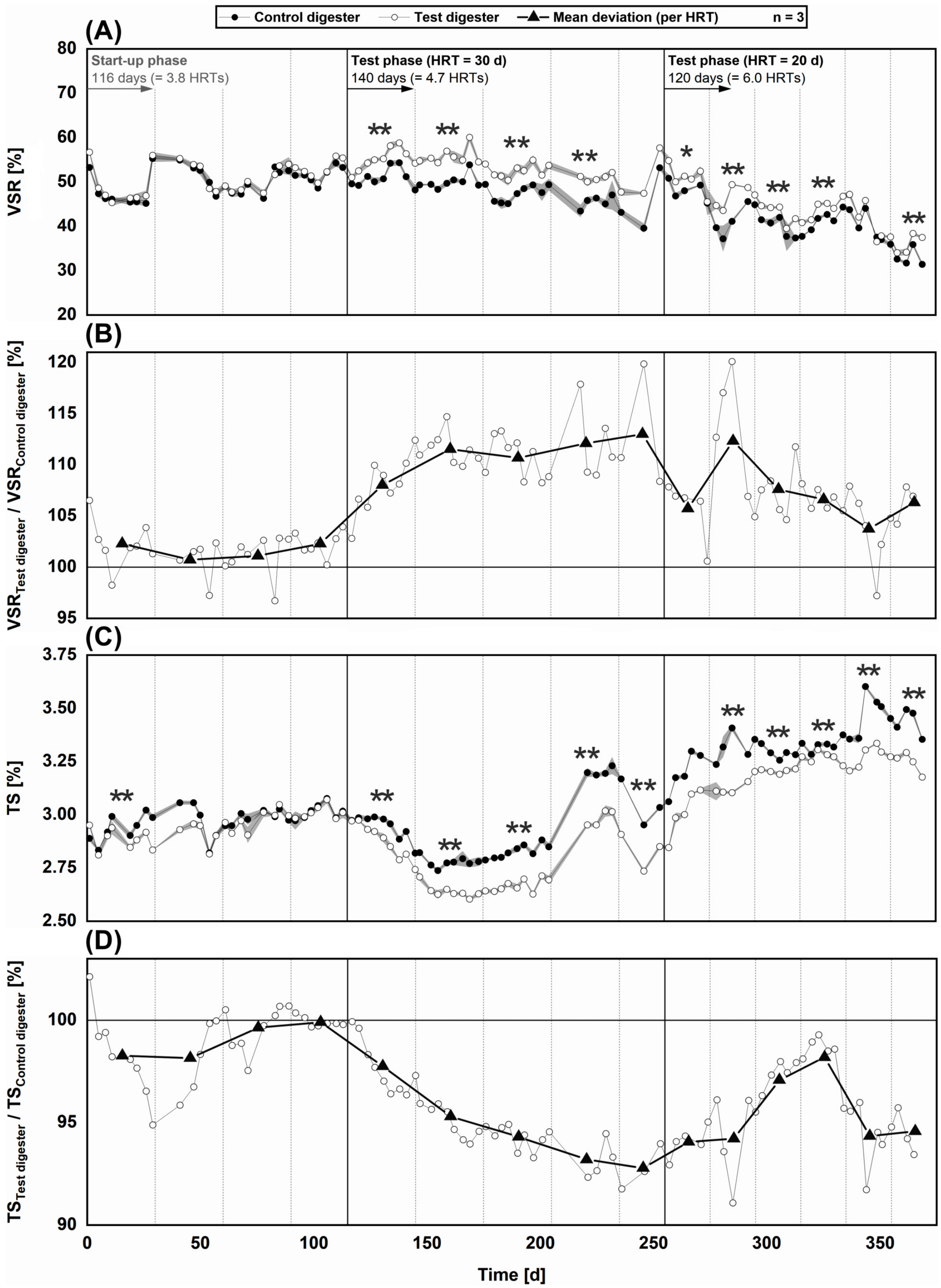

Figure 8.3: Development of volatile solids removal (VSR) (A) and total solids (TS) concentration of the digestate (C) of the control and the test digester, and percentage differences between both digesters (B and D). Significant and highly significant differences between the two data sets were denoted with one or two asterisks, respectively, for each hydraulic retention time (HRT). Error bands denote the standard deviation of the mean (n = 3).

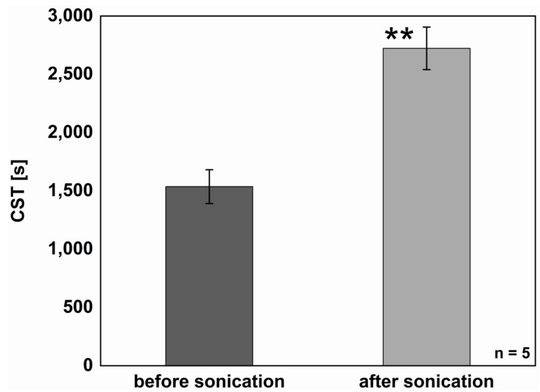

Figure 8.4: Capillary suction time (CST) of digested sludge before and after batch sonication at a specific energy input of 2,000 kJ/kg_{TS}. Error bars denote the standard deviation of the mean (n = 5).

with the lowest increase in methane production (test phase #1, HRTs four and five). This observation further indicates that the disruption of the microbial community during the co-treatment might cause elevated substrate partitioning to cell synthesis and maintenance.

8.3.3 Effect of the co-treatment on digestate dewaterability

The effect of sonication on dewaterability was examined directly after the treatment (i.e., for sludge exiting the US reactor) and for the mixed sludge in the sonicated test digester. The direct effect of the sonication was determined in an additional batch sonication test, as sample collection from the US reactor was not possible in the continuous system. The batch sonication was conducted at the same specific energy input of 2,000 kJ/kg_{TS}, following the experimental procedure reported in [133] and using an identical US reactor as in the continuous experiment. The resulting CSTs of the sample before and after the batch sonication are presented in Figure 8.4.

The treatment led to a highly significant increase in CST values by almost 77%, indicating a severely deteriorated sludge dewaterability immediately after sonication. The findings confirm previous studies, similarly reporting a strong increase in CST values after US treatments at comparable specific energy inputs [71, 73, 103].

To examine whether the constant intake of sludge with elevated CSTs also deteriorates the overall dewaterability of the reactor content, the CST of the digestates from both the control and the test digester were additionally analyzed, as displayed in Figure 8.5. Due to the different solids concentrations in the two digestates (see Figure 8.3), CST values were normalized according to their TS concentration (i.e., CST was divided by TS as recommended in standard methods [126].

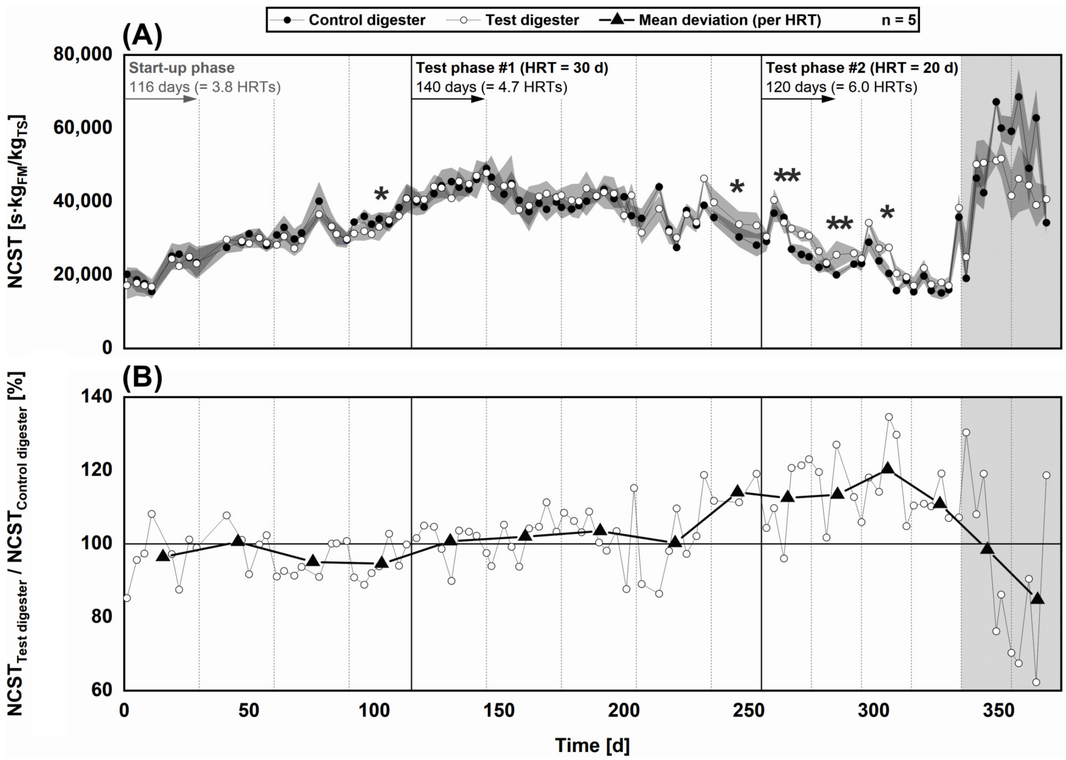

Figure 8.5: Development of normalized capillary suction time (NCST) for the control and the test digester (A) and percentage difference between both digesters (B). Error bands denote the standard deviation of the mean (n = 5). Significant and highly significant differences between the two data sets were denoted with one or two asterisks, respectively, for each hydraulic retention time (HRT). Due to massive variation, the last two HRTs (highlighted in grey) were not considered in the significance tests.

At first glance, the difference between the normalized CSTs (NCSTs) appears verifiably small despite the strong direct effect observed in the batch sonication. The attenuation of the effect can possibly be explained by the side-stream sonication (only about 10% of the sludge are sonicated per day), but also by a degradation of compounds responsible for the impaired dewaterability during anaerobic digestion [34].

Nonetheless, the NCST of the test digester showed a relative increase over time. In the last HRT of test phase #1, the value was already elevated, while in the third HRT of test phase #2, the average NCST of the sonicated digester was more than 20% higher than in the control digester. Averaged over all HRTs that showed significantly impaired dewaterability, the percentage increase in NCST amounted to 14%. Thus, results suggest that in the long-run, the co-treatment also caused a significantly impaired dewaterability of the whole reactor content.

Steady-state analysis indicated that NCST showed a comparably slow response to changing operating conditions. During start-up, NCST kept constantly increasing, indicating that even

3.8 HRTs were not enough to establish steady conditions. The reason for the observed increase may be the change in feeding conditions, i.e., from a blend of PS and WAS at the WWTP to WAS only in the lab. A second indication for the parameter´s slow response is the decrease in NCSTs observed in the first four HRTs in test phase #2, despite the 50% increase in OLR. In the fifth HRT, however, a sharp increase in NCSTs was observed, which was deemed a delayed effect of the increase in OLR. As the parameter clearly did not reach steady-state within the two last HRTs of test phase #2, the difference between the two digesters was not considered in the significance tests during that time. Results suggest that a meaningful investigation of sludge dewaterability requires testing of a larger number of HRTs, probably even more than in the present study. The result moreover demonstrates that there is not one steady-state condition for all parameters, but that each measurement variable requires an individual adaptation time to reach steady performance. At the same time, it should be noted that CST tests cannot quantitatively predict the dewatering performance of full-scale centrifuges [185], so that even a steady CST value can only serve as a qualitative indicator for digestate dewaterability.

8.3.4 Estimation of economic feasibility and full-scale applicability

For assessing the economic feasibility of the treatment, the electricity costs of the US reactor were balanced against the benefits due to additional methane recovery and a reduced residual sludge mass for disposal. Electricity costs were calculated based on the reactor´s electric power (= 300 W), the daily operating time (= 600 s), and an electricity price of 30.43 Cent/kWh (according to the German Federal Ministry for Economic Affairs and Energy, www.bmwi.de/en). The amount of current that could be generated due to additional methane was calculated assuming an energy content of methane of 10 kWh per m^3 and a conversion efficiency of the combined heat and power plant of 40% [133]. The resulting benefits were determined based on the same electricity price of 30.43 Cent/kWh, assuming savings due to reduced energy demand. For the benefits due to improved solids degradation, a range of disposal costs of 200 €, 300 €, and 400 € per ton of TS (including costs for transport and dewatering to a TS concentration of 25%) was assumed, according to Wiechmann et al. [23]. Neither investment, amortization nor maintenance costs of the US equipment were considered in the simplified assessment. The observed changes in sludge dewaterability could not be included in the calculations, either, due to the aforementioned limitations of CST tests [185]. A detailed example calculation can be found in Appendix F, Table F.1.

