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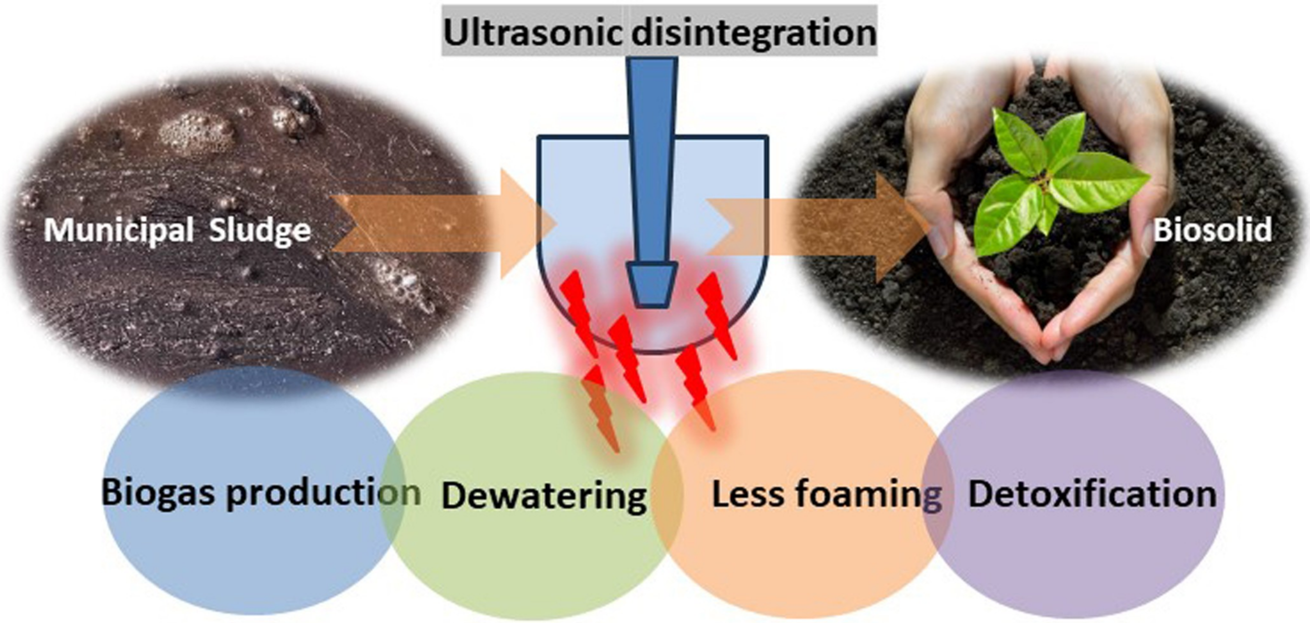
Ultrasonic Disintegration of Municipal Sludge: Fundamental Mechanisms, Process Intensification and Industrial Sono-Reactors

Ridha Djellabi,[a] Peidong Su,[b] Teklit Gebregiorgis Ambaye,[c] Giuseppina Cerrato,[d] and Claudia L. Bianchi*[e]



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Sludge disintegration is an environmental and industrial challenge that requires intensive research and technological development. Sludge has a complex structure with a high yield of various chemical and biological compounds. Anaerobic digestion is the most commonly used process for sludge disintegration to produce biogas, detoxify sludge, and generate biosolids that can be used in agriculture . Biological cell lysis is the rate-limiting cell lysis. This review discusses the application of sonolysis as a sludge pretreatment for enhanced anaerobic digestion via three combined processes: thermal destruction, hydrochemical shear forces, and radical oxidation. The mechanistic pathways of sono-pretreatment to enhance biogas, sludge-enhanced dewatering, activation of filamentous bacteria, oxidation of organic pollutants, release of heavy metals, reduction of bulking and foaming sludge, and boosting ammonia-oxidizing bacterial activity are discussed in this review. This article also discusses the use of ultrasound in sludge disintegration, highlighting its potential in conjunction with Fenton and cation-binding agents, and reviews common large-scale sonoreactors available on the market..

1. Introduction

Due to the huge increase in worldwide pollution and human activities, many environmental issues have been raised. Water, air, and soil pollution are major problems in the world. Large amounts of toxic municipal sludge are produced in wastewater treatment plants, which requirecareful treatment. Accumulationof toxic sludge in the environment is associated with many environmental and health risks. Dried sludge can be evaporate, leading to air pollution with harmful particulate matter as well as contamination of nearby soils. Unlike water treatment, purification of toxic sludge is more complicated.[1] The treatment of produced sludge at wastewater treatment stations could reach up to 60% of the total cost.[2] It is important to mention that the type, composition, and amount of produced sludge depend on the type of wastewater and treatment techniques.[1,3] Therefore, many suggestions have been proposed for the use of free or less sludge production wastewater treatment technologies.[4–6] In general, the production of sludge is still inevitable on a large scale. The sludge is formed typically of 5 to 20% solid, and the rest is water. Sludge contains a huge amount of organic matter. In general, four classes of sludge disintegration are available such as mechanical disintegration,[7] thermal disintegration,[8] chemical and thermo-chemical disintegration[9] and biological disintegration.[10] Sludge disintegration at large scale is passed through several steps by using successive mechanical, chemical, and biological techniques. Mechanical techniques are used for different reasons, such as dewatering sludge, thickening sludge, mixing sludge for enhanced biogas production, etc. The mechanical treatment could include high-pressure homogenization, high-pressure jet and collision, centrifugation, ball mill stirring, rotor-stator mixing, and emerging ultrasonic technology.[11]

Thermal treatment of sludge is based on the increase of temperature to disintegrate sludge cells. In general, thermal disintegration at temperatures ranging up to 100 °C occurs at normal pressure, while the thermal treatment at above 100 to higher values requires the application of pressure.[12] Thermal disintegration is applied mainly at a large scale using several systems such as Cambi® process (Norwegian company),[13] BioThelys® process, developed by Veolia Water.[14] Microwave disintegration is one of the thermal techniques that is used as an alternative to classical conventional thermal techniques because of its fast treatment, low cost, and efficiency. It is based on the application of frequencies of 300 MHz to 300 GHz to initiate thermal and athermal effects.[15,16] The microwave thermal effect results from heating generation, while athermal is the effect of an electromagnetic field to break bonds among macromolecules.[17,18] Thermo treatment is often combined with chemical techniques for enhanced sludge disintegration, achieved at lower temperature values. Of these combinations, the thermal treatment is associated with the addition of acids,[19] alkaline[20] and so on. Chemical techniques can be used to disintegrate sludge by the addition of oxidative agents such as O_3,[21] Potassium ferrate,[22] or systems that are able to generate reactive oxidative agents such as Fenton agents,[23] photo-Fenton.[24] Biological disintegration involving aerobic and anaerobic processing are commonly at large scale to reduce the volume of sludge and biogas generation.[25,26] Several biological mechanisms are used for sludge treatment, including enzymatic hydrolysis,[27] Protozoa and metazoa predation,[28] autothermal thermophilic aerobic digestion,[29] etc.

Cavitation is a highly promising physicochemical phenomenon characterized by the formation, expansion, and subsequent collapse of vapor-filled cavities caused by a sudden and significant decrease in pressure. Cavitation can be classified into hydrodynamic and acoustic cavitation, which can be used as pre-treatment techniques. Hydrodynamic cavitation (HC) is a sustainable and cost-effective technology that initiates by pressure differences in the medium, resulting in violent bubble collapse characterized by high pressure up to 5000 bar and

[a] R. Djellabi
Department of Chemistry, Alfaisal University, Al Zahrawi Street, Al Maather, Al Takhassusi Road, 11533, Riyadh, Saudi Arabia
[b] P. Su
School of Chemical and Environmental Engineering, China University of Mining and Technology (Beijing), Beijing 100083, China
[c] T. G. Ambaye
Department of Environment and Resource Engineering, Technical University of Denmark, 2800 Lyngby, Denmark
[d] G. Cerrato
Department of Chemistry, University of Turin, Via P. Giuria 7, 10125 Torino, Italy
[e] C. L. Bianchi
Department of Chemistry, University of Milan, Via Golgi 19, 20133 Milano, Italy
E-mail: claudia.bianchi@unimi.it

extreme temperature values ranging from 727 to 9727 °C within a fraction of seconds.[30,31] Hydrodynamic cavitation demonstrated high efficiency in many bioprocesses, such as anaerobic digestion.[32] Hydrodynamic cavitation is an increasingly popular technique for treating solids using waste-activated sludge (WAS).[33] This method uses different designs that can be categorized into static and dynamic devices. Static devices encompass equipment such as orifice plates, venturi tubes, whirling jet-induced hydrocarbons, and high-pressure homogenizers.[34] Rotating generators of hydrodynamic cavitation (RGHC), which are dynamic devices, exert significant shear pressures that disrupt the flocs of waste-activated sludge (WAS).

Ridha Djellabi is chemist with interests and expertises in processes and systems for sustainable environmental remediation and energy generation. He is one of the World's Top 2% Scientists issued by Stanford University & Elsevier. Ridha Djellabi received his PhD from Badji-Mokhtar University (Annaba, Algeria) in Analytical Chemistry and Environment. A Part of his thesis was conducted at the University of Milan, Italy. He carried out a postdoc at LSRE- University of Porto, Portugal. After that, Ridha worked at RCEES, the Chinese Academy of Sciences (Beijing, China) and Shenzhen University (China). Then, he worked as researcher at the University of Milan, Italy. Ridha worked as Personal Docent i Investigador at Universitat Rovira I Virgili, Tarragona in Spain for 2 years. Currently, Ridha is assistant professor of chemistry at Alfaisal University, Riyadh in Saudi Arabia. He co-authored more than 110 scientific publications and several book chapters.

Peidong Su is an assistant professor at the China University of Mining and Technology (Beijing). He obtained his Ph.D degree in Jackson State University in the United States in 2019 and has published more than 60 SCI papers. His research focused on the treatment of wastewaters, and he served as the Early Career Board Member of Reviews of Environmental Contamination and Toxicology (IF = 7.9) and Eco-Environment & Health, and he also served as Guest Editor of Toxics (IF = 4.6).

Teklit Gebregiorgis Ambaye is a post doctorial researcher in the field of environmental remediation, life cycle assessment, circular economy, and energy production with international experience in different countries, including Ethiopia, Belgium, Netherlands, Italy, and Denmark. Teklit received his PhD from the University of Brescia, Italy in Environmental Engineering. Teklit researched soil (bio) remediation, solid waste management, life cycle assessment, nanotechnology, wastewater, circular economy, water Quality, and Resource recovery. Teklit has published more than 60 peer-reviewed scientific papers in its relevant journals. He has been a reviewer for many journals. He taught several subjects, including solid waste management, environmental engineering, data analysis, and water treatment processes. He is currently a postdoctoral researcher in the field of integration of circular economy practice with LCA to quantify the environmental impacts and climate mitigation modeling of the most carbon-intensive construction material at the Technical University of Denmark.

Giuseppina Cerrato is an Associate Professor in the Department of Chemistry at the University of Turin (UNITO). She specializes in general and inorganic chemistry (SSD: CHIM/03). Her educational background includes a PhD in Chemical and Materials Sciences, and she is involved in both teaching and research activities at the university. Prof. Cerrato's research interests encompass a variety of topics, including the development of "smart" biomaterials for medical applications, surface characterization of various materials, and the use of photocatalysis for pollution abatement. Some of her notable research work includes studies on the structural and surface properties of yttria-stabilized tetragonal zirconia and the adsorption characteristics of different chemical species on modified zirconias. In addition to her research, Prof. Cerrato is actively involved in several teaching programs. She teaches General and Inorganic Chemistry courses with Laboratory, Didactics of Chemistry, and Solid State Chemistry: From the Macro to the Nano.

Claudia L. Bianchi is full professor of Industrial Chemistry at the University of Milan, where she coordinates the Master Degree in Industrial Chemistry and leads the Innovative Sustainable Materials for Environmental Remediation (ISMER) Laboratory. She is also vice-President of the European Society of Sonochemistry. With a deep expertise in Life Cycle Assessment (LCA), her research prominently features the development of photocatalytic materials, leveraging her knowledge of surface modification techniques to enhance photocatalytic efficiency. She has significantly contributed to designing, synthesizing, and characterizing micro- and nanostructured materials for pollutant degradation and sustainable energy production. Bianchi's expertise extends beyond academia, providing consultancy services to the industrial sector and advising on chemical processes, environmental compliance, and sustainable development. Her collaboration with academic and industrial partners ensures the practical application of her research, translating into scalable and economically viable solutions. Her substantial publication record includes over 300 papers in international journals, three patents, and an impressive H-index of 56 on Scopus, underscoring her authority in the field.

Rotary gas heat exchangers (RGHCs) offer distinct benefits compared to traditional heat exchanger devices, which has prompted the creation of innovative designs such as serrated discs, dimpled rotors, and pinned discs.[35]

Nevertheless, it is crucial to consider the phenomenon of cavitation erosion and the wear of parts subjected to cavitation. Design and development can be accomplished using either experimental or numerical modeling methods. Although these methods are faster and more cost-effective, they require validation. Despite significant breakthroughs, the issues of cavitation erosion and wear of cavitating elements continue to be a concern in the design of HC devices.[36]

The ultrasound process has been used over decades in labs and on large scales to disintegrate sludge.[37] It focuses on the generation of ultrasonic cavitation because of the growth of water bubbles up to a certain size, which causes them to collapse, resulting in physical and/or chemical effects based on the applied frequencies (Figure 1). In general, low frequencies produce a physical effect, while high frequencies can generate reactive oxygen species, i.e., •OH radicals. Ultrasonic cavitation can enhance biogas generation, reduction of foaming, digestion, dewatering, sludge detoxification, and enhancement of ammonia-oxidizing bacteria. In addition, the ultrasonic process has been combined successfully with many technologies, such as alkalization,[38] ozonation,[39] Fenton[40] and so on. This review discusses the application of the ultrasonic process to sludge disintegration and the role of ultrasonic cavitation in enhancing different mechanistic pathways during sludge disintegration using other technologies. The application of the ultrasonic process on a large scale by emphasizing some ultrasonic reactors available in the market will be discussed. Recommendations and suggestions are provided to further enhance research to meet industrial requirements.

2. Ultrasonic Disintegration of WWTP Sludge

Municipal sludge is semi-solid waste that results from the purification of wastewater. So far, three types of sludge can be produced in wastewater purification plants, including the primary sludge (PS), waste-activated sludge (WAS), and digested sludge (DS) (Figure 2a). PS mainly consists of suspended particles and organics which are produced from the mechanical (primary) clarification of wastewater, such as gravitational sedimentation and filtration. PS is fast-biodegradable because it contains easily digestible compounds, including carbohydrates and fats. Thus, PS has a high production of biogas. Waste-activated sludge (WAS), known as secondary sludge, is more complicated because secondary wastewater purification consists of large amounts of biological species, pathogens, and chemical pollutants. Therefore, WAS digestion is more complicated than PS and usually produces lower biogas since most of its energy is consumed in the secondary treatment stage.

Digested sludge (DS) is the obtained solid though the digestion of PS and WAS into reduced and purified non-odorous mass and less pathogens content. Anaerobic digestion is a commonly used technology to digest and stabilize sludges, which can reduce the mass of the sludge, removal the odor, decrease pathogens, and, more importantly, recover the biogas energy. However, the biological cell lysis is the rate-limiting step of the anaerobic digestion process. In terms of WAS, operators are also facing the hard treatment of combating bulking and foaming, which pose issues for the growth of plants in nearby zones. Therefore, a pretreatment of sludge to rupture the cell wall, to release the intracellular material, and facilitate the biodegradable of sludge is required before the anaerobic digestion. On the market, many methods are available for the pretreatment of municipal sludge, such as chemical, biological, thermal, and mechanical, for enhanced anaerobic digestion.[41] Among them, the ultrasonic process, as an emerging technology, has demonstrated efficient and fast sludge pretreatment processing.[42,43] Ultrasonic disintegration of

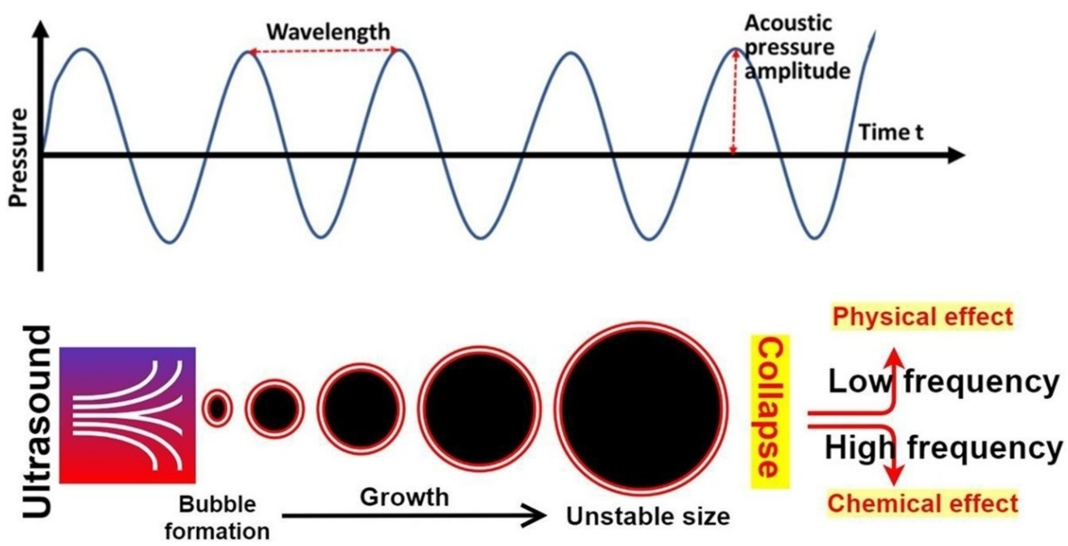

Figure 1. Acoustic cavitation process for the generation of physical and chemical effects, reproduced with permission from ACS@2022.[37]