The resulting average cost recoveries for all HRTs are displayed in Figure 8.6. The maximum energy recovery amounted to ∼18%, while the average recoveries over both test phases ranged from 6.2% to 8.6%, depending on the assumed disposal costs. The finding

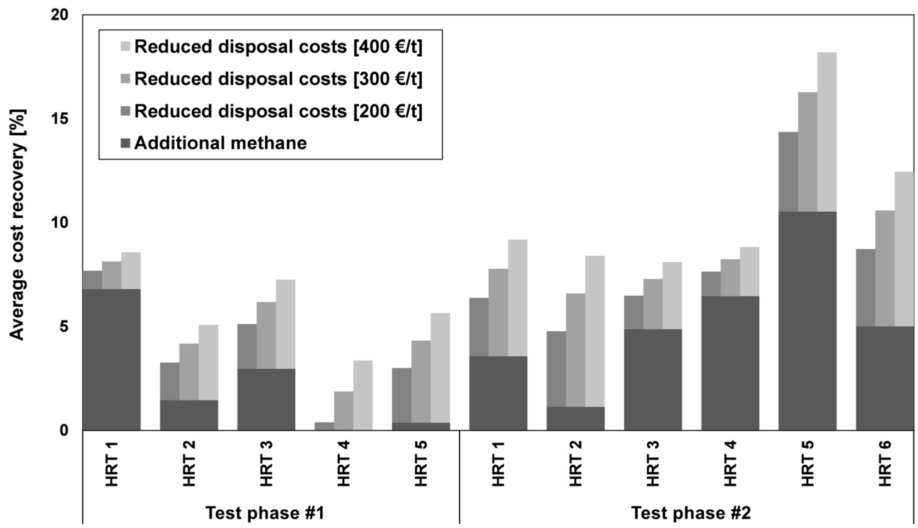

Figure 8.6: Estimation of average cost recoveries (i.e., ratio of electricity costs vs. cost savings due to additional methane and reduced residual sludge mass) for each HRT of both test phases.

demonstrates that even the combined benefits due to additional methane and reduced residuals could not compensate for the energy costs of the US reactor by far. Even when doubling SMP enhancement (to clearly exclude any negative impact of the offset between the digesters), the average energy recovery would still remain at a relatively low level of approximately 12%, assuming the highest disposal costs of 400 € per ton of TS.

Moreover, the calculated savings due to reduced residual sludge assumed constant dewatering performance. However, based on the conducted CST analysis, it is likely that dewatering would be impaired so that the advantages due to improved solids removal may be canceled out again. Such negative side effect would further hamper the overall treatment economics, as the enhanced solids removal was responsible for roughly half of the benefits when assuming medium disposal costs of 300 € /t.

The findings of a negative cost (or energy) balance for co-treatment are in line with the study by Garoma and Pappaterra [65] who reported energy recoveries between 4% and 11% for municipal digestate sonication in lab-scale. Also, Azman et al. [64] concluded that the energy requirements for the sonication of manure digestate clearly exceeded the energy content of the additionally produced methane.

To achieve a more economical cost recovery, a viable strategy could be a reduction of specific energy inputs. In a study on WAS pre-treatment, low energy inputs (for instance, 200 kJ/kg_{TS}) were shown to cause a similar enhancement of specific methane yield than high energy inputs (for instance, 2,000 - 3,000 kJ/kg_{TS}) [133]. While the findings were made for WAS pre-treatment and by the use of BMP tests so that a direct transferability seems questionable, a reduction in specific energy inputs still seems a reasonable next step to drive co-treatments to a more economic range. Yet, when applying much lower energy inputs, the anyway small stimulation of the process might be further reduced.

When co-treatments are used to avoid the costly construction of an additional digester by increasing the capacity of an existing digester, the treatment could be economical despite the negative cost recovery. However, as was observed from the reduction of HRT from 30 to 20 days, co-treatments could not maintain the same performance that was achieved with an HRT of 30 days without treatment. Thus, it seems questionable whether the moderate enhancement observed would suffice to avoid the construction of a new digester. A more distinct effect of the treatment might only be noticeable for severely reduced HRTs (such as 10 days), where the digestion process might fail completely without additional treatment.

In addition to examining cost recovery, it was deemed worthwhile to consider the practical implications of a 2,000 kJ/kg_{TS}-treatment regarding full-scale applicability. In the lab, the selected energy input was achieved by operating a 300 W reactor for only 10 minutes per day. However, to obtain the same energy input in a full-scale plant (assuming a digester volume of 2,500 m^3, a side-stream sonication of 10% of the reactor content per day, and identical sludge properties), a reactor system with an electrical power of 180 kW operating 24 hours per day would be necessary. However, this by far exceeds the electrical power of commonly installed full-scale reactors (for instance, 4 kW in [186], 10 kW in [45], 13.5 kW in [187], and 15 kW in [110]). Thus, it seems that an investigation of the potential of low energy input treatments is inevitable when an application of the technology in full-scale WWTPs is targeted. To conclusively assess the economics of such treatments, further possible treatment effects, such as viscosity reduction or foaming control, should also be considered.

8.4 Conclusion

The potential of ultrasonic co-treatment of digested sewage sludge was investigated using two continuously operated biogas test systems (comprising one control and one test digester) at HRTs of 20 and 30 days. The treatment (conducted at 2,000 kJ/kg_{TS}) led to significant, but only moderate enhancements of methane production (+6%, on average) and (volatile) solids removal (increase in VSR by 9%, and decrease of digestate TS by 5%, on average). The realized improvements could not recover the operating costs of the US reactor by far, and average cost recovery remained below 10%. Moreover, as the treatment impaired the dewaterability of the digestate, potential gains due to reduced residual sludge may be diminished again by increased dewatering costs. Thus, at the conditions investigated, ultrasonic co-treatment seemed to have only limited potential for enhancing anaerobic digestion performance. To render the treatment economically viable, future studies should explore the potential of low energy input sonication but also investigate further possible applications, such as viscosity reduction or foaming prevention.

8.5 Acknowledgments

This work was supported by the German Federal Ministry for Economic Affairs and Energy (BMWi), Grant 03ET1396B. The company BANDELIN electronic GmbH & Co. KG is kindly acknowledged for providing the US reactor system. Special thanks go to the staff members of the Freising WWTP for the outstanding support and to Meriam Muntau for the assistance at the continous test plant.

8.6 Appendix - Supplementary material

The graphical abstract and supplementary material to this article can be found in Appendix F of this thesis.

CHAPTER 9

Summary of research outcome and hypothesis testing

The main research outcomes of this dissertation are summarized in the following, along with a testing of the initially formulated research hypotheses. A summary of the results of the conducted hypothesis testing is presented in Table 9.1 at the end of this chapter.

9.1 Research objective #1

The first research objective was to evaluate whether the novel US reactor is capable of enhancing specific methane production and COD solubilization of substrates with industry-typical TS contents of 4% - 7% already at low, industrially feasible specific energy inputs (proof-of-concept). Regarding an economic use of the new reactor type, a closely related objective was to identify energy inputs that furthermore allow for an energy-positive sonication, meaning that the reactor´s energy consumption is lower than the energy production from the additionally generated methane gas. The first hypothesis was formulated as follows:

> ***Hypothesis #1:*** *"Ultrasonication of waste activated sludge at low specific energy inputs using a novel ultrasonic flatbed reactor leads to significant enhancement of the sludge´s biomethane potential so that the pre-treatment is energy-positive."*

Results obtained in **Paper I** demonstrated that the new reactor type is able to significantly enhance both COD solubilization and BMP of the tested substrates. While the relationship between specific energy input and DD_{COD} was found to be almost linear for all sludge types (ranging from 1% to 22%), no linear relationship was observed between energy input and BMP enhancement. In fact, low energy inputs (200 kJ/kg_{TS}) led to the same maximum BMP increase of 12% as high energy inputs (2,000 kJ/kg_{TS} and 3,000 kJ/kg_{TS}), despite verifiably small DD_{COD} values of only 1% - 5%. Intermediate inputs ranging from 400 kJ/kg_{TS}

to 1,000 kJ/kg$_{TS}$, on the other hand, could not realize significant BMP enhancement. This interesting effect, which has considerable economic implications, could be reproduced in a subsequent study within this research project [145] and has also been observed before [29, 43], albeit not in a systematic manner and without discussing this effect in more detail. As a potential explanation, it was hypothesized that the release of coagulating biopolymers at intermediate energy inputs leads to the re-flocculation of previously de-agglomerated sludge flocs, which would negate the beneficial treatment effects observed at low energy inputs. Naturally, such ineffective and uneconomic ranges of energy inputs should be avoided in any full-scale sonication application.

An energy-positive treatment was only obtained for one of the three tested sludges at a specific energy input of 200 kJ/kg$_{TS}$, with an energy recovery of 122%. For both other sludges, on the other hand, the treatment consumed more energy than was regained for all energy inputs tested. Hence, it can be concluded that an energy-positive treatment of WAS is generally possible with the novel reactor system but only at very low specific energy inputs and for individual sludge types. Hypothesis #1 was, therefore, considered as ***partially accepted***. At the same time, it should be mentioned that an economic assessment solely based on BMP tests remains largely simplified, as it does not account for the reactor´s investment and maintenance costs or other treatment effects such as potentially improved biosolids removal or digestate dewaterability. The distinct differences among the three WAS samples examined demonstrate that different sludges respond differently to US pre-treatment, thus emphasizing that testing of multiple samples is required for obtaining reliable conclusions. The finding that 'less is more' concerning specific energy input can be considered a crucial outcome, especially with respect to economic full-scale applications. The result furthermore emphasizes that not cell lysis, but floc de-agglomeration is the major working principle of US treatment in full-scale environments.

9.2 Research objective #2

The second research objective addressed the optimization of the reaction chamber design of the new reactor type in order to identify designs that allow for maximum disintegration efficiency at a given energy input. A suitable US reactor design seems particularly important for the treatment of viscous substrates such as sewage sludge, as the high sludge viscosity may cause both strong sound wave attenuation and a prevention of cavitation-induced mixing. A potentially inhomogeneous cavitation field might, therefore, not be compensated for by substrate mixing during the US treatment. Thus, before investigating the impact of reactor geometry on primary and secondary effects of cavitation, an examination of the fluid dynamics of sewage sludge during US treatments was conducted according to Hypothesis #2.1:

> ***Hypothesis #2.1:*** *"Sludge flow in ultrasound flatbed reactors is mostly laminar despite the impact of cavitation-induced micro-turbulences due to the high viscosity of the sludge."*

The hypothesis was tested by visual examination of the flow field during ultrasonic irradiation by using a digital camera and a US reactor with a transparent panel. To render the opaque sewage sludge flow visually accessible, the tested WAS and DS samples were substituted by transparent xanthan solutions with similar, non-Newtonian flow behavior. For visualizing flow laminae and treatment effects, dye streams were injected into the flow at the inlet of the reaction chamber. The experiments were carried at a flow rate of approximately 100 L/h, which corresponded to laminar, generalized Reynolds numbers of 0.03 for $WAS_{Xanthan}$ and 0.14 for $DS_{Xanthan}$, and theoretical specific energy inputs of 250 kJ/kg_{TS} and 430 kJ/kg_{TS} for WAS and DS, respectively.

The results of this investigation published in **Paper II** demonstrated that the flow regimes of both sludge surrogates remained largely laminar during the US treatment, except for an eruption-like branching of the dye streams that appeared locally as a consequence of individual cavitation events. Due to the high viscosity of the sludges, however, such movement was strongly damped and did not translate to macro-scale fluid mixing. The branching of the dye streams was much more pronounced for DS than for the more viscous WAS, where the dye laminae showed almost no disruption. Thus, given the strong suppression of cavitation-induced fluid movements and the accordingly mostly laminar flow during the treatment, Hypothesis #2.1 was ***accepted***. Results imply that a potentially inhomogeneous cavitation field is not compensated for by a cavitation-induced mixing, which emphasizes the critical role of US reactor design for sewage sludge treatment.