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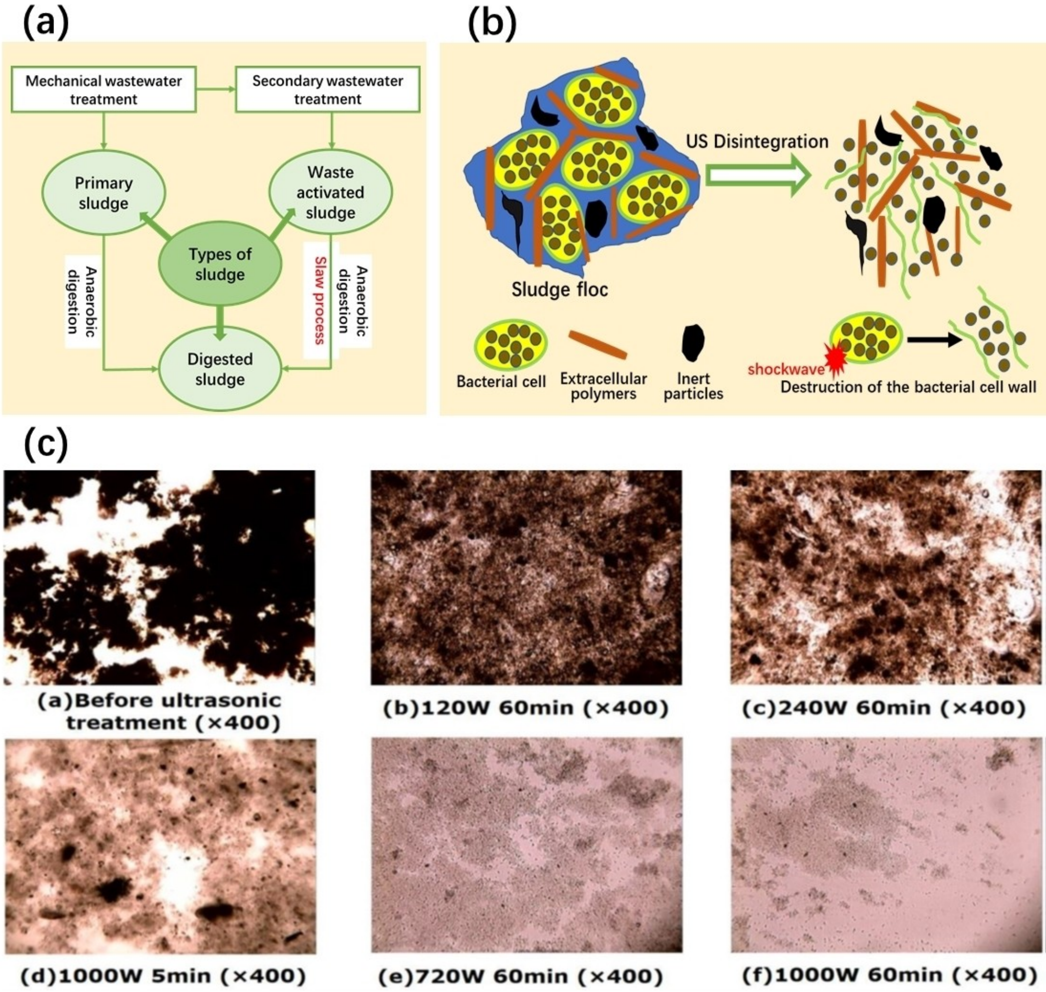

Figure 2. (a): Types of sludge generated in municipal wastewater treatment plants (WWTPs); (b): mechanism of ultrasonic disintegration of activated sludge; (c): Micromirror photographs of sludge floc after ultrasonic treatment at different powers, Reproduced with permission from Elsevier (n. 5830801401457. July16, 2024) Ref.[55]

sludge involves three pathways: free radical oxidation, hadrochemical shear force, and thermal degradation.[44] The mechanical pathway is predominant at low frequencies, while the chemical pathway via the generation of reactive radical species (ROSs) is more pronounced at high frequencies. Free radical oxidation is one of the mechanisms used to disintegrate sludge using sonication. In this method, bubble implosions can cause chemical reactions because they create highly reactive radicals (•H, •OH) and break down substances at high temperatures (pyrolysis), primarily via sonochemical reactions. The intense temperatures and pressures during the collapse of cavitation bubbles lead to the creation of highly reactive molecules, which then combine to produce hydrogen peroxide in a water-based environment.[45] Aqueous sonochemistry includes a variety of oxidation and reduction processes that result from secondary interactions with highly energetic intermediates. These radicals undergo reactions with biomolecular entities analogous to those generated by ionizing radiation. Pyrolytic processes within cavitation bubbles primarily degrade volatile pollutants, whereas interactions with hydroxyl radicals in the bulk liquid destroy nonvolatile pollutants. Sonochemical degradation processes occur across a broad range of ultrasonic frequencies, with the most effective results found at frequencies ranging from 200 to 1000 kHz.[46] Thermal pretreatment is another mechanism used in wastewater treatment facilities (WWTPs) to break down sludge cells by subjecting them to elevated temperatures and pressures.[47] Implementing this technology increases biogas generation and less volatile solids in the digested sludge, leading to smaller outputs with improved dewatering qualities. Thermal pretreatment also decreases the presence of components that are difficult to break down, thus enhancing the overall effectiveness of organic substance removal during digestion.[47] Moreover, high temperatures can result in the breakdown of sludge owing to the breakdown of lipids in the cytoplasmic membrane. This breakdown can lead to puncturing of the membrane and release of intracellular substances into the surrounding liquid. The thermal impact on sludge disintegration is negligible because of the low level of solubilization and extended duration of the reaction.[48] In general, the degree of sludge disintegration is influenced by the amount of energy absorbed, highest temperature reached, and duration of exposure.

Hydromechanical shear forces, which create intense localized pressures and shockwaves at the bubble's position, affect the breakdown of sewage sludge and cause mechanical harm to surrounding substances.[49] When the bubble is close to a solid surface, a liquid jet with a high velocity can collide with and harm adjacent cells.[50] Initially, the collision causes disintegration of clusters of sediment particles, liberating individual bacterial cells that have the potential to generate cavitation bubbles, which can expand to a size of up to 175 μm.[51] Forceful collapse generates intense hydromechanical shear pressures inside the liquid, rupturing nearby microbial cells and their cell membranes. Ultrasonic cavitation generates hydromechanical shear forces that break apart macromolecules weighing more than 40,000 Da.[52] The frequency range of 20–40 kHz, in which mechanical forces are the most efficient, achieves optimal disintegration. Hydromechanical shear forces were the main cause of sludge disintegration when the ultrasonic density was 0.384 W/mL or lower.[42] The impact of sonochemical phenomena, specifically oxidizing radicals, became more prominent as the ultrasonic density increased. In general, the mechanical forces generated by sonication have a more significant impact on sludge breakdown than the chemical processes caused by sonication.[53]

Compressions and rarefactions will be generated in situ when the ultrasound is applied in a sludge medium at a low frequency ranging from 20 to 40 kHz. The compression cycles push the liquid molecules together via a positive pressure. In contrast, the rarefaction cycles pull molecules from each other via a negative pressure, resulting in cavitation microbubbles with successive growth till an unstable diameter, wherein they violently collapse, giving a shockwave energy with a distractive potential.[54] As represented in Figure 2b, this mechanical energy is able to effectively destroy the strong bacterial wall of the sludge and disintegrate the sludge solid into a smaller mass. Figure 2c shows the micromirror photographs (magnification of 400 times) of municipal sludge before and after ultrasonic at different ultrasound powers ranging from 120 to 1000 W.[55] It is obvious that the sludge flocs were destroyed by very small particles, where the degree of damage to the flocs structure increases gradually with the increase of power.

Applying ultrasonic vibration on sludge can significantly change its physicochemical characteristics such as the pH, temperature, particle size of sludge, conductivity, chemical and biological composition, etc.[56–58] Three mechanistic pathways can take place during the ultrasonic disintegration of activated sludge: thermal destruction, hydro-chemical shear forces (predominant) and (iii): radical oxidation.[59] Lower frequencies (20 to 40 kHz) produce a high amount of cavitation bubbles, having a strong shearing force that can destroy the sludge flocs mechanically. There is an optimum between the yield of bubbles and the shear degree produced via such bubbles.[60] At the same time, the change in the frequency results in different mechanisms, including nucleation, micro-mixing, degradation, and fragmentation.[61] The efficiency of sludge destruction also depends on the energy input, nature and composition of sludge, and the reactor design. Therefore, the control and the optimization of operating factors and designing are a must for enhanced ultrasonic treatment of a specific sludge. In a sludge digestion station, the ultrasonic process can achieve many advantages (Figure 3), such as enhanced degree of digestion, enhanced volatile solids reduction, enhanced biogas production, reduced sludge mass, enhanced dewatering, activation of filamentous bacteria, degradation of organic pollutants and recovery of heavy metals, enhanced of ammonia-oxidizing bacteria activity, reduction of bulking and foaming sludge. In the following sections, the role of ultrasonic cavitation on sludge will be discussed in depth.

2.1. Intensification of Biogas Production

Biogas, which consists of methane, carbon dioxide, and hydrogen sulfide, is a renewable alternative energy obtained from the degradation of organic load.[62] Unlike other techniques, anaerobic digestion is very efficient for biogas generation because all organic matter, including carbohydrates, lipids, and proteins, can be converted to methane.[63–66] The sludge is very rich with organic matter. Consequently, the use of an ultrasonic process as a pre-treatment before anaerobic digestion is very efficient in breaking down the sludge flocs, releasing the

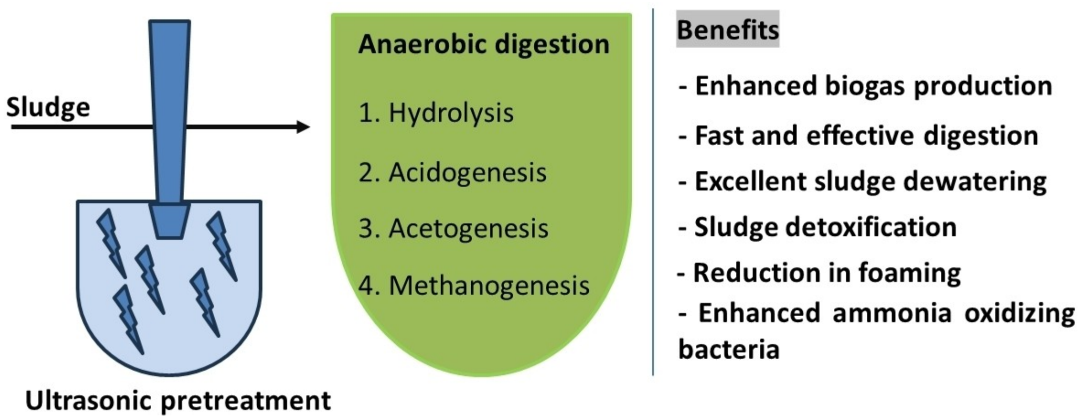

Figure 3. The advantages of the application of the use ultrasound process on the sludge pre-treatment.

digestible organic matter.[59,66–69] Matthijs et al.[70] found that the ultrasonic process was the most efficient technique compared to ozone and hydrogen peroxidation processes in biogas production. Alagöz et al.[71] studied the comparative efficiency of ultrasound and microwave irradiation as a sludge pretreatment to spend up the hydrolysis stage and enhance the anaerobic digestion for improved biogas production. Their results demonstrated that the ultrasonic treatment was much more efficient and low-cost than microwave irradiation for methane production, wherein the applied energy in microwave was almost 9 times superior to that used in the ultrasonic process. G. Erden et al.[72] have studied the effect of ultrasonic treatment at low ultrasound densities on the anaerobic biodegradability of biological sludge. The methane production was enhanced by 44% for the sample treated with a power density of 0.09 WmL^{-1} and energy input of 9690 $kJ\,kg^{-1}$ TS, compared with the untreated sample. The enhanced degree of disintegration of sludge and methane production was due to efficient cell lysis by ultrasonic treatment. However, at higher energy more than 9690 $kJ\,kg^{-1}$ TS, the sludge was mineralized preceding a solubilization which decreased the sludge disintegration. Ultrasonic field can increase the temperature which helps to destroy the sludge flocs, while a higher increase in temperature limits the cavitation intensity and the sludge disintegration. Additionally, authors reported that chemical oxygen demand (COD), dissolved organic carbon (DOC), total nitrogen (TN), and total phosphorus (TP) rates were significantly increased in the sludge as a result of ultrasound treatment. Paulista et al.[73] reported that the coupling US and anaerobic digestion enhances the methane production from a mixture of crude glycerol and biological sludge because US can promote the hydrolysis of long-chain fatty acids and deagglomerates the microorganisms in the sludge flocs. Wu et al.[74] reported that the increase in ultrasonic power from 30 to 300 W significantly improves the COD release in the supernatant. The results showed that there was a relationship between the released COD yield and heterotrophic bacterial activity, wherein, an increase in released COD results in lower heterotrophic bacterial activity, suggesting that the released organic matter is from the lysed cells of heterotrophic microorganisms. It is important to point out that the increase in biogas production via the ultrasonic pre-treatment process depends not only on the applied conditions, such as the energy input but also on the types and sludge characteristics, especially the load of organic matter that exist in the sludge.[64,67]

Utilizing low-intensity ultrasonics can increase the amount of product produced and decrease the time it takes for anaerobic digestion by boosting the activity of enzymes in microbes and improving the rate at which mass is transferred. Both laboratory- and pilot-scale studies have demonstrated the efficacy of ultrasonic-assisted anaerobic digestion treatment, with multiple investigations demonstrating improved biological activity.[75] For example, Zhang and Jin[76] used online ultrasonic equipment to break down sludge in an anaerobic membrane reactor. This improved the performance of the reactor and led to a 51.3% rise in the amount of volatile solids that were used. In addition, one study measured the effects of low-intensity ultrasonics on anaerobic digestion processes, specifically examining TTC-dehydrogenase activity and coenzyme F420 factor concentration. Exposure to ultrasonics with a low intensity (0.2 W/cm2) and a frequency of 35 kHz led to an increase in biological activity. However, when the intensity of the UV light exceeded 0.4 W/cm2, there was a significant drop in activity.[77] One study demonstrated that the application of low-intensity ultrasonic (US) can enhance methane output by 40% during biogas generation. This increase was observed in ambient and mesophilic settings as well as in up-flow anaerobic sludge blanket reactors, with improvements ranging from 38% to 43%. In addition, subjecting the USBr system to a continuous application of power at a density of 0.05 W/mL for 1 second every minute resulted in a 43% increase in methane output. A study of cell morphology has revealed a multitude of indentations and fractures in cells exposed to low-intensity ultrasound.[78,79] Furthermore, the physical effects of ultrasonication provide advantageous conditions for the delivery of nutrients and substrates to cellular structures, which in turn activate intracellular calcium channels and accelerate the log phase in microorganism growth. It is used in the pretreatment of feedstock and the generation of biogas, resulting in enhanced solubilization of organic matter and intensification of the biogas production phase. Ultrasonication (US) causes beneficial alterations in microorganisms and enhances mass transfer efficiency, leading to a significant increase in biogas production within a reduced timeframe.[80]

2.2. Ultrasonic Dewatering of Sludge

In WWTP, dewatering sludge flocs, which are repositories for water, is of paramount interest since it results in less and easier-handily amounts of sludge. Four categories of water can be found in sludge: free water, interstitial water, bound water, and intracellular water.[81] Sludge dewatering is very costly and a serious challenge because of the large quantities of residual sludges produced annually.[82–84] Several technologies are applied to dewater sludges at WWTPs, such as mechanical,[85] thermal,[85] solar drying,[86] and electro-dewatering.[87] Sludge dewatering by ultrasonic process is also an emerging economical and efficient technology. The shockwave produced by bubble implosion breaks down the extracellular polymeric substances (EPS) and bacterial cell walls in the sludge flocs. Such destruction results in lower sludge particle size and improves its dewaterability. However, very small particle sizes may lead to clogging of the cake and limit the dewatering of the sludge. Such counteractive measures must be investigated in full-scale applications to optimize the value of energy power for better sludge dewaterability.

Ultrasound enhances sedimentation and dewatering efficiency by inducing a sponge-like action on the sludge, which facilitates the aggregation of sludge particles and increases particle size. Additionally, it reduces the size of floc particles and disrupts the arrangement of bacterial micelles, thereby transforming bound water into unbound water.[55] Ultrasound effectively eradicates viruses, bacteria, and other detrimental

Review
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entities in sludge, thereby enhancing heavy metals' leaching and recovery rates. Various aspects, such as ultrasound frequency, duration, acoustic energy density, pH value, mode of action, and coupling mechanism, determine the effectiveness of ultrasound therapy in enhancing sludge dewatering performance. In addition to shorter treatment durations, medium- and low-frequency ultrasound is advantageous for sludge dewatering. Ultrasound treatment improves organic matter dissolution, reduces sludge thickness, and promotes sludge distribution.[88]

Na et al.[89] for example, reported that the ultrasonic treatment significantly improves the dewatering of waste-activated sludge by decreasing the capillary suction time from 53 s (untreated sludge) to less than 10s (treated sludge with $>$2000 kJ/l ultrasonic energy). The ultrasonic dewatering of sludge depends on the applied energy dosage, wherein, a low energy dosage is not enough to effectively dewater the sludge, while a higher energy dosage can deteriorate sludge dewatering. Feng et al.[90] studied the mechanistic pathways behind the ultrasonic dewatering of waste-activated sludge. The capillary suction time (CST) and specific resistance of filtration (SRF) decreased, respectively, from 94.2 s and 2.35×10^{10} m/kg (untreated sample) to 83.1 s and 1.30×10^{10} m/kg (US treated sample with 800 kJ/kg TS). However, a negative sludge dewatering was observed at a very high energy dosage (4400 kJ/kg TS). The ultrasonic energy dosage can negatively or positively influence sludge's dewatering via the changing of the extracellular polymeric substance (EPS) and particle size of sludge. Therefore, an optimum energy dosage, depending on the working conditions and the characteristics of the sludge, should be obtained for enhanced sludge dewatering. Besides the mechanical effect of US on the reduction of particle size in sludge, the ultrasonic process can decrease the particle size of sludge through the breakdown of cell membranes by US-produced radicals at a higher frequency.[53] The optimization of operating factors is the key to sludge dewatering via ultrasonic cavitation, as reported by several research groups. The ultrasonic power density should be optimized for optimum particle size and enhanced sludge dewatering. Sahinkaya et al.[91] reported that the most effective ultrasonic density is within the range of 0.75–1 W/mL. Deviating from this range by raising the density beyond 1.2 W/mL or decreasing it below 0.5 W/mL resulted in a decrease in the capacity to dewater sludge.

Mobaraki et al.[92] demonstrated that ultrasonic time and power impacted the protein and polysaccharide content in sludge dewatering. The sample underwent a 15-minute power set at a concentration of 5.0 W/mL, resulting in a protein release of 1.4 mg/L and a polysaccharide content of 21 mg/L. Generally, low-to medium-frequency and short conditioning periods have positive effects. However, increasing the ultrasonic frequency and prolonging the conditioning time can decrease efficiency. High-power ultrasonic treatment can reduce efficiency by increasing the surface area of the sludge that retains water. Low-energy and short-duration ultrasonic therapy can enhance efficiency; however, any deviation from the optimal settings can decrease efficiency.