The optimization of US reactor design for maximum treatment homogeneity and disintegration efficiency at a given energy input was conducted by the use of a size-adjustable US reactor (with five RCH settings, ranging from 20 mm to 100 mm). To simulate treatments feasible in full-scale plants and according to the results of **Paper I**, all sonications were conducted at a constant specific energy input of 200 kJ/kg_{TS}, while thickened WAS with a TS concentration of approximately 4.5% was used as a substrate. Treatment effects were densely mapped throughout all reaction chamber geometries by the use of three different measurement techniques, including hydrophone measurements (recording cavitation noise as a primary effect of sonication), and aluminum foil tests and soluble COD analyses (both assessing secondary effects of sonication). This trifold approach enabled a thorough examination of the relationship between reactor design and sonication performance, and furthermore allowed assessing whether cavitation noise measurement is a suitable tool for optimizing US reactor designs or not. In addition to the sonications in batch mode, the flow of the sludge through the different reaction chambers was also accounted for by the use of a laminar CFD simulation (according to the findings of **Paper II**), in order to estimate how fluid dynamics further affect treatment (in)homogeneity. The according hypothesis for the reactor design optimization was formulated as follows:

> ***Hypothesis #2.2:*** *"Ultrasonic reactor designs with maximum average cavitation noise levels provide both maximum treatment homogeneity and maximum disintegration efficiency."*

The obtained results are presented in **Paper III** and confirm that US reactor design has a critical impact on both sonication performance and treatment (in)homogeneity. The study revealed that the smallest RCH of 20 mm allowed for comparably high average CNL and DD_{COD} values of 38 dB and 4%, respectively, while at the largest RCH of 100 mm, average CNL and DD_{COD} values critically dropped to 24 dB and 1%, respectively, despite the same energy input of 200 kJ/kg_{TS}. The qualitative aluminum foil measurements further substantiated the obtained results. The reason for the strong influence of the reaction chamber geometry was found to be severe sound wave attenuation towards the channel center due to the high solids content of the sludge. While RCHs of 20 mm and 40 mm showed no notable attenuation of cavitational effects, a decline of CNL by 34% (relative to the intensity at the transducer surface) was already observed in the channel center at an RCH of 60 mm, whereas CNL dropped by 63% when RCH was increased to 80 mm. For the largest gap distance of 100 mm, CNL even fell below the detection limit. Flow-through sonication aggravates this treatment inhomogeneity further, as the flow rates in the laminar flow regime are highest in the channel center where cavitation intensity is lowest. For instance, in batch sonication at RCH = 100 mm, approximately 43% of the sludge volume fell below the detection limit of the cavitation noise measurement, while in flow-through sonication, the share of non-exposed substrate increased to approximately 53%. Thus, the findings emphasize that identical energy inputs do not imply equal treatment intensities. As a rule of thumb, the obtained results suggest that RCHs of US reactors should be as small as possible (enabling high disintegration efficiency and treatment homogeneity), but as large as necessary (enabling low clogging risk and high operational stability).

The agreement between CNL measurements and secondary treatment effects (assessed via COD solubilization and aluminum foil erosion) was found to be quite high, as was substantiated by a strong linear correlation between average CNLs and average DD_{COD} values ($R^2 > 0.8$). Thus, results suggest that cavitation noise is a more reliable signifier for disintegration effects than the electrical power of a US reactor and that cavitation noise measurement is a suitable, time-efficient, and easy-to-use tool for optimizing reactor designs for both maximum disintegration efficiency and treatment homogeneity. Hypothesis #2.2 could , thus, be ***accepted***.

9.3 Research objective #3

Based on the outlined potential of low-energy-input sonication using the novel flatbed reactor (**Paper I**) and the previous reactor design optimization (**Paper III**), a full-scale testing of the reactor was conducted. To realize a pressure-resistant, tube-shaped reactor configuration that also features a relatively small gap distance for an intense cavitation field, a double-tube reactor with an RCH of 57 mm was manufactured. Such distance was seen as a good compromise between disintegration efficiency (RCH as small as possible) and operational stability (RCH as large as necessary). The setup featured a tallied power rating of 13.5 kW and was installed at the Starnberg WWTP (located 20 km south of Munich, Germany) for full-stream pre-treatment of thickened WAS at the previously identified most economic specific energy input of 200 kJ/kg_{TS} (**Paper I**). The mostly typical operating conditions of the Starnberg WWTP and the availability of two identical full-scale digesters operated in parallel (non-sonicated control digester vs. sonicated test digester) allowed for obtaining both transferable and reliable results. Treatment effects were monitored with respect to (i) methane production, (ii) (volatile) solids removal, (iii) sludge viscosity, and (iv) digestate dewaterability. The full-scale test was conducted according to the following hypothesis:

> ***Hypothesis #3:*** *"Low energy input sonication of thickened waste activated sludge allows for significantly increased methane production, enhanced organic matter removal, lower sludge viscosity, and improved digestate dewaterability in a full-scale wastewater treatment plant."*

The results of this investigation published in **Paper IV** suggest that the full-scale pre-treatment had only minor effect on anaerobic digestion performance. The only parameter that was significantly improved by the US treatment was the apparent viscosity of thickened WAS, which dropped by approximately 6% due to the treatment. However, the resulting cost savings were negligible, as the reduced pumping costs were less than 1‰ of the electricity costs of the US reactor.

For all other parameters tested, US treatment did not lead to a significant improvement.

Only methane production showed a slight enhancement by trend in the last phase of the experiment with a maximum incremental increase in methane yield by 6% relative to the control digester. While the increase was verifiably small, it is interesting to note that already such a small gain allowed to compensate the energy expenses of the US reactor. Nonetheless, the investment costs of the reactor could not be regained, even when assuming a very long reactor lifetime of six years (total investment and operating costs roughly three times larger than savings due to increased methane yield). Thus, the treatment seemed overall not economic, and Hypothesis #3 was ***rejected***. At the same time, it should be noted that the full-scale trial was conducted under rather challenging boundary conditions (TS of WAS of over 6%, HRT of over 40 d, share of WAS to RS less than 40%), which might explain the only moderate effects observed and the unfavorable treatment economics. Nonetheless, the results challenge the majority of the promising lab results (including the results of **Paper I**) and suggest that an application of US treatment to regular, well-performing plants seems not an effective or economically feasible sludge management strategy.

9.4 Research objective #4

Due to the miscellaneous experiences for WAS pre-treatment, the fourth research objective explored ultrasonic co-treatment of DS as an alternative to WAS pre-treatment. The examination was conducted by the use of a continuously operated lab-scale biogas test system comprising a non-sonicated control digester and a sonicated test digester. Operating conditions were closely adapted to the conditions of the WWTP the sludge was sampled from, especially with respect to HRTs (20 - 30 d) and organic loading rates (1.0 - 1.5 $kg_{VS}/(m^3 \cdot d)$). For better comparability to previous studies ([43, 64, 67]) and for presumably stronger treatment effects, a specific energy input of 2,000 kJ/kg_{TS} was selected for the co-treatment. Treatment effects were monitored based on (i) methane production, (ii) (volatile) solids removal, and (iii) digestate dewaterability. The study was conducted according to the following hypothesis:

> ***Hypothesis #4:*** *"Ultrasonic co-treatment of digested sludge economically enhances methane production and (volatile) solids removal, without negative effects on digestate dewaterability."*

The results of this investigation published in **Paper V** demonstrate that US co-treatment was able to significantly enhance both methane production and (volatile) solids removal. On average, daily methane yields increased by 9%, while VS and TS removal increased by approximately 9% and 5%, respectively. At the same time, the cost recovery of the treatment (savings due to additional methane and decreased residual sludge masses vs. electricity costs of the US reactor) remained below 10% for most of the experiment due to the only moderate enhancement and the comparably high energy input. Hence, an economically viable improvement of anaerobic digestion performance was not achieved under the experimental conditions applied. Moreover, sonication caused a significant impairment of digestate dewaterability (increase in normalized CST by almost 20%), which would most likely further deteriorate economic feasibility due to presumably worsened dewatering efficiency. Based on the unsatisfactory treatment economics, Hypothesis #4 was ***rejected***. Therefore, the potential of ultrasonic co-treatment of DS seems similarly limited as for conventional WAS pre-treatment. At the same time, testing of only one (rather high) specific energy input does not allow for general conclusions and an additional testing at low energy inputs is recommended.

Table 9.1: Summary of hypothesis testing.

Hypotheses	Status
Hypothesis #1 "Ultrasonication of waste activated sludge at low specific energy inputs using a novel ultrasonic flatbed reactor leads to significant enhancement of the sludge´s biomethane potential so that the pre-treatment is energy-positive."	Partially accepted
Hypothesis #2.1 "Sludge flow in ultrasound flatbed reactors is mostly laminar despite the impact of cavitation-induced micro-turbulences due to the high viscosity of the sludge."	Accepted
Hypothesis #2.2 "Ultrasonic reactor designs with maximum average cavitation noise levels provide both maximum treatment homogeneity and maximum disintegration efficiency."	Accepted
Hypothesis #3 "Low energy input sonication of thickened waste activated sludge allows for significantly enhanced specific methane production, enhanced organic matter removal, lower sludge viscosity, and improved digestate dewaterability in a full-scale wastewater treatment plant."	Rejected
Hypothesis #4 "Ultrasonic co-treatment of digested sludge leads to economic enhancement of methane production and (volatile) solids removal, without negative effects on digestate dewaterability."	Rejected

CHAPTER 10

Overall discussion

10.1 Potential of tubular US reactors with surface transducers for enhancing anaerobic digestion performance

A novel tubular US reactor with surface transducers was investigated regarding its potential to enhance anaerobic digestion performance. The performance assessment was conducted with respect to potential improvements in (i) methane production, (ii) (volatile) solids removal, (iii) sludge viscosity, and (iv) digestate dewaterability. The key outcomes are presented in the following, while a discussion on the economic implications of the observed treatment effects is provided in Section 10.3.

10.1.1 Effects on methane production

In line with previous research [29, 43, 76], US treatment using the novel tube-shaped US reactor was able to enhance the methane production of sewage sludge. Yet, the achieved enhancements were comparably low, and the highest increase in methane yield observed amounted to only 12% (**Paper I**). This result was furthermore obtained using batch tests, which were shown to overrate treatment effects as compared to continuous anaerobic digestion tests [92–94]. Confirming this appraisal, the methane production enhancement observed in the continuous full-scale test on WAS pre-treatment was even smaller and amounted to a maximum (yet insignificant) increase of 6% (**Paper IV**). The continuous test on DS co-treatment yielded significant, but similarly moderate average enhancements of only 6% (**Paper V**). Based on those relatively small and partially insignificant increases, the potential of US treatment for enhancing methane production seems, therefore, limited.

10.1.2 Effects on (volatile) solids removal

The effect of US treatment on (volatile) solids removal was assessed for full-scale WAS pre-treatment (**Paper IV**) and lab-scale DS co-treatment (**Paper V**). In the latter case, US treatment led to a significant increase in organic matter removal (+9%, on average) and, accordingly, to a significant reduction of the TS content of the digestate (-5%, on average). Yet, the enhancement was rather low, despite a relatively high energy input of 2,000 kJ/kg_{TS}. In the case of the full-scale test at the Starnberg WWTP, US pre-treatment had no measurable impact on (volatile) solids removal (corresponding to the only insignificant increase in methane production) so that savings due to reduced residual sludge masses could not be realized. Based on this outcome, the potential of US treatment for stimulating organic matter degradation seems limited.

10.1.3 Effects on sludge viscosity

The potential of US treatment to reduce sludge viscosity was assessed for the full-scale test at the Starnberg WWTP. The obtained results demonstrated that sonication led to a significant, but only minor decrease in WAS viscosity by approximately 6% (**Paper IV**). Given the small improvement, neither pumping nor stirring costs could be notably reduced (reduced pumping costs less than 1‰ of the electricity costs of the US reactor). An increased biogas production due to better mixing of the less viscous WAS and DS (as suggested by Amani et al. [37]) could not be realized, either.

10.1.4 Effects on digestate dewaterability

The impact of US treatment on digestate dewaterability was examined for both the full-scale test on WAS pre-treatment and the continuous test on DS co-treatment. In the full-scale test, US pre-treatment did not cause any effect on digestate dewaterability (**Paper IV**). Hence, a negative impact of sonication on dewatering performance is not expected. Such a result agrees with previous literature, indicating that low energy inputs are not detrimental to digestate dewaterability [73, 177]. It seems that for low energy inputs feasible at full-scale plants, changes in digestate dewaterability are relatively unlikely (i.e., neither improvements nor impairments). In the case of DS co-treatment, on the other hand, the relatively high energy input of 2,000 kJ/kg_{TS} led to a significant increase in normalized CST values by almost 20% (**Paper V**), thus indicating a significant impairment of digestate dewaterability. While the effect could not be included in the economic assessment due to the limited transferability of CST tests to centrifugation performance [185], it is likely that the elevated CST values would have entailed a negative impact on the dewatering process, and, thus, operating costs.

10.1.5 Overall potential

Overall, the results obtained suggest that US treatment is a relatively ineffective measure for enhancing anaerobic digestion performance of well-functioning digesters. Methane yields and (volatile) solids removal were significantly increased, albeit to a very limited extent. The effects on sludge viscosity were significant but still small and could not entail beneficial effects on anaerobic digestion performance. Effects on dewaterability were either negative or insignificant, indicating that treatments at higher energy inputs might even have adverse side effects. The potential of US treatment for enhancing anaerobic digestion performance seems, therefore, limited. At the same time, the conducted experiments are by no means exhaustive, and US treatment might be an effective strategy for other conditions or fields of application that were not covered in this dissertation.