The cost of activated sludge dewatering by ultrasonic process varies from case to case depending on the type of the activated sludge, the ultrasonic system itself, and the wanted goals. Some studies at the lab scale reported that US sludge treatment could be a cost-effective process due to the short treatment time and the co-generation of valuable biogas. The dewatering is a perfect option to reduce the sludge amounts. The sludge reduction time (SRT) characterizes the speed of the sludge quantity reduction over time. One of the industrial benefits of using the ultrasonic process is the short period, which is considered a huge industrial advantage. Nico et al.[43] reported the demand for electrical energy declined from 89.920 to 15.517 kWh per ton of dry sludge avoided, reflecting a significant energy win, researching 80%. Many worldwide companies have involved ultrasonic sludge treatment in their stations due to the cost-effectiveness, efficiency, short time, and enhanced biogas production. To boost further ultrasonic dewatering and reduce the treatment cost, the ultrasonic process was also combined with other methods for enhanced sludge dewaterability. Mei-Qiang et al.[93] have integrated the acidification and ultrasound processes to enhance the dewatering of sludge at lab and pilot scales, as shown in Figure 4. The authors found out that the US can improve the porosity and break down the extracellular polymeric substances (EPS) by shockwaves and oxidation via produced radicals, which improves the mass transfer and hydrolysis via acidification. Ultrasonic-based devices for smart-drying and dewatering materials have been developed as well. Zinovy Plavnik et al.[94] invented an apparatus by means of ultrasonic for effectively breaking down the boundary humidity-water layers and facilitating the transfer of heat. Devices of US transduces based on a pneumatic type or combined with a UV light system for further boundary layer disruption were developed.

2.3. Ultrasound-Assisted Sludge Detoxification and Disinfection

WWTP sludge contains a high content of various toxic pollutants, including heavy metals, organic contaminants, pathogens, and unpleasant odors. The valorization of sludge as fertilizer is one of the most famous approaches. For example, the European Commission reported in 2010 that 39% of sewage sludge generated in the European Union was recycled into agriculture.[95] Therefore, the detoxification of sludge is rate-limiting and an issue restricting its general application in agricultural fields. The ultrasonic process or, combined with chemical techniques, has been widely used for the removal of toxic elements from sludge due to the power of the cavitation effect, thermal effect, mechanical effect, and oxidation via the generated ROSs.[96–102] Detoxification and disinfection occur simultaneously during dewatering, and sludge blocks destruction. The release of organic, biologic, and metallic species due to the cavitation forces is a manner of detoxification. These species will be extracted and might be used for biogas production or concentration and separately treated. Guangming et al. reported energy input's effect on releasing heavy metals from sludge.[103] The applied energy for the release of each metal was quite different due to their different phys-

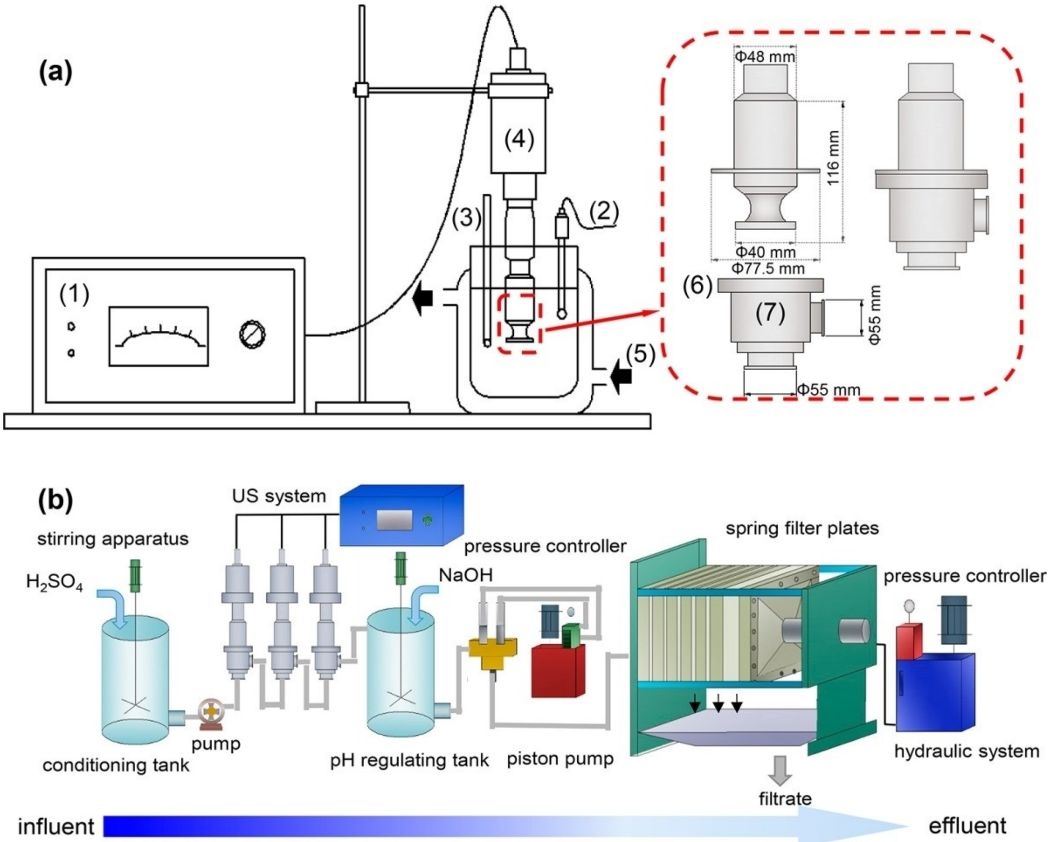

Figure 4. Scheme of ultrasonic system: ultrasonic generator, (2) pH-meter, (3) thermometer, (4) ultrasonic probe, (5) cooling system, (6) receiving ring, and (7) reactor. (b) Sludge conditioning and dewatering pilot employing acidification/ultrasonic combined system, reproduced with permission from ACS@2018.[93]

icochemical characteristics and the bonding of metals in sludge. A power density ranging from 0.8–1.6 $W\,ml^{-1}$ was effective for leaching the metals as mentioned above, while too high or too low power density negatively affected the release of metals. Xie et al.[104] reported the ultrasound-assisted acid leaching of Fe and Cu from printed circuit board waste sludge. It was found that this combined system is very efficient for removing and separating metallic cations compared to conventionally heated acid leaching without ultrasound. Ultrasonic cavitation bubbles can simultaneously improve the dissolution of copper hydroxides into liquid and precipitation of iron oxides into solid. This efficient technology has been successfully applied to the industrial scale for the recovery of heavy metals (Huizhou city, China). Samuel et al.[105] also studied the ultrasound-assisted leaching of metals such as Cu and Zn from sludge in combination with chemical leaching. Rumky et al.[106] integrated Fenton with ultrasound to remove Cd, Cr, Cu, Pb, and Zn from sludge. The authors stated that the co-production of radical species via the Fenton reaction, ultrasonication, and acoustic cavitation work synergistically to destroy the membrane cell and intracellular organic substances, resulting in higher metal leaching.

Cr(VI) removal from tannery sludge by ultrasonic cavitation was reported by Hongrui et al.[107] The authors stated that almost all Cr and COD in the sludge were extracted from the sludge at optimum conditions. Pham et al.[108] studied the comparative efficiency of the ultrasonic process and Fenton reaction as a pre-treatment to facilitate the biodegradation of bis(2-ethylhexyl) phthalate (DEHP) in WWTP sludge. Ultrasonic treatment was operated at 20 kHz, while the Fenton reaction was carried out after the acidification of the sludge (pH 3) and via the addition of H_2O_2 and Fe(II) Fenton reagents. Even though the Fenton is a chemical-consuming reaction, it was found that the ultrasonication gives a better DEHP oxidation than the Fenton.

Sono-advanced Fenton-like combined system has been widely used as synergistic processes for the release and degradation of many types of organics pollutants in sludge such as textile dyes,[109] aromatic amines,[110] PAHs[111] and polyaromatic hydrocarbon.[112] A combined ultrasound-Mn(VII) system was used for the oxidation of 3,3′-dimethoxybenzidine in sludge.[113] Ultrasound-assisted ozonation synergistic process was also used to release and remove organic pollutants such as volatile solids[97] and fluoroquinolone contaminants levofloxacin.[114] The enhanced nitrogen and phosphorus removal from activated sludge was performed by combining ozone and ultrasound processes.[115]

2.4. Reduction of Bulking and Foaming Sludge

In WWTPs, the primary and the activated produced sludge are usually associated with an excessive generation of bulking and foaming, which is mostly a result of many causes, such as the accumulation of acetic acid, hydrophobic substances, and the presence of a high amount of filamentous microorganisms *Microthrix parvicella*.[116] Excessive foams in the digestion station cause a significant inhibition in the stabilization degree and biogas formation. One reported advantage of sludge pre-treated with an ultrasound process is the significant reduction of foaming incidents. For example, Isik et al.[117] reported that the sonication process can control and optimize the foam during sludge treatment at low energy input, wherein, at higher energy input, a foaming generation issue was observed due to the increased solubilization and degradation rates. Wünsch et al.[116] studied the degradation of filamentous microorganisms by ultrasonication at a lab and full scales (Reinfeld sewage treatment plant, Germany). The authors reported that generating high local shear stresses by low-frequency is very efficient in destroying the filamentous structures of the sludge.