10.2 Operational aspects of the novel reactor type

10.2.1 Installation and space requirements

The novel tube-shaped reactor system allowed for similarly compact reactor dimensions as conventional sonotrode or radial horn reactors, and the full-scale reactor installed at the Starnberg WWTP required only small space requirements of about 20 m^2. The retrofitting of the plant was, hence, easily feasible. A photograph of the installed setup is depicted in Figure 10.1.

Due to the modular design of the reactor systems, the installation process was moreover relatively easy to conduct and required only moderate labor. The complete installation of the reactor setup was realized within 2 - 3 working days.

10.2.2 Resistance to clogging

Due to the unobstructed, tube-shaped reactor layout, clogging risk could be effectively minimized. During the full-scale trial, no clogging event was encountered over an experimental period of 12 months, despite the high TS content of the treated WAS (> 6%) and the omission of reactor cleaning throughout the experimental period (**Paper IV**). Thus, concerning clogging risk, results clearly suggest that tube-shaped US reactors enable trouble-free treatment of sewage sludge at full-scale WWTPs.

Figure 10.1: Photograph of the full-scale US reactor system depicting individual reactors, pipes, and the control cabinet at the Starnberg wastewater treatment plant.

10.2.3 Susceptibility to cavitation erosion

The double-tube reactor employed at the Starnberg WWTP exhibited an erosion-induced micro-leakage after approximately one year of operation (or 1,440 hours, according to the operating time of 4 hours per day). Due to the irreversible leakage, the full-scale trial had to be terminated. After cutting the reactor open for inspection, severe pitting could be detected throughout the reactor´s inner surfaces, as can be seen in Figure 10.2.

This result challenges the appraisal that low-intensity US reactors with surface transducers feature improved resistance to erosion as compared to high-intensity sonotrode or radial horn reactors. Moreover, in contrast to sonotrode reactors, where a reactor refurbishment can be conducted by simply replacing eroded sonotrode tips, surface transducer systems need to be replaced entirely once erosion pitting occurs. In the present example, this would entail yearly maintenance costs (i.e., reactor replacement costs) of approximately 100,000 €, which by far exceeds typical annual replacement costs for sonotrodes (typically several thousand € per year, assuming costs of about 700 € per sonotrode). From this point of view, the full-scale applicability of tube-shaped US reactors in their current form is questionable. A clear

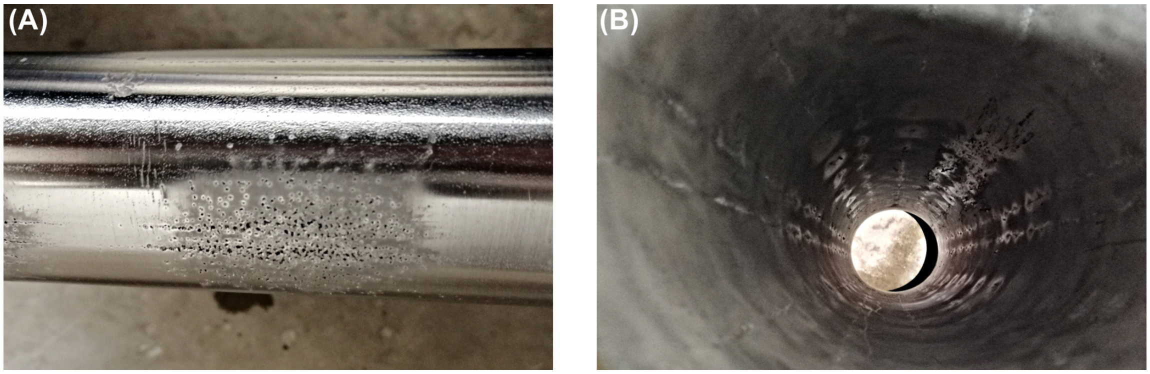

Figure 10.2: Erosion damage on the inner (A) and on the outer transducer surfaces (B) of the full-scale ultrasound reactor employed at the Starnberg wastewater treatment plant.

understanding of the relationship between substrate properties (especially TS content) and erosion susceptibility and further research on lifetime-enhancing reactor modifications (such as protective coatings of the sound-emitting surfaces) is, therefore, of crucial importance.

10.3 Economics of (full-scale) US treatment

Based on the only moderate treatment effects and the high erosion susceptibility, an economic application of ultrasonic sludge treatment using the novel reactor type seems challenging.

The results of the conducted lab-scale studies (**Paper I** and **Paper V**) suggest that an economic and energy-positive application of US treatment is only possible at low specific energy inputs and low electricity consumption. For instance, results of **Paper I** demonstrated that an energy-positive WAS pre-treatment was only possible at the lowest energy input tested (200 kJ/kg_{TS}), while all other inputs (400 kJ/kg_{TS} to 3,000 kJ/kg_{TS}) consumed more energy than was regained through additional methane for three different WAS samples tested. Results of **Paper V** confirmed such outcome and indicated that the chosen energy input of 2,000 kJ/kg_{TS} for DS co-treatment was far too high for economic reactor operation, and electricity costs were roughly 20 times higher than the benefits due to additional methane and reduced residual biosolids for disposal. Moreover, it should be noted that above mentioned lab-scale studies did not consider investment and maintenance costs for the US reactors (which are naturally directly proportional to the chosen energy input) so that overall treatment economics might have been even worse.

According to this outcome, the full-scale test at the Starnberg WWTP was conducted at a low specific energy input of 200 kJ/kg_{TS} (**Paper IV**). Yet, the results of the case study demonstrate that even such low energy input already requires a quite powerful reactor system

(13.5 kW), thus causing investment costs of roughly 100,000 € (assuming 7,500 €/kW) and yearly electricity costs of about 4,000 € (assuming an industrial electricity price of 21 Cent/kWh and 4 h of operation per day). Given the observed short lifetime of the reactors of only one year, this would add up to annual operation and maintenance costs of 104,000 €. The result makes clear that the short lifetime of the novel US reactors (and the necessity to replace eroded systems completely) is financially much more challenging for operators than the electricity consumption of the US reactors, thus emphasizing that further research on wear resistance is crucial.

The economic assessment presented in **Paper IV** demonstrated that such cost ranges cannot be regained through the realized treatment effects. For instance, to compensate for both electricity and reactor replacement costs (i.e., 104,000 € per year), a constant percentage increase of methane yield by roughly 30% would have been necessary. Such value is, however, in strong contrast to the actual (and insignificant) increase of a maximum of 6%. To fully compensate for the investment and electricity costs at such low methane yield increases, a reactor lifetime of more than 6 years would have been necessary. This seems, however, relatively unlikely, especially when considering that operating times during the test phase amounted to only 4 h per day. In normal operation, operating times of at least 8 h (or even 24 h in case a permanent WAS feeding is conducted) seem realistic, which would presumably further shorten the reactor lifetime.

Regarding other potential merits of full-scale US treatment (such as decreased sludge viscosity or improved digestate dewaterability), results of **Paper IV** demonstrate that slightly reduced WAS viscosity (6%) is economically irrelevant when it does not translate into reduced viscosity of the digester content. Concerning potentially improved digestate dewaterability, the findings of **Paper IV** suggest that full-scale US treatment has no measurable impact on CST values so that savings due to improved dewatering performance cannot be expected.

All the same, it should be considered that the boundary conditions at the Starnberg WWTP were somewhat challenging for conducting a pre-treatment (low energy input, high substrate TS, and long HRTs). On the other hand, the conditions were also not far off from other WWTPs (see survey among 17 plants in **Paper IV**) so that the obtained results may also apply to other, rather typical plants. In consequence, results suggest that US treatment may not be economically viable at WWTPs with well-performing anaerobic digesters.

Thus, the most likely scenario for an economic application of the technology seems US treatment for poorly performing digesters operating at their capacity limit. Assuming that the implementation of a US reactor could maintain proper digester operation despite shortened HRTs and avoid the construction of an additional digester, the treatment may be economically feasible, even at negative energy balances. Yet, without such impending multi-million € investment that could be avoided through ultrasonic sludge treatment, an economic application of the technology seems highly challenging.

10.4 Possible explanations for the limited performance

By indicating that US treatment is a rather ineffective and uneconomic measure to enhance anaerobic digestion performance, obtained results are in contradiction to many previous studies (see Chapter 2). In the following, potential explanations for the differing results are elucidated.

10.4.1 Low US intensity of the novel reactor type

As all hitherto full-scale studies and the majority of the lab-scale studies were performed using high-intensity sonotrode or radial horn reactors [4, 10, 19, 45, 84, 111], the application of low-intensity US reactors with surface transducers appears to be a likely reason for the only moderate enhancement. In contrast to sonotrodes and radial horn reactors which exhibit US intensities of up to 50 W/cm^2 [45, 97], surface transducer systems are typically limited to US intensities of less than 1 W/cm^2 [76], thus leading to comparably low oscillation amplitudes (6 – 8 μm wall oscillation vs. 60 – 80 μm sonotrode tip oscillation, [43]). Such low intensity (and oscillation amplitude) could be detrimental to sonication efficiency, as Neis et al. [99] reported an almost linear relationship between US intensity and COD solubilization.

At the same time, Bandelin et al. [145] have recently shown that the performance of low-intensity tube reactors is not categorically different from high-intensity sonotrode reactors. At low energy inputs, tube reactors even achieved stronger COD solubilization than the tested sonotrode reactor. Possibly, the more homogeneous induction of US waves through multi-transducer systems in tube reactors is able to compensate for the lower intensity. Such finding is substantiated by further lab-scale studies, which similarly found significant enhancement of methane yields through US treatment with tube reactors [29, 43, 76]. Thus, the application of novel US reactors with surface transducers may be a contributor, but not the main reason for the limited performance observed.

10.4.2 Application of low, industrially feasible energy inputs

For obtaining results with high transferability to full-scale US treatment, a relatively low range of specific energy inputs was examined. Thus, energy inputs of around 200 kJ/kg_{TS} were employed in most of the conducted studies (**Papers I, II, III**, and **IV**), which is in line with energy inputs reported in previous full-scale trials (for instance, approximately 400 kJ/kg_{TS} in

Xie et al. [84], or 150 kJ/kg_{TS} in Neis [45]). Yet, such input is considerably lower than typical energy inputs of previous lab-scale studies, which mostly ranged between 1,000 kJ/kg_{TS} and 16,000 kJ/kg_{TS} [10, 26], or sometimes even reached values above 100,000 kJ/kg_{TS} [80, 86]. Regarding this distinct offset, it seems that the low inputs applied are an obvious reason for the moderate enhancements observed. Even the 'high' potential of low energy inputs (for instance, 12% BMP increase at 200 kJ/kg_{TS}) can still be regarded as low when compared to previously observed BMP increases between 20% and 140% at high energy inputs [4, 10]. (Naturally, a BMP increase as high as 140% strongly suggests that the digestion performance of the non-sonicated blank was severely compromised in the first place so that the transferability to well-functioning digesters is fairly limited anyway.)

In the light of this offset, it must be emphasized that treatments with several thousand kJ/kg_{TS} have nearly no relevance for full-scale applications. For instance, a verifiably low lab-scale input of 1,000 kJ/kg_{TS} would have already necessitated a reactor system with a total electric power of approximately 70 kW at the Starnberg WWTP, thus causing investment costs of roughly 500,000 € and yearly electricity costs of about 44,000 €. Considering again a reactor lifetime of approximately one year, annual operation and reactor replacement costs would amount to 544,000 €. To recover these costs through enhanced methane production, a percentage increase in methane yield of about 150% would have been necessary. The example illustrates that even a comparably low lab-scale energy input of 1,000 kJ/kg_{TS} would already require extraordinary, or even unrealistic enhancements of digester performance at full-scale WWTPs. Besides, it was shown in **Paper I** that an energy input of 1,000 kJ/kg_{TS} showed the weakest BMP enhancement within a range of energy inputs from 200 kJ/kg_{TS} to 3,000 kJ/kg_{TS} so that presumably, much higher energy inputs are needed to take full advantage of the effects of high energy input sonication. Naturally, this would drive investment and operating costs even further away from economic viability. In addition to the financial aspects, the installation of a 70 kW reactor would entail considerable space requirements, which would challenge the retrofitting of existing plants.

Given this offset in energy input, it seems furthermore worth mentioning that lab- and full-scale sonications are based on different working principles. Low energy inputs mostly utilize effects of floc de-agglomeration [25, 72], while only high energy inputs (above 1,000 kJ/kg_{TS}) were reported to cause notable cell lysis [10, 26, 81]. Thus, studying high ranges of energy input is not only detached from economic and technical feasibility, but it also investigates a functional principle that cannot be attained at full-scale WWTPs. To develop US reactors applicable in full-scale plants, future research should put a clear focus on low, industrially feasible energy inputs.

10.4.3 Treatment of sludges with a high TS content

Another critical boundary condition that may have contributed to the low performance enhancement observed is the high solids concentration of the treated substrates of up to 7% (**Paper I**), as previous lab-scale studies on WAS pre-treatment were mostly carried out using dilute WAS samples with TS contents of about 2% (see reviews by Tyagi et al. [19] and Le et al. [12]). Such range is, however, unrealistic at full-scale WWTPs, as thickened WAS typically exhibits TS contents of above 5% [36, 105, 133]. At the same time, it is well-known that sonication efficiency declines above a certain solids content due to sound wave attenuation, and, for instance, Show et al. [14] demonstrated that COD solubilization already decreases at TS contents above 3.2%. Thus, just like in full-scale ultrasonic WAS pre-treatment, most of the conducted studies were outside the ideal TS range for US treatment (2.3% to 3.2% [14]).

The results of **Paper II** moreover demonstrate that the high TS content of sewage sludge has a defining impact on the fluid dynamics, as the high viscosity of the solids-rich substrate almost completely suppresses cavitation-induced flow disturbances. Therefore, an inhomogeneous cavitation field is not compensated for by fluid mixing during US treatments, thus causing an inhomogeneous exposure of the sludge to cavitation.

The findings in **Paper III** confirmed that the high TS levels of thickened WAS cause severe sound wave attenuation, which substantiates that high TS contents are detrimental to US treatments and that cavitation fields in US reactors are, in fact, highly inhomogeneous. At a distance of less than 50 mm to the transducer surface, the sludge remained completely unaffected by the treatment. It has to be noted that such sharp decline in cavitation intensity was already encountered at a TS concentration of around 4.6%, while TS contents of thickened WAS can easily reach values of above 6% (**Papers I** and **IV**, and [36, 105, 133]). Thus, large parts of the treated sludge might not experience notable cavitation intensity, which especially applies to up-scaled US reactors for industrial use, where small reactor gaps (such as 20 mm or 40 mm) are hardly feasible due to clogging risk or overly high pressure losses.

The gained operational experience during the full-scale test at the Starnberg WWTP (**Paper IV**) moreover showed that the tube-shaped US reactor was subjected to severe erosion damage, despite the lower ultrasonic intensity as compared to sonotrode systems. The occurrence of the severe erosion patterns in the low-intensity system may also be explained by the high solids content of the treated WAS, as solids-rich substrates are well-known to promote early-onset erosion damage in US reactors [188, 189]. Besides, the strong sound wave attenuation prevalent during WAS sonication concentrates the destructive forces of cavitation events to the close vicinity of the reactor walls, thus further promoting fast reactor erosion.

Hence, with respect to treatment homogeneity, sonication efficiency, and resistance to erosion, the obtained results strongly suggest that sonication of substrates with lower TS concentrations is highly recommendable and much better adapted to the requirements of ultrasonic treatment.

10.4.4 Examination of digesters with long HRTs

It is well-known that the effectiveness of sludge pre-treatment decreases with increasing HRTs, as an acceleration of reaction kinetics becomes less important once sufficient digester capacity is available [10, 26]. Accordingly, most previous studies examined the potential of US treatment at comparably low HRTs, such as 7.5 d - 15 d in [70], 4 d - 16 d in [99], or 10 d - 20 d in [91]. Thus, it seems that the selected HRTs of $>$ 40 d (**Paper IV**) and 20 d - 30 d (**Paper V**) were already too high for achieving strong treatment effects. On the other hand, the conducted field survey among 17 south German WWTPs (**Paper IV**) revealed that average digester HRTs amounted to 33 days and that HRTs below 25 days were only observed in 3 out of 17 plants. Hence, for obtaining results that are transferable to full-scale WWTPs, selection of comparably long HRTs as test condition seems justified (at least with regard to the group of WWTPs surveyed).

10.5 Future research needs

10.5.1 In-depth investigation of the potential of low energy input sonication

The economically interesting result of **Paper I** that low energy input sonication leads to the same BMP increase as high energy input sonication should get further attention, as well as the performance drop at intermediate inputs at around 1,000 kJ/kg_{TS}, where no significant improvement was achieved. A similar pattern was recently observed by Bandelin et al. [145], who realized incremental BMP enhancements of up to 25% at 300 kJ/kg_{TS}, but only of 6% and 11% at energy inputs of 437 kJ/kg_{TS} and 3,500 kJ/kg_{TS}, respectively.

Both the high performance at low energy inputs and the impaired performance at intermediate inputs are highly relevant with respect to full-scale sonication, which takes place at exactly such low ranges of energy input. Possibly, the distinct BMP enhancement at low energy input can be explained by the de-agglomeration of sludge flocs and the accordingly increased surface area. To test this hypothesis, BMP tests could be accompanied by microscopy investigations of the treated sludge or by laser diffraction measurements for determining the particle size distribution after treatment [35, 190]. The same techniques could be applied to test whether the impaired performance at intermediate inputs is a cause of re-flocculating effects at the onset of cell lysis, as was hypothesized in **Paper I**. A clear understanding of the 'performance gap' at intermediate inputs is seen as crucial, as it could help to avoid such potentially uneconomic ranges of energy input in full-scale plants. For obtaining more transferable results, the potential of low energy input sonication should furthermore be reproduced in a continuously operated digester system.

10.5.2 Systematic optimization of reactor design for different substrates

Results of **Paper III** demonstrated that hydrophone measurements are a suitable tool for optimizing US reactor design. Thus, the existing setup could be further used to investigate the relationship between sludge properties (especially TS content), reactor specifications (such as US intensity) and optimum reactor geometry, in order to develop a design guideline for a wide variety of substrates. Such data set could moreover be of great use for modeling sound wave attenuation as a function of substrate TS content and US intensity. Once such relationship would be established, a computer-based optimization of US reactor design could be conducted based on substrate properties for any kind of reactor type.

10.5.3 Investigation of the effect of elevated operating pressures

To date, most studies on sewage sludge disintegration are conducted at ambient pressure [12]. Yet, in sludge pipes at WWTPs, pressure levels are commonly slightly elevated and amount to approximately 1 - 2 bar [125]. At such elevated pressures, however, the violence of the bubble implosions might be enhanced, leading to higher temperatures, pressures, and an increased release of shear forces during collapse events. At overly high pressures, on the other hand, bubble formation (or bubble growth) is impaired due to an increased cavitation threshold. Thus, there seems to be an optimum pressure for every sonication condition [191].

In their study, Le et al. [192] investigated the impact of hydrostatic pressure (0 - 16 bar) on the degree of sludge disintegration for various specific energy inputs (7,000 - 75,000 kJ/kg_{TS}). Independent of the specific energy input applied, the strongest DD_{COD} was constantly observed at a slight overpressure of 2 bar. Compared to ambient pressure, performance enhancements of over 50% were attained due to such optimized pressure conditions [192]. The finding of a performance optimum at intermediate pressure levels is substantiated by a study of Bandelin et al. [125], who reported that cavitation noise in US reactors increased at an overpressure of 1 bar but decreased once the pressure was elevated above 2 bar.

While the results seem promising, all hitherto studies on pressure optimization were only conducted using surrogates parameters for sonication efficiency (such as DD_{COD} or cavitation noise), while especially DD_{COD} was shown to be a relatively unreliable parameter to predict sonication efficiency [103, 133]. Thus, for a more holistic assessment of the impact of pressure, further studies employing BMP or continuous tests seem a logical and worthwhile next step. A pressure-driven performance enhancement of ultrasonic sludge treatment seems particularly advantageous, as sludge pipes in WWTPs seem to already provide favorable pressure conditions, at no added cost and without further engineering effort. At the same time, it should be noted that the pressure conditions at the Starnberg WWTP (approximately 1.5 bar) were already at such favorable range, without causing a notable performance boost. Given the boundary conditions at the plant (long HRT, high substrate TS), it is, however, also a possibility that the potential pressure-driven performance enhancement was rendered non-detectable. Overall, it seems that elevated operating pressures certainly do not guarantee an effective US treatment, but an already well-performing treatment process may be further improved through optimum pressure conditions.

10.5.4 Conduction of lab-scale studies at full-scale conditions

For a meaningful assessment of the potential of US treatments, future lab-scale studies should be conducted with experimental settings that would also be possible at full-scale WWTPs. Especially the selection of specific energy inputs and sludge solids content should be adapted more closely to full-scale conditions (i.e., $< 1{,}000\ kJ/kg_{TS}$ and $> 5\%$, respectively), in order to obtain more meaningful and transferable results. As numerous research articles have already demonstrated the potential of US treatment [4, 10, 12, 19], the key research question for future studies is not whether US treatment can significantly enhance anaerobic digestion performance or not, but whether this enhancement can be achieved economically and under conditions feasible in full-scale plants.

10.5.5 Conduction of full-scale tests under favorable boundary conditions

By selecting the WWTP Starnberg for the conducted full-scale test, the potential of tube-shaped reactors was tested according to Frank Sinatra´s New-York-principle ("*If I can make it there, I´ll make it anywhere*"). A high TS content of the thickened WAS of over 6%, a long average HRT of over 40 days, and a comparably low share of WAS to RS of less than 40% created rather challenging test conditions. By showing only an insignificant enhancement of anaerobic digestion performance and unfavorable treatment economics (**Paper IV**), the results of the full-scale test suggest that US treatment is not a viable option for well-performing WWTPs.

Thus, with regard to the experimental costs, time, and labor force, it might be worthwhile to consider more favorable boundary conditions for future full-scale trials. Hereby, especially plants with short HRTs and a low TS content of WAS or DS might represent well-suited test locations. Based on the obtained experience, high controllability of the digesters and highly dedicated plant personnel are further critical requirements for a meaningful test over a sufficiently long experimental phase.

Generally, the importance and the research need for further full-scale trials cannot be overemphasized, as it is the only possibility to reliably determine operational performance and treatment economics. At present, peer-reviewed literature on full-scale trials is, however, scarce, and a study clearly confirming the potential of US treatment to enhance anaerobic digestion performance under full-scale conditions is yet to be conducted.

10.6 Alternative approaches

While the potential of tube-shaped US reactors with surface transducers seems limited for sewage sludge pre- or co-treatment, the technology might be well-suited for other areas of wastewater treatment. Especially substrates with a lower TS concentration and smaller treatment volumes might be more amenable to US treatment.

10.6.1 Enhancement of biological nitrogen removal

Biological nitrogen removal at WWTPs traditionally comprises a nitrification and a denitrification step. Nitrification is a two-stage biological oxidation process, comprising an initial and rate-limiting oxidation step where ammonium (NH_4^+) is oxidized to nitrite (NO_2^-) through the activity of ammonia-oxidizing bacteria (AOB), and a subsequent and rapidly occurring step where nitrite is oxidized to nitrate (NO_3^-) through the activity of nitrite-oxidizing bacteria (NOB). In the subsequent denitrification step, nitrate is successively reduced to gaseous nitrogen and removed from the wastewater [193].

A common approach to render the above process more energy- and resource-efficient is a switch from nitrification to partial nitrification, i.e., to prevent the oxidation of nitrite to nitrate [194]. Benefits of partial nitrification are, for instance, an increase of nitrate reduction rates, a reduction of oxygen demand (by up to 25%), and a significant reduction of biomass generation in the subsequent denitrification process [193]. The key to attain partial nitrification is the enhancement of AOB activity and the simultaneous impairment of NOB activity. Among several control strategies for achieving this goal (including regulation of temperature, pH, or dissolved oxygen concentrations [193]), US has gained weight as another effective control strategy [162, 163, 195].

Through cavitational effects, US was reported to selectively enhance the enzymatic activity of ammonia monooxygenase and hydroxylamine dehydrogenase (i.e., enzymes involved in the oxidation of ammonium to nitrite), which has been shown to have beneficial effects on AOB activity [163, 196]. NOB activity, on the other hand, was found to be effectively impaired through ultrasonic irradiation, as the NOB community is more sensitive towards US-induced cell disruption than the AOB community [197, 198]. Thus, US seems a well-suited control strategy for partial nitrification. Moreover, non-thickened activated sludge offers more favorable treatment conditions than thickened WAS, due to the considerably lower TS content and the accordingly lower sound wave attenuation and treatment inhomogeneity. At the same time, hitherto lab-scale studies similarly employed relatively high energy inputs (for instance, 66,000 kJ per kg of mixed liquor suspended solids [194]) so that an economic application in full-scale WWTPs seems again challenging. A careful optimization of energy inputs and an alignment of experimental conditions to full-scale feasibility seems, therefore, also relevant for such alternative US treatment approach.