In general, the overgrowth of filamentous bacteria causes filamentous bulking and foaming in wastewater stations.[42,55] To handle the activated sludge, it is important to provide conditions that allow the formation of robust floc. In this manner, a moderate amount of filamentous bacteria is necessary.[118] Cavitation can limit the overgrowth of filamentous bacteria in the medium, but it does not disintegrate it totally. A small number of filaments, connect to each other, forms a mesh that provides a support structure for other bacteria to attach to as they form flocs.[119]

2.5. Activity Enhancement of Ammonia-oxidizing Bacteria

Ammonia-oxidizing bacteria (AOB) is a limiting factor in achieving the transformation of toxic ammonia to nitrite in biological WWTPs.[120] In contrast, nitrite-oxidizing bacteria (NOB) are used as a counter-reaction to inhibit the conversation of ammonia. Since it is difficult to control or limit NOB,[121] research studies currently focus on controlling and enhancing AOB/NOB abundance.[26,122–124] The use of the ultrasonication process to enhance AOB activity for ammonia nitrogen removal has drawn much attention. Huang et al.[125] stated that the use of low-intensity ultrasound = 0.25 W/mL shorted the start-up period partial nitrification and enhanced the nitrite accumulation ratio due to the enhancement of AOB activity. Many reports stated that the use of the ultrasound process for a longer time has a positive effect on the population of AOB of the genus *Nitrososphaera* and a reduction in the population of NOB (*Nitrospira*) was observed.[122,125,126]

3. Combination of Ultrasound with Chemical Processes for Sludge Disintegration

Chemical oxidation treatments, such as the Fenton process, ozone oxidizing method, and ultrasonic technology, can be used to achieve high sludge reduction efficiency. The ultrasonic application can enhance the digestibility and dewaterability of the sludge by transferring the organic matter of the sludge into the liquid phase. For instance, according to Golbabaei et al.,[127] in their research to improve dewatering using ultrasound, ultrasound can be used to break up microbial cell walls and sludge flocs, which could cause the sludge to disintegrate and release organic compounds into the liquid phase and enhance the biodegradability and dewaterability of the sludge. Hydro-mechanical shear forces mostly cause the breakdown of sludge. Bardi et al.[128] observed that long-term application of ultrasonic waves at higher power density or intensity while using a high specific energy input (> 5000 kJ kg TS1) leads to high sludge reduction and the formation of radicals ($H^\bullet$ and $^\bullet OH$), which contribute to sludge disintegration. According to Lambert et al.,[43] in an SBR system, ultrasonic waves with a specific energy of 6000 kJ/kg DS were used to reduce the yield of sludge by 15 % to 45 % by applying them to 15 % to 30 % of the total amount of sludge produced each day. Although pricey, using these waves at high energy density could negatively impact the sludge's characteristics. Therefore, ultrasonic could be combined with another treatment technique, such as the Fenton treatment, to reduce energy consumption while also increasing the degree of sludge disintegration.

The use of the Fenton reaction, one of the most efficient advanced oxidation processes, is another successful method for sludge reduction. A ferrous iron catalyst speeds up the breakdown of hydrogen peroxide during the Fenton reaction. Hydroxyl radicals are potent and non-selective oxidizing agents, and this can hasten their production. To release intracellular organic compounds into the liquid phase, hydroxyl radicals can oxidize the membrane of a microbial cell.[129] While this is happening, hydroxyl radicals can transform the resistant substances into biodegradable ones that can be used as nutrients in additional biological treatments.[130] Fenton oxidation is a highly effective for treating sludge in various industries, including petroleum, municipal, and aluminum wastewaters.[131] Its high oxidation potential, simplicity, and short reaction time make it suitable for mineralizing WAS into carbon dioxide and water and transforming the remaining part into a biodegradable solution. Dark Fenton is a type of Fenton oxidation process that occurs in a dark condition, with a moderate scavenging effect and better degradation even without UV radiation.[132–134] The Fenton process enhances sludge's biodegradation and dissolution, which enhances sludge dewatering.[135] However, this Fenton integration currently has some limitations, including the need for adjustment of pH, dose, and concentration of hydrogen peroxide and the generation of a high amount of sludge at the end of the process, which greatly restricts its use. Moreover, due in part to new restrictive legislation on sludge disposal and economic evaluations of wastewater treatment procedures, these flaws prevent the technology from being widely used.[136] On top of that, Fenton can face some issues in sludge disintegration, such as the accessibility of •OH through the microbial cells and sludge blocks,[137] which in some cases requires the high use of Fenton agents. Hence, the single use of disintegration of the Fenton and ultrasonic processes presented several difficulties, including high energy consumption and high reagent dosages that resulted in low efficiency. The related drawbacks must be addressed to make the technology more sustainable, as they are the main barrier to its general use.

Ultrasonic combined with Fenton is an advanced oxidation process that has been used for water and wastewater treatment, especially for sludge treatment.[46,106,138] For instance, Qiu et al.[139] found that using ultrasound as a pretreatment for the Fenton-acclimation treatment increased the sludge reduction rate from 26.53 to 63.59% with an operating cost reduction of 51.46%, showing that ultrasonic irradiation was efficient in enhancing both sludge reduction efficiency and operating cost-effectiveness. Ning et al.[140] studied the effect of ultrasound-assisted Fenton treatment on the physicochemical properties of textile dyeing sludge, and the results showed that the ultrasound-assisted Fenton treatment sludge presented obvious advantages over the Fenton-treated sludge for disrupting sludge floc structure, but its efficiency depends upon the ultrasonic density and pH, needs to optimize these condition to apply in large scale. Bao et al.[138] studied the physicochemical pretreatment methods for waste-activated sludge hydrolysis and acidification, and they found that the ultrasonic and Fenton treatments combined had a synergistic effect on enhancing the hydrolysis and subsequent acidification of activated sludge. In general, due to the increased production of OH• radicals by cavitation during ultrasonic irradiation, ultrasound can speed up the pace at which Fenton oxidizes. Additionally, the use of OH• radicals produced by either transient cavitation or Fenton's reagent is made easier due to the strong micro-mixing caused by a variety of mechanisms, such as microstreaming through microturbulence and ultrasound propagation via cavitation bubbles. However, it is still too early to apply to sludge disintegration management. It requires further research investigation on the development of optimized conditions that enhance sludge digestibility and dewaterability technologies, including low consumption and high efficiency of simple process and environmentally friendly using life cycle assessment to make the process more sustainable, as well as on the use of these approaches to nutrient recovery and energy production in the context of the circular economy concept.

In general, sodium citrate is used as a cation binding agent (CBA) to boost the solubilization of WAS flocs, improving the disintegration process. Specifically, CBA plays a crucial role in removing the bridging cations in sludge, such as Ca, Mg, Fe(III), and Fe(II), which provokes the release of proteins, carbohydrates, and humic matter from the sludge mass. The release of these products is important to enhance the generation of methane. However, this chemical process requires the use of large volumes of sodium citrate with relatively high concentrations to obtain effective treatment. Ultrasound-assisted sodium citrate treatment could lead to synergistic effects, allowing an effective and fast sludge treatment. JR Banu et al.[141] reported that the combination of sodium citrate and ultrasound leads to boost the production of methane, wherein biomethane yield was found to be in combined ultrasound-assisted sodium citrate (0.26±0.009 L/g COD) in comparison to ultrasound (0.145±0.006 L/g COD) and sodium citrate (0.03±0.004 L/g COD) (Figure 5).

A recent study by Phong et al.[142] examined the combined effects of ultrasound in conjunction with various disruptive chemical methods such as H_2O_2 oxidation, ionic liquid extraction, supercritical fluid extraction, alkaline/acid treatment, and

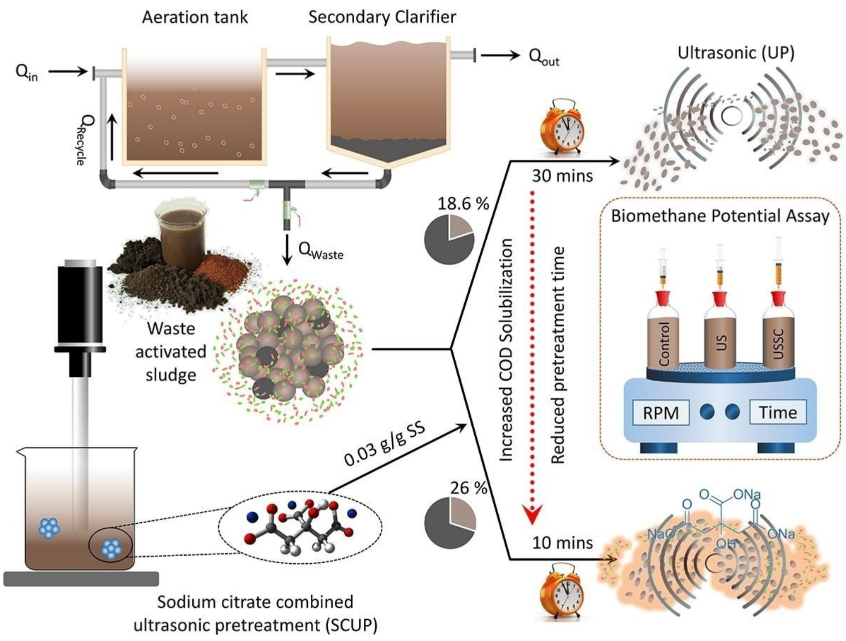

Figure 5. Ultrasound-assisted sodium citrate for enhanced biogas production, reproduced with permission from Elsevier (n. 5019019934 July 16, 2024).[141]

ozonation. Simultaneously applying both procedures greatly improved the release of carbohydrates and lipids, resulting in extraction yields of lipids and carbohydrates almost twice as high as those achieved with ozonation alone. The lipid extraction yields also increased from 46.5% to 59% compared to the exclusive utilization of ultrasound. The use of H_2O_2 in combination with ultrasonic treatment resulted in an increased efficiency of cell lysis. Overall, combining ultrasonic and advanced chemical techniques efficiently decreases energy usage, chemical quantities, and operation time while also recovering key intracellular components. This makes it a practical option for large-scale commercial extraction in microalgal biorefining.[143] Table 1 summarizes several studies regarding the recombination of ultrasound with other agents/technologies for enhanced sludge disintegration, biogas production, and saving energy and chemicals.