10.6.2 Digestate post-treatment

While several studies already examined how ultrasonic pre-treatment affects sludge dewaterability (both directly after sonication and after the anaerobic digestion of the sonicated sludge) [73, 82, 83, 102, 177, 199], the use of US as a digestate post-treatment for enhanced dewaterability is a relatively novel approach. Several post-treatment schemes were tested and, for instance, Gallego-Juarez [200] examined the potential of US-assisted sludge dewatering for a rotary vacuum filter and found that the application of short US irradiation (2 s) already increased the TS content of the filter cake by 6% relative to the untreated sample. The author claimed that the alternating pressure cycles under US irradiation squeezed the sludge like a sponge for an enhanced water release (sponge effect) and created channels in the filter cake for an improved water efflux (channel effect). Mobaraki et al. [201] investigated US-assisted drying by the use of a laboratory dryer setup with vacuum suction, hot air convection, and an ultrasonic vibrating plate. In all examined process combinations, US-irradiation was found to be beneficial with respect to drying performance. Zhu et al. [164] studied a process combination more closely related to wastewater treatment and employed mild US irradiation to the sludge before adding a cationic polyacrylamide coagulant and rice husk for a re-flocculation of the dispersed sludge and a skeleton building for enhanced filter cake permeability, respectively. The authors demonstrated that such process combination led to significantly enhanced dewatering performance as compared to non-conditioned sludge, already at short sonication times of 12 s and an ultrasonic density of 300 W/L. Assuming a solids content of the digestate of 3%, the specific energy input would amount to only 120 kJ/kg_{TS}. The results were recently confirmed by Bień and Bień [165] who found that the strongest reduction of CST values was obtained when dosing coagulants to a previously sonicated sludge sample. Unfortunately, no information on the electrical power of the US sonotrode reactor was given so that the energy input could not be calculated. Both studies claimed that the stripping of bound water from the flocs and the release of interstitial water from the sludge flocs through US irradiation caused the enhanced dewatering performance [164, 165].

The process combination of US and coagulant dosage has recently been commercialized by the company VTA Austria GmbH (Rottenbach, Austria). According to the manufacturer, the initial sonication of the sludge (by the use of a US reactor termed mudinator®) followed by coagulant dosage enables an increase of TS concentration of the dewatered residue by up to 5 percentage points (see http://www.vta.cc/de/Anlagentechnik/VTA-mudinator%C2%AE).

While such promising outlook has not been confirmed by peer-reviewed scientific literature yet, the concept certainly justifies further investigation, due to the highly favorable boundary conditions for US treatment. First, the treatment scheme takes advantage of the low TS concentration of DS, just like co- or inter-treatments. Second, the approach relies on both economic and industrially feasible low energy inputs (such as 120 kJ/kg_{TS}, calculated based on data from Zhu et al. [164]), as full disintegration of the sludge would not entail an improvement, but an impairment of dewaterability [82, 102, 199]. Third, the effectivity of the treatment can be investigated right after the post-treatment using lab-scale or full-scale centrifuges, without being subjected to long waiting times as for pre- or co-treatments. In case the innovative treatment scheme holds up against thorough scientific testing, it would certainly be a useful addition to existing sludge management strategies.

10.6.3 Industrial wastewater treatment

Another field of application for US reactors might be industrial wastewater treatment. By causing sonochemical pyrolysis, US could be a promising treatment technology for substances that cannot be effectively degraded by conventional biological processes, or chemical treatments including advanced oxidation processes (AOPs). For instance, emerging contaminants such as poly- and perfluorinated alkyl substances (PFASs) were reported to be recalcitrant towards chemical and biological degradation, as well as towards oxidative processes using ozone (O_3), hydrogen peroxide (H_2O_2), or combined approaches utilizing Fenton´s reagent (H_2O_2/Fe^{2+}) [202, 203].

For such persistent organic pollutants, the extreme heat generated during cavitation bubble collapses could be an effective means of treatment. As waste streams containing PFASs (such as perfluorooctanesulfonate, PFOS or perfluorooctanoate, PFOA) are typically aqueous solutions that are not causing notable sound wave attenuation [204, 205], US treatment could unfold its unmitigated potential. Besides, as PFASs are strong surfactants, they tend to accumulate at the bubble-liquid-interface so that the contaminant is efficiently concentrated at the pyrolytic cavitation sites [202, 203]. In contrast to sewage sludge treatment, PFAS sonication would, therefore, occur under highly favorable boundary conditions. Accordingly, promising results were already obtained in lab-scale studies, and, for instance, Panda et al. [206] reported a complete mineralization of PFOS and PFOA after 80 min of treatment (at energy inputs of 7,200 - 18,000 kJ per liter of PFAS-solution). Similar results were obtained by Lin et al. [205] who achieved an almost complete decomposition of PFOA after 90 min of ultrasonic irradiation (at 2,700 kJ per liter of PFAS-solution). From a technical point of view, even such long treatment times and high energy inputs seem feasible, due to the comparably low production volumes of PFAS-containing wastewaters (as compared to daily amounts of sewage sludge). Yet, with respect to treatment economics, a benchmarking of US-induced PFAS degradation against other degradation methods (such as thermolysis or photocatalysis [207]) should be conducted. Based on the promising lab results and the globally emerging challenges arising from PFAS contamination, the use of US as treatment strategy should certainly get further attention in the future.

CHAPTER 11

Overall conclusion and outlook

The obtained results suggest that the treatment of sewage sludge using novel tubular US reactors with surface transducers has limited potential for enhancing anaerobic digestion performance. Due to the only moderate enhancement of methane yield (+ 12% at maximum), solids removal (-5% on average), and sludge viscosity (-6% on average), and the limited lifetime of the US reactors (approximately one year or 1,440 hours of constant operation), conducted US treatments were found to be largely uneconomical.

Also, the optimization of specific energy inputs (low inputs seemed equally performant as high energy inputs) and geometric reactor design (as small as possible, as large as necessary) did not result in an impactful, economic treatment.

A possible explanation for the only moderate potential may be the clear focus on industrially feasible treatment conditions. In contrast to many previous lab-scale studies, selected experimental settings were restricted to mostly low specific energy inputs ($< 1{,}000$ kJ/kg_{TS}), high substrate TS contents ($> 5\%$), and relatively long HRTs in the digesters (20 d - 40 d). The findings suggest that an economic application of ultrasonic sewage sludge disintegration is highly challenging, at least for common WWTPs with largely typical operating conditions.

Thus, future research should put a clear focus on underperforming WWTPs with digesters operating at capacity limit. Assuming that a pre- or co-treatment can avoid the expensive construction of an additional digester, US treatment may be economic despite a negative energy balance and a short reactor lifetime. Future lab-scale research should be aligned more closely to full-scale conditions (especially with regard to applied energy inputs) in order to obtain more meaningful and transferable results.

Further possible applications fields with possibly more amenable boundary conditions for US treatment may be (i) US-assisted nitrogen removal in biological wastewater treatment, (ii) digestate post-treatment for enhanced dewatering efficiency, and (iii) sonication of industrial wastewaters for pyrolytic degradation of persistent contaminants.

APPENDIX A

List of publications

A.1 List of peer-reviewed journal articles

A.1.1 First-author contributions

Paper I:

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

Paper II:

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

Paper III:

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

Paper IV:

Lippert, T., Bandelin, J., Vogl, D., Alipour Tesieh, Z., Wild, T., Drewes, J.E., Koch, K. (2021) Full-scale assessment of ultrasonic sewage sludge pre-treatment using a novel double-tube reactor. *ACS ES&T Engineering*, 1, 298-309.

Paper V:

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

A.1.2 Second-author contributions

Paper VI:

Koch, K., Lippert, T., Hauck Sabadini, N., Drewes, J.E. (2017) Tube reactors as a novel ultrasonication system for trouble-free treatment of sludges. *Ultrasonics Sonochemistry* 37, 464-470.

Paper VII:

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

Paper VIII:

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

Paper IX:

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

A.1.3 Additional peer-reviewed journal articles outside of the study focus

Paper X:

Koch, K., Lippert, T., Drewes, J.E. (2017) The role of inoculum's origin on the methane yield of different substrates in biochemical methane potential (BMP) tests. *Bioresource Technology* 243, 457-463.

Paper XI:

Horstmeyer, N., Lippert, T., Schön, D., Schlederer, F., Picioreanu, C., Achterhold, K., Pfeiffer, F., Drewes, J.E. (2018) CT scanning of membrane feed spacers – Impact of spacer model accuracy on hydrodynamic and solute transport modeling in membrane feed channels. *Journal of Membrane Science* 564, 133-145.

Paper XII:

Horstmeyer, N., Thies, C., Lippert, T., Drewes, J.E. (2020) A hydraulically optimized fluidized bed UF membrane reactor (FB-UF-MR) for direct treatment of raw municipal wastewater to enable water reclamation with integrated energy recovery. *Separation and Purification Technology* 235, 116165.

A.2 Conference contributions

A.2.1 Oral presentations

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

Bandelin, J., Lippert, T., Drewes, J.E., Koch, K.: Cavitation field analysis for a more effective substrate pre-treatment with ultrasound, *The 15th IWA Conference on Anaerobic Digestion 2017*, Beijing, China.

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

Lippert, T., Bandelin, J., Koch, K., Drewes, J.E.: Ökonomie der Klärschlammdesintegration mittels Ultraschall. *46. Abwassertechnisches Seminar 2018*, Ismaning, Germany.

Bandelin, J., Lippert, T., Koch, K., Drewes, J.E.: Energieeffiziente Klärschlammvorbehandlung durch innovative Ultraschall-Desintegration, *46. Abwassertechnisches Seminar 2018*, Ismaning, Germany.

Lippert, T., Bandelin, J., Drewes, J.E., Koch, K.: „Power-to-cavitation (PTC)“: Proposing a new parameter to assess the energy-efficiency of ultrasonic reactors for sewage sludge pretreatment. *Biogas Science 2018, International Conference on Anaerobic Digestion*, Torino, Italy.

Koch, K., Lippert, T., Drewes, J.E.: The role of inoculum's origin on the methane yield of different substrates in BMP tests *Biogas Science 2018, International Conference on Anaerobic Digestion*, Torino, Italy.

A.2.2 Poster presentations

Lippert, T., Drewes, J.E., Koch, K.: Ultrasonic treatment of sewage sludge - Principles, experimental setup, research targets (poster with short presentation). *Doctoral Candidates Day 2017*, Department of Civil Geo and Environmental Engineering, Technical University of Munich.

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

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

APPENDIX B

Supplementary material Paper I

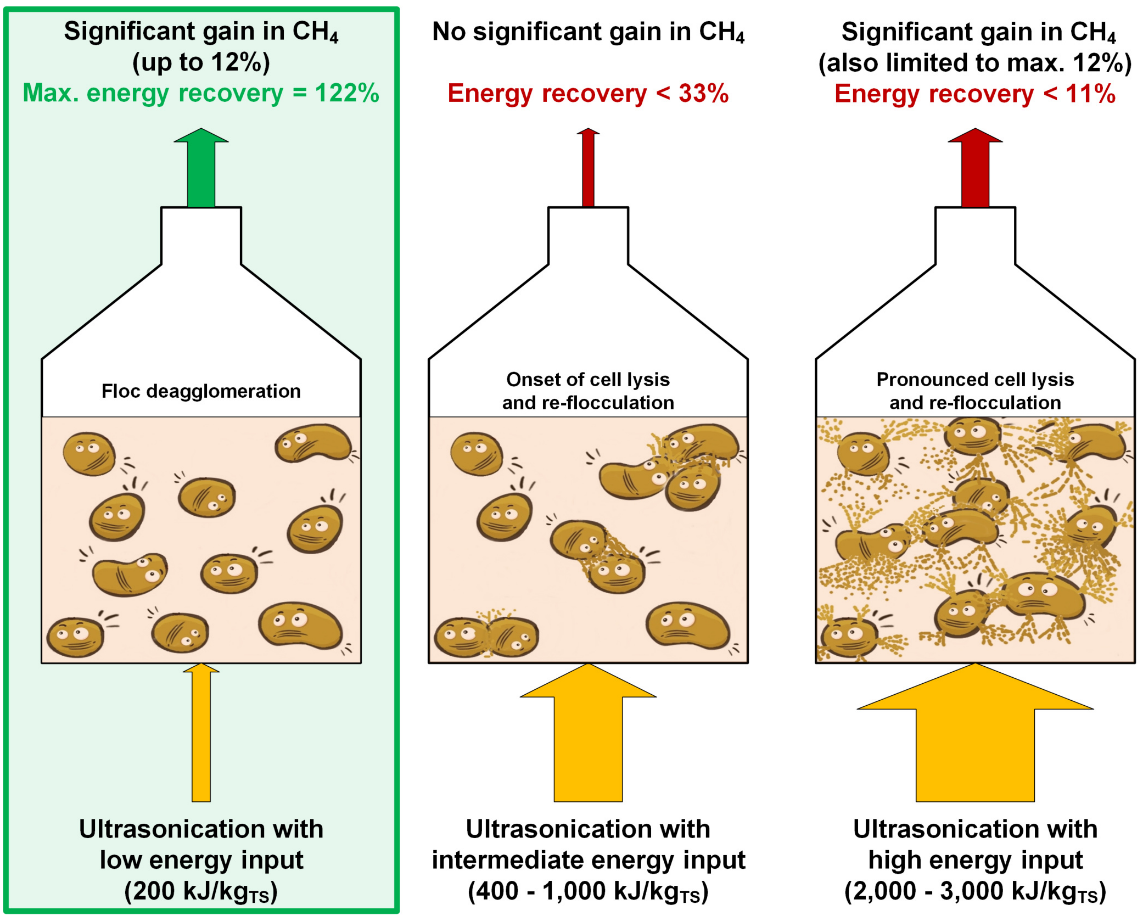

Figure B.1: Graphical abstract Paper I (*Energy-positive sewage sludge pre-treatment with a novel ultrasonic flatbed reactor at low energy input*).