Table 1. Combination of ultrasound with other technologies for enhanced sludge disintegration

Combined process	Observations
Fenton + US	The presence of US leads to higher performance at lower H_2O_2 and Fe^{+2} and shorten time. Effective destruction of the microstructure of sludge was obtained due to enhanced generation of •OH via the Fenton-US system.
Fenton + US	The introduction of US to Fenton increases the generation •OH from 0.26 to 0.43 mM. Fenton + US provide significant higher concentration of soluble chemical oxygen demand (SCOD), total organic carbon (TOC) and extracellular polymeric substances (EPS) in sludge supernatant.
Fenton + US	Increase of •OH and effective release of carbon, nitrogen and phosphorus by combined Fenton + US
Fenton + US	Effective odours elimination by combined Fenton + US.
Alkaline + US	The combination of Alkaline + US leads to significant increase in the solubilization (SCOD/TCOD) and speeds up the disintegration process.
Alkaline + US	Boosted dissolution of chemical oxygen demand and use of low NaOH quality and short time as compared to bare alkaline system. The destruction of organic matter improved from 38.0% to 50.7%.
Acid + US	The combination of acid and ultrasound allows fast and effective sludge disintegration as compared to single systems.
O_3 + US	O_3 + US results in a high degree of composting quality which was reflected by superior reduction in volatile solids, total organic carbon, C/N ratio, total coliform along with the enhanced availability of nutrients elements.
Thermal + US	Thermo-sonication led to destroy hydrolysis barriers of the sludge and enhanced the anaerobic digestion compared to single systems. The generation of biogas improved by 19%, while the removal of volatile solid increased by 50%.
Coagulants + US	The combination of US with different coagulants ($Al_2(SO_4)_3$, $FeCl_3$, cationic polyacrylamide and combined ferric chloride and lime) led to significant increase in sludge dewatering. Cationic polyacrylamide and combined ferric chloride and lime combined with US showed better performance.
Microwave + US	Combined microwave–ultrasonic treatment showed excellent release of organic matter and digestion performance compared to single process. Methane production was found to be 147 mL by combined, while US and microwave alone showed methane production 30 and 16 mL, respectively.
K_2FeO_4 + US	The use of K_2FeO_4 under US irradiation provides excellent sludge disintegration. K_2FeO_4 can be cheap and safe agent alternative to O_3. K_2FeO_4 byproducts leads to high flocculation and settleability of the sludge.
Sodium citrate + US	COD solubilization was increased by 26% when the combined system is used. A significant increase in biogas of about 50% was recorded in combined system along with huge decrease in energy consumption compared to US alone.

4. US Reactors Available in the Market for Sludge Disintegration

Variables such as reactor geometry, intensity, and frequency affect cavitation activity in ultrasonic reactors. Although the effects of frequency and intensity on sewage sludge sonication have been well studied, there have been few studies on the influence of reactor geometry. Design factors are significant for sludge pretreatment. An increased concentration of solids in sludge results in the absorption of sound waves, and subsequently, the formation of cavitation decreases as the distance from transducers increases. Increased viscosity inhibits the mixing caused by cavitation, leading to uneven exposure of the sludge during treatment. Ultrasonication is commonly performed using flow-through reactors that rely on fluid dynamics within the reactor. In unfavorable reactor designs, the flow condition might worsen the unevenness of the treatment. This is because the flow rates are likely to be the highest in the middle of the reaction chamber, where the cavitation field is the least intense.[155] Prior research has demonstrated that the configuration of US reactors in the United States substantially influences the breakdown of sludge, with full-scale US reactors exhibiting more than double the disintegration efficiency compared to laboratory-scale batch systems.[156]

Nevertheless, the effectiveness of sonication differed significantly among the settings examined, even if the energy inputs remained similar. The lack of a systematic approach to reactor design and structural variations among reactor systems prevents the establishment of a definitive correlation between geometry and performance. These findings emphasize the significance of considering the geometry of US reactors when assessing the effectiveness of sludge disintegration.[155]

Extensive research has been conducted on ultrasonic pretreatment methods. However, it is important to note that most of these studies have been carried out in laboratory environments, which do not accurately reflect the conditions observed in full-scale wastewater treatment facilities (WWTPs).[157] Full-scale wastewater treatment plants (WWTPs) often use modest amounts of energy, because they handle massive amounts of sludge and have short treatment durations. During laboratory experiments, the amount of energy applied varies between 1.000 and 16,000 kJ/kgTS, with certain cases reaching values as high as 27,000 or 108,000 kJ/kgTS.[158] Because treatment effects are generally enhanced as energy input increases, the applicability of lab-scale investigations to full-scale applications is restricted. In addition, the test methodology employed in laboratory-scale investigations differs from that used in full-scale digesters, as most are designed as continuous systems.[159] Moreover, obtaining data on treatment economics and operational stability, such as the clogging behavior and lifetime of sound-emitting surfaces, is challenging to achieve through lab-scale investigations alone. This necessitates testing in full-scale WWTPs.[160]

Although there is a need for comprehensive trials, few studies have been conducted on the use of pretreatment for raw sewage. A study conducted by Xie et al.[161] showed that utilizing a 20 kHz radial horn reactor at the Ulu Pandan Water Reclamation Plant in Singapore significantly increased methane yield, ranging from 13% to 58%. Neis conducted another comprehensive experiment at the Bamberg WWTP in Germany, showing that biogas production and volatile solids degradation rose from 1.5 million cubic meters per year to 2.2 million cubic meters per year, from 34% to 50%, respectively.[155] This facilitated consistent digestion despite the reduced hydraulic retention time (HRT). However, it was challenging to ascertain whether the beneficial progress was exclusively attributed to US pre-treatment since no control digester was employed.[162]

Ultrasonic WAS pretreatment has been found to have minimal effects on biogas production and organic matter degradation in water treatment plants. A second full-scale trial at the Swedish Ernemar WWTP did not show significant anaerobic digestion enhancement owing to repeated reactor malfunctions. Operational challenges were also encountered, with the radial horn reactor at the Ulu Pandan Water Reclamation Plant exhibiting clogging susceptibility and requiring manual cleaning. Previous full-scale trials were not always peer-reviewed and faced inconsistent methodologies, making interpreting the current information on full-scale sonication tests challenging.

Over the last two decades, many efforts have been made to transfer the ultrasonic process to full-scale real-world applications. Several requirements for successful industrial-scale application must be fulfilled, including large volume processing, efficiency and timeliness, low-cost process, and low maintenance. To solve such concerns, the reactor configuration and mixing degree and the capability of transducers are key parameters to design a US plant for long-term, large-scale applications. DesiUS, an abbreviation of "Disintegration Ultrasound System", was developed by Weber Ultrasonics manufactory in collaboration with the Fraunhofer Institute (Germany), and then it was optimized by Entec via the use of the BioPush ultrasonic reactor. BioPush, the technological heart of DesiUS, is a maintenance-free, simple structure and high-quality machine according to DIN EN ISO9001:2015. Unlike conventional rod transducer-based US systems, which can treat only the species in the immediate proximity of the ultrasound source, DesiUS is able to produce extended and homogeneous cavitation field via powerful planar transducers as shown in Figure 6a–d. For enhanced and proper ultrasonic sludge treatment, multi-BioPush-based systems can be designed. In the turnkey constructed DesiUS, the BioPush US reactors are fed via a progressive cavity pump. The sludge is routed at a given speed inside the BioPush reactor, wherein it is subjected to a powerful ultrasonic field so that the provided energy input needed to break down the sludge flocs is properly accomplished. A mechanical pre-treatment macerator protects the equipment from foreign objects like stones and ensures a high homogenization of the sludge for effective ultrasonic treatment. The plan is equipped with a PLC-based control system, temperature, pressure and volume sensors for better and easy control. Over 100 Weber Entec DesiUS were installed worldwide in WWTPs to purify sludges and for biogas production. A test Weber Entec DesiUS equipped with 6 BioPush of US power of 2 kW was integrated in 2013 at a Swiss WWTP with a population capacity of 80000. After a year, enhanced gas production of 17%, improved flow properties, and reduced sludge mass, Weber Entec DesiUS was tested on waste-activated sludge at Moscow WWTPs (Kuryanovsky and Luberetsky). It was reported that the ultrasonic treatment of 40% of waste-activated sludge at 2.2–2.5 kWh/m^3 energy enhanced gas yield up to 17%, while a 10% reduction of volatile solids during the digestion was found. According to their report, this technology is very economical, easy to proceed, and worth further consideration for implementation compared to conventional technologies. DesiUS, equipped with 14 US reactors, was installed in WWTP in Lithuania in 2017. In this station, the biogas (methane) production of ultrasonic-treated and non-treated samples was evaluated, and it was found that the gas yield was enhanced by up to 34% after ultrasonic treatment in the DesiUS machine. Many WWTPs benefited from DesiUS in treating sludges in different countries such as Germany, Switzerland, Italy, Thailand, Russia, Singapore, Lithuania, and many individual installations.

At the lab scale, Lippert et al.[163] recently studied comparatively the dynamics of water, thickened waste-activated sludge, and digested sludge within an ultrasonic flatbed reactor (BANDELIN electronic GmbH & Co. KG, Berlin, Germany), which is similar to DesiUS reactor. The visualization of the flow was investigated via the injection of dye streams into the flow. The authors found out that the ultrasonic effect in a flatbed reactor causes a strong fluid dynamic disturbance in water (turbulent flow). However, in sludge flow, the dye mainly remained in laminar form, which indicates that sludge flow is nearly laminar in ultrasonic flatbed reactors (Figure 6e).