APPENDIX C

Supplementary material Paper II

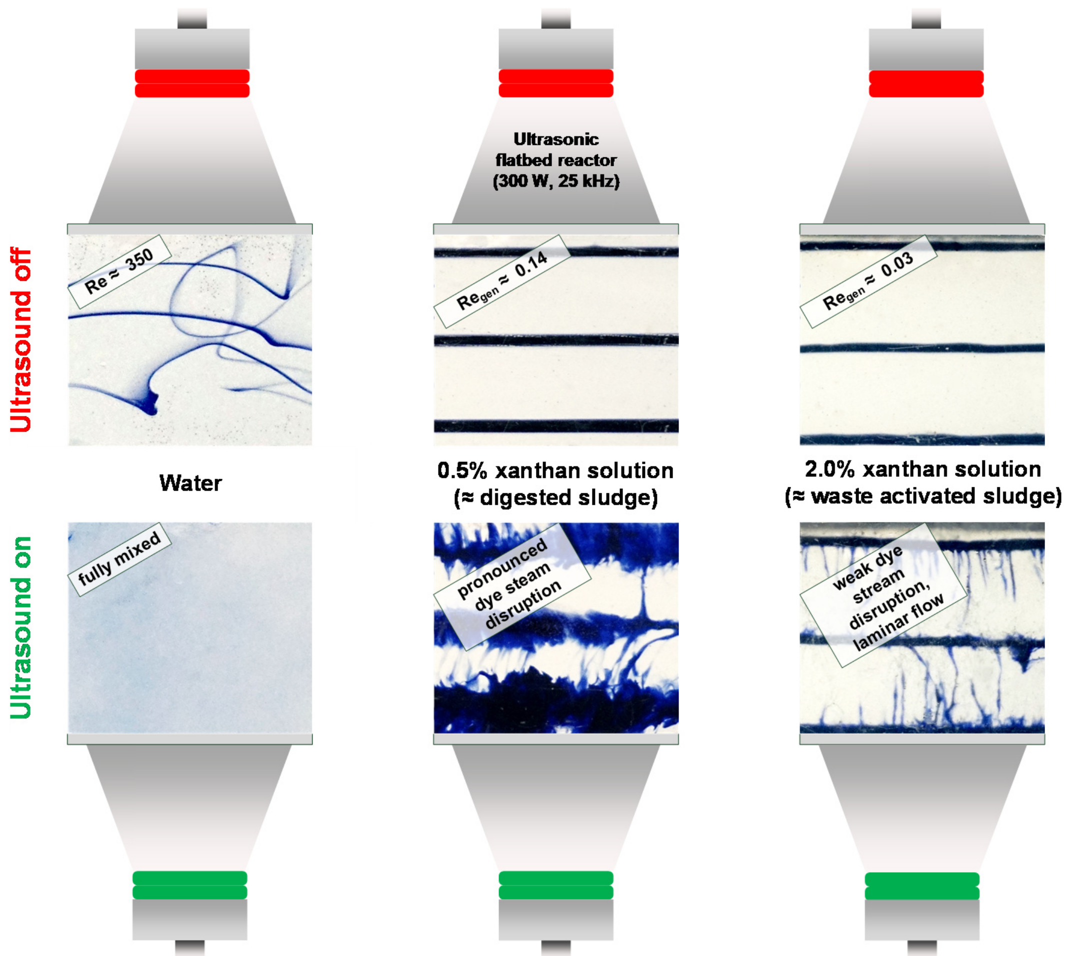

Figure C.1: Graphical abstract Paper II (*Impact of ultrasound-induced cavitation on the fluid dynamics of water and sewage sludge in ultrasonic flatbed reactors*).

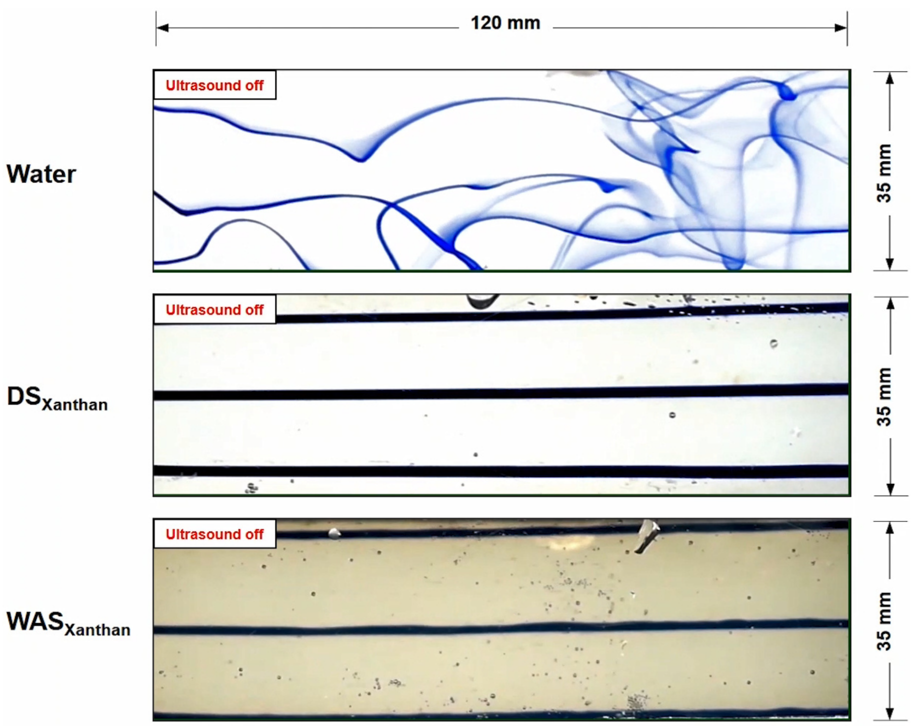

Figure C.2: Visualization of the flow behavior of water, $DS_{Xanthan}$ and $WAS_{Xanthan}$ in an ultrasound flatbed reactor before and during treatment by the use of dye streams. Video is available at https://doi.org/10.1016/j.ultsonch.2019.01.024.

APPENDIX D

Supplementary material Paper III

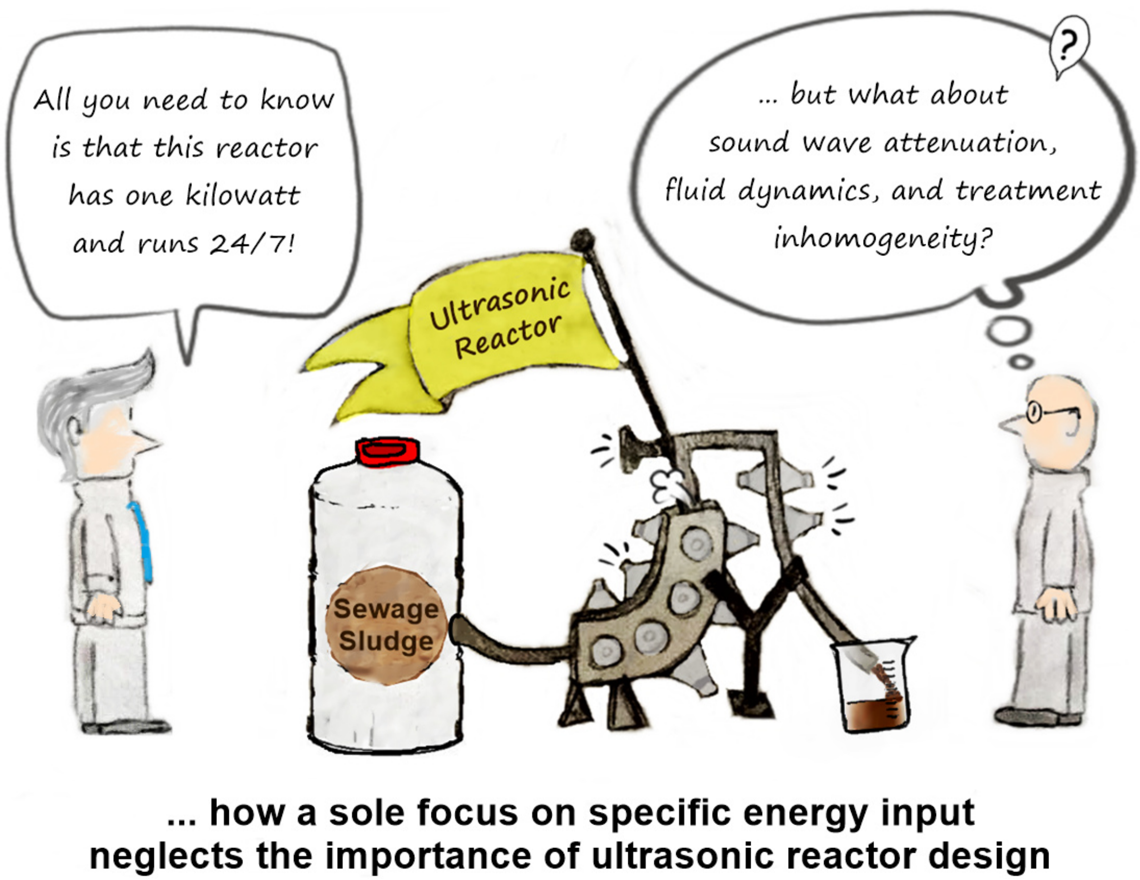

Figure D.1: Graphical abstract Paper III (*Effects of ultrasonic reactor design on sewage sludge disintegration)*.

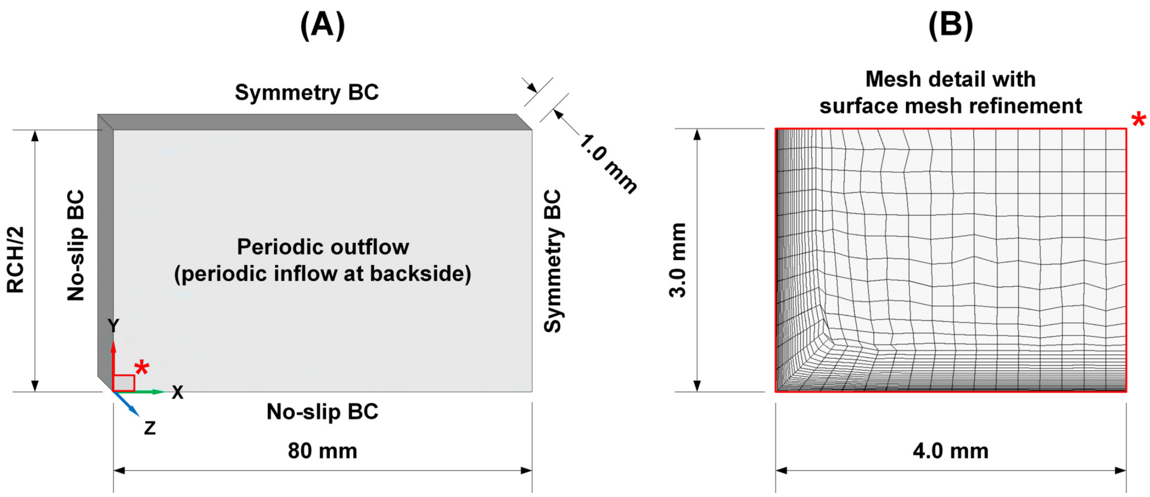

Figure D.2: 3-D periodic fluid domain and applied boundary conditions (BCs, subfigure A) and detail of the final mesh showing hexahedron elements and surface mesh refinements at the no-slip walls (subfigure B), for a reaction chamber height (RCH) of 100 mm. The location of the mesh cutout within the domain is marked by a red frame in subfigure (A).

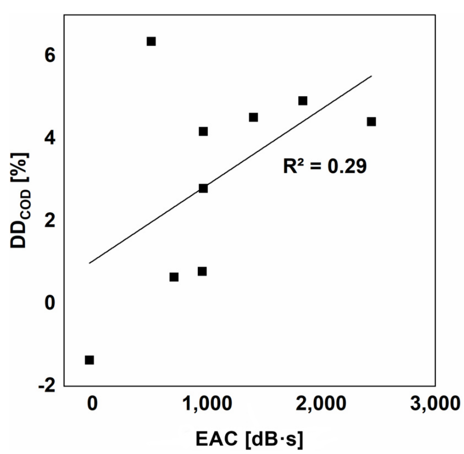

Figure D.3: Linear regression between exposure to acoustic cavitation (EAC, i.e., cavitation noise level multiplied by exposure time) and degree of disintegration (DD_{COD}).

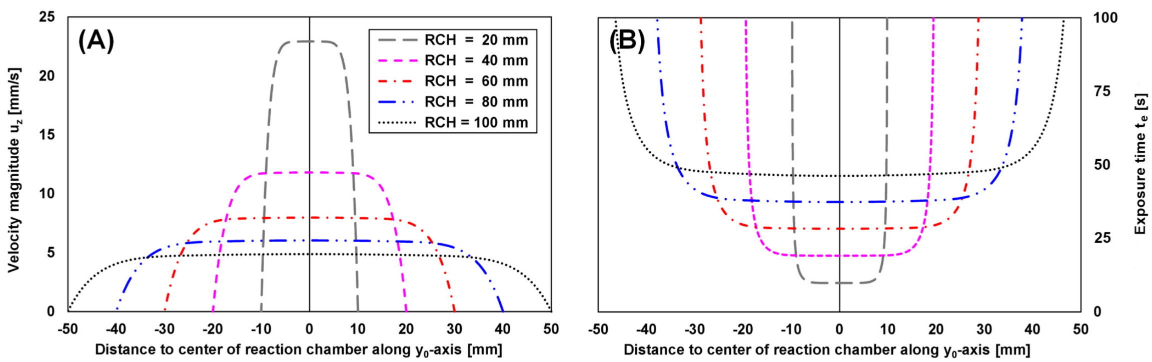

Figure D.4: Velocity profiles in u_z-direction (subfigure A) and affiliated exposure times t_e (subfigure B) along the y_0-axis for all tested reaction chamber heights (RCHs). For better visibility, the scale for exposure times was cut at 100 s as theoretical exposure times close to reactor walls were infinitely high due to the no-slip condition ($u_z = 0$). Exposure times for the slowest 2% of the flow are therefore not shown).

APPENDIX E

Supplementary material Paper IV

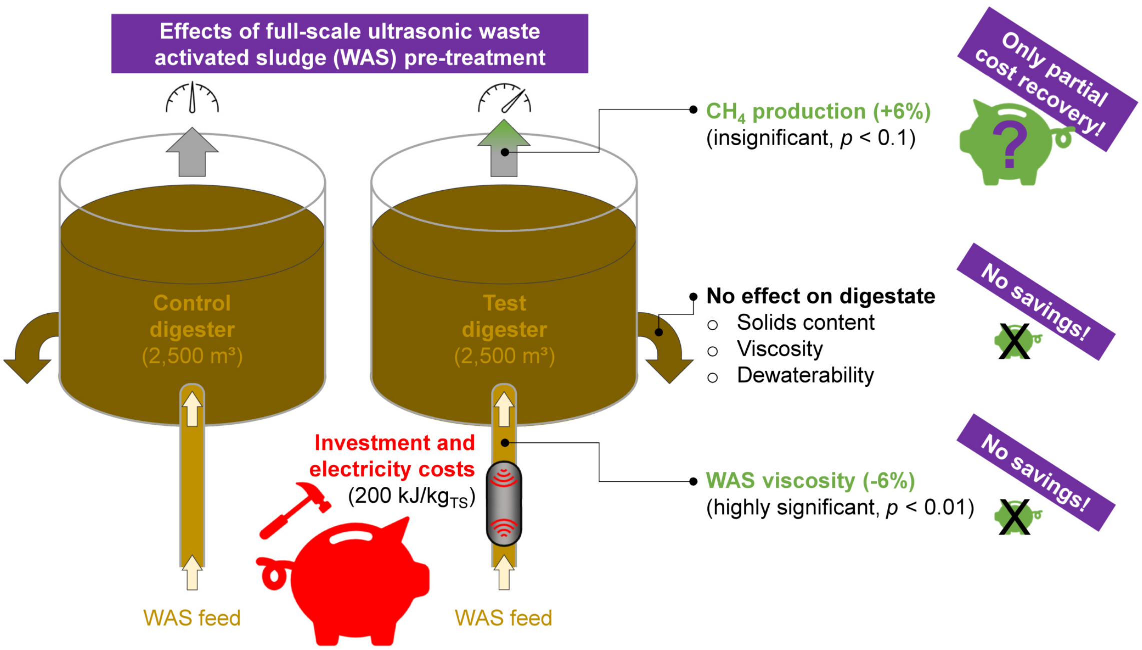

Figure E.1: Graphical abstract Paper IV (*Full-scale assessment of ultrasonic sewage sludge pre-treatment using a novel double-tube reactor*).

Table E.1: 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]	Share of WAS to RS [% of VS_{fed}]
Average	**154,800**	**164**	**91**	**4.1**	**81**	**6.4**	**73**	**8,100**	**38**	**33**	**43**
Median	**100,000**	**108**	**59**	**3.9**	**80**	**6.7**	**73**	**5,000**	**38**	**32**	**43**
WWTP #1 (Starnberg)	**100,000**	**89**	**31**	**3.8**	**86**	**6.3**	**74**	**5,000 (2 · 2,500)**	**38**	**42**	**33**
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 equivalent, PS = Primary sludge, WAS = Waste activated sludge, TS = Total solids, FM = Fresh matter, VS = Volatile solids, HRT = Hydraulic retention time, RS = Raw sludge.

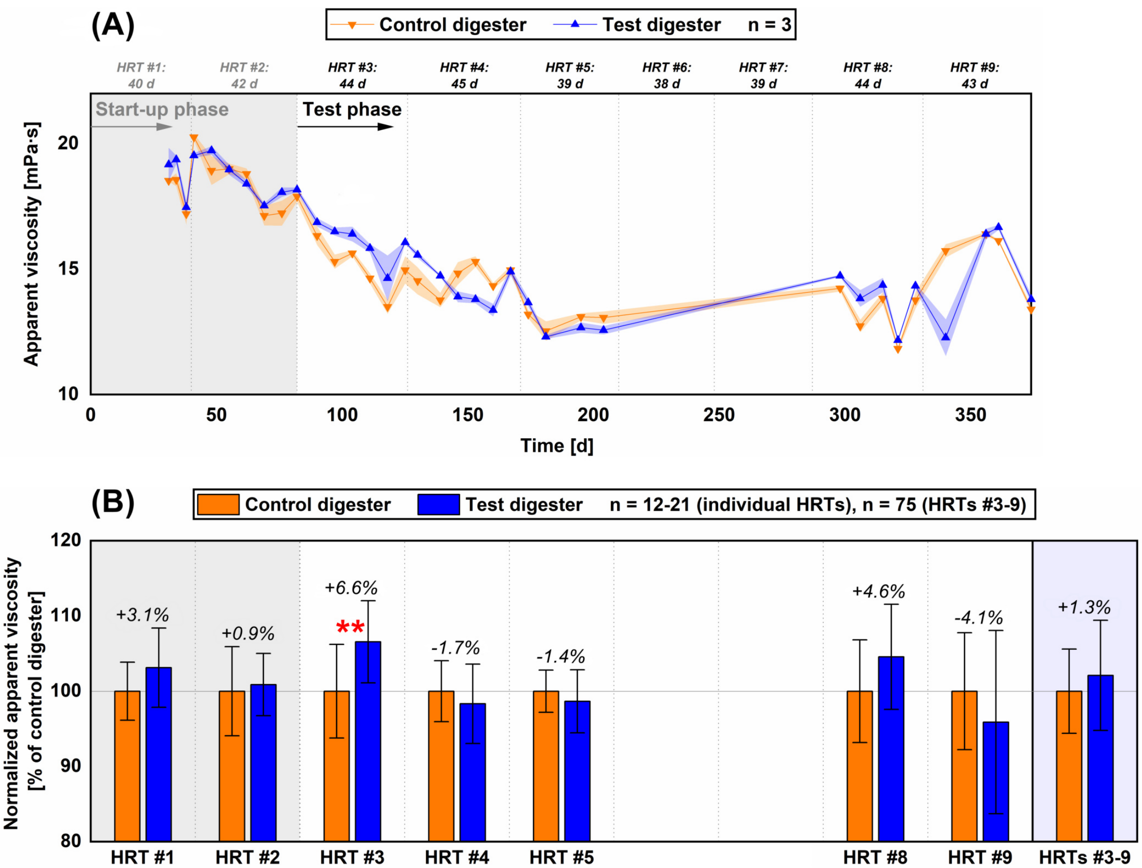

Figure E.2: Development of apparent viscosity of digested sludge from the control and the test digester at a shear rate of 644 1/s (A) and normalized apparent viscosity relative to the control digester (B). Highly significant differences between two data sets are denoted with two asterisks. Error bands and error bars denote the standard deviation of the mean.

APPENDIX F

Supplementary material Paper V

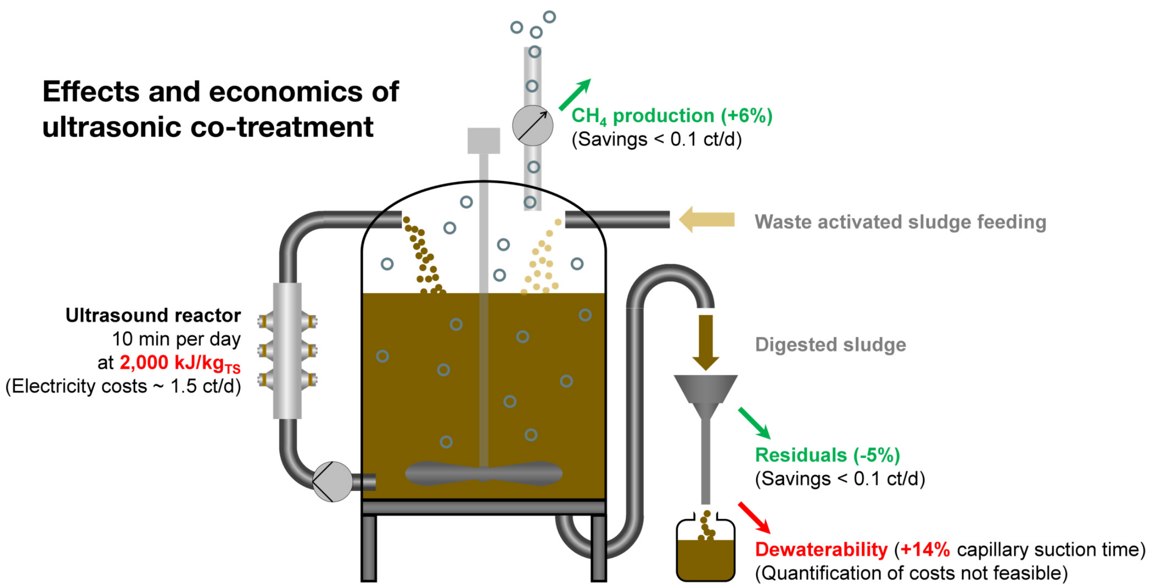

Figure F.1: Graphical abstract Paper V (*From pre-treatment to co-treatment - How successful is ultrasonication of digested sewage sludge in continuously operated anaerobic digesters?*).

Table F.1: Estimation of cost recovery.

	Unit	Test phase #1	Test phase #2
Electricity consumption of US reactor			
Power of US reactor	[kW]	0.3	
Operating time	[h/d]	0.17	
Energy used	[kWh/d]	0.05	
Electricity price[a]	[Cent/kWh]	30.43	
Electricity costs	[Cent/d]	1.52	
Methane production			
Energy content CH_4	[kWh/L]	0.01	
Conversion efficiency	[%]	40	
Avg. CH_4 production (control digester)	[L_{CH_4}/d]	6.14	8.48
Avg. CH_4 production (test digester)	[L_{CH_4}/d]	6.41	9.05
US-induced additional CH_4 production	[L_{CH_4}/d]	0.27	0.57
Savings due to additional CH_4	[Cent/d]	0.033	0.068
Residual sludge			
Assumed disposal costs[b]	[Cent/kg_{TS}]	0.30	
Avg. dry residuals (control digester)	[kg_{TS}/d]	0.029	0.050
Avg. dry residuals (test digester)	[kg_{TS}/d]	0.028	0.048
US-induced reduction in dry residuals	[kg_{TS}/d]	0.001	0.002
Savings due to reduction in dry residuals	[Cent/d]	0.043	0.063
Total costs	[Cent/d]	1.52	
Total savings	[Cent/d]	0.075	0.132
Net loss	[Cent/d]	1.45	1.39
Savings/costs	[%]	4.94	8.65

US = Ultrasound, [a] electricity price for 2019 according to the German Federal Ministry for Economic Affairs and Energy (www.bmwi.de/en), [b] assumed disposal costs for dry residue including transport and dewatering (to a total solids concentration of 25%), according to [23].

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