The Austrian VTA group developed and patented the VTA GSD device for the ultrasonic disintegration of sewage sludge.[164] The VTA GSD system is focused on destroying sludge flocs via ultrasonic oscillators with low energy usage. It is a pressure-free and fully automatic operation, allowing easy integration into any sludge purification station. The device consists of a container with a mechanical agitator and rod-shaped US transmitters parallel to the container's axis (Figure 7a and b). High-quality VTA GSD sonotrodes consist of titanium and stainless-steel components designed for a long time use over 45000 h. Figure 7c shows a photograph of sonotrodes used for 6000 h at WWTP of Hajdów, Lublin (Poland). Micro-

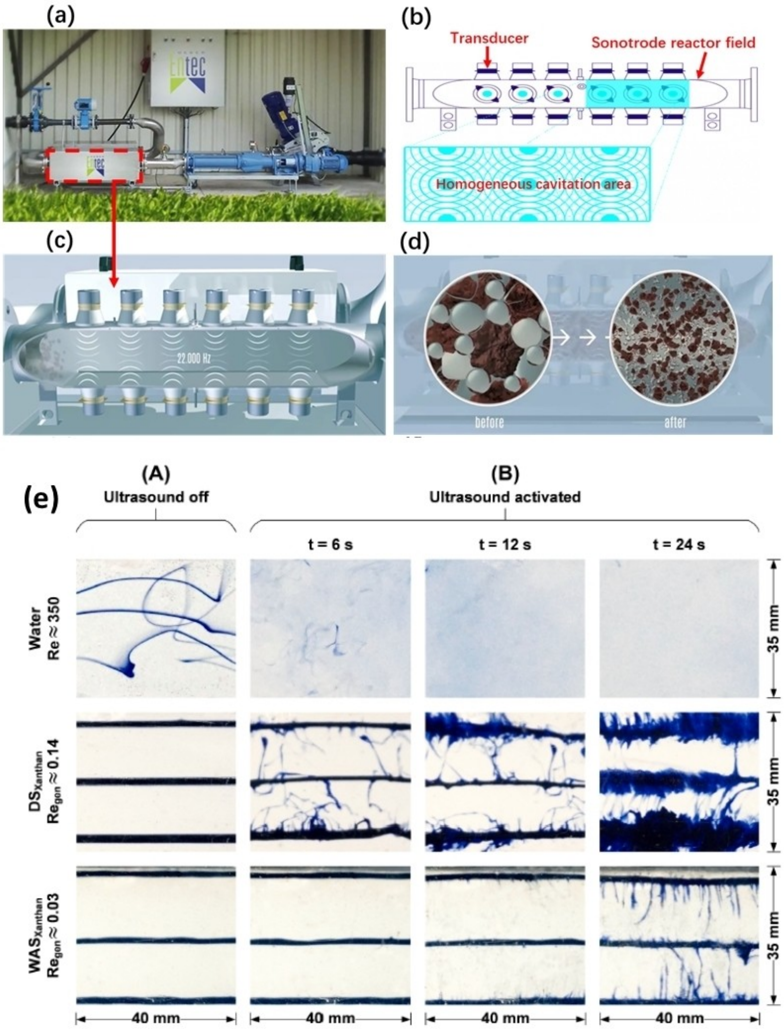

Figure 6. (a): Weber Entec DesiUS with single US reactor; (b-d) Operation of US reactor for sludge disintegration. (e): Images showing flow dynamics behavior of water and sludges in ultrasonic flatbed reactor, reproduced with permission from Elsevier (n. 5830811158412 July 16, 2024).[163]

mirror photographs of two types of sludge flocs before and after ultrasonic treatment by VTA GSD system (×100) as shown in Figure 7d. VTA GSD technology has mainly been applied worldwide in WWTPs, such as in Austria, Germany, Italy, Lithuania, France, Switzerland, Spain, South Korea (Figure 6e), Belgium, and Poland. Very encouraging results have been reported, wherein up to 29% more gas production and 19% lower sludge rates were found. On top of that, the use of this system led to avoiding digester foaming.

ULTRAWAVES ultrasound reactors were designed by ULTRAWAVES company (Germany) in collaboration with SONOTRONIC Nagel GmbH (Germany).[165] The standard ULTRAWAVES reactor (Figure 8a) consists of a 30 L container with five ultrasound oscillating units (each has a converter, a booster, and a sonotrode), which produces a continuous typical output of 1000 Watts, a frequency of 20 kHz, and intensity of 25 to 50 W/cm^2. ULTRAWAVES systems generate a hard cavitation within a few seconds, capable of destroying activated and filamentous sludges. It operates as a plug flow system with a quick volumetric flow rate of streaming medium up to 2 m^3/h. ULTRAWAVES can run continuously (24/7). ULTRAWAVES systems have been applied successfully to digest WWTP sludge. In 2005, it was installed in Meldorf WWTP, Germany, to overcome the foaming formation observed in the station due to the huge growth of filamentous bacteria (*Microthrix parvicella*) in WAS.[166,167] The foaming sludge was no longer an issue after a short time of integration of the US reactor.

Additionally, a 60% to 45% reduction in the volatile solid was observed for the period of tests, while an improvement of 30% was evaluated in biogas production. The first long-term application of the ULTRAWAVES plan for sludge digestion appeared at Bamberg STP.[168] Today, ULTRAWAVES plans are installed in many countries, including Germany, Denmark, Italy, Austria, Great Britain, Netherlands, Hong Kong, China, Japan, USA, India, Australia, Brazil, etc. Many experimental research studies and real tests have led to the development of a patented high-power ultrasound system.[165] A new ULTRAWAVES design was fabricated, as shown in (Figure 8b) for enhanced sludge disintegration and lower maintenance. In this system, the sludge is subjected successively to hard cavitation

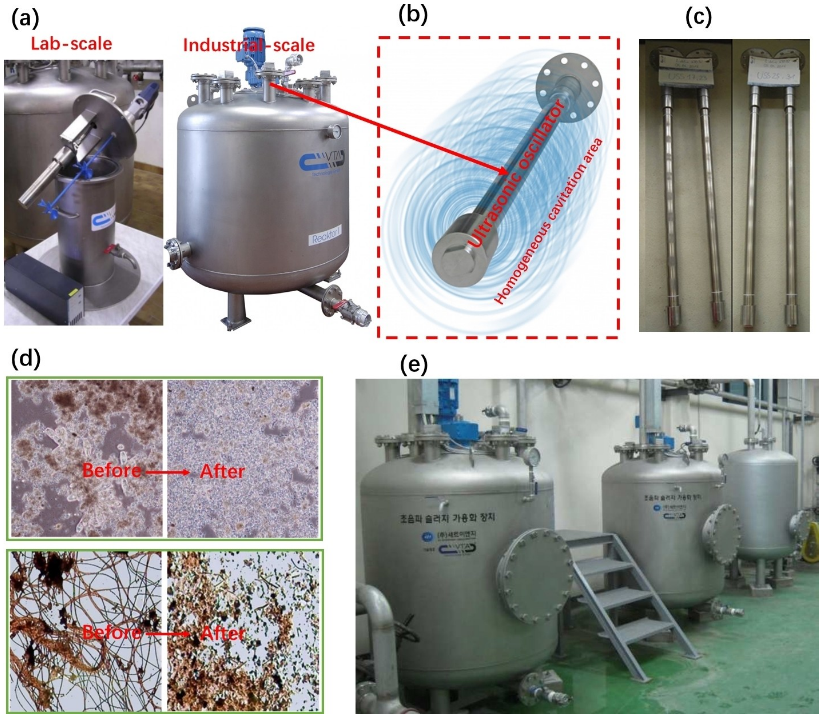

Figure 7. (a): Examples of lab-scale and industrial-scale of VTA GSD devices for the ultrasonic sludge disintegration; (b): VTA GSD sonotrodes; (c): Photograph of VTA GSD sonotrodes after using for 6000 h at WWTP of Hajdów, Lublin (Poland); (d): Micromirror photographs of two types of sludge flocs before and after ultrasonic treatment by VTA GSD system (×100); (e): VTA GSD plan installed in WWTP in South Korea

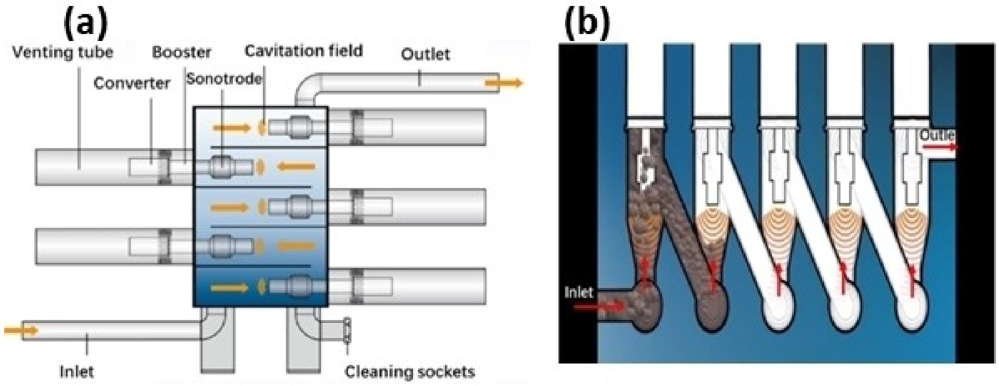

Figure 8. (a) scheme shows the ULTRAWAVES ultrasound reactor; (b): A new ULTRAWAVES design for enhanced sludge disintegration and avoiding the accumulation of sludge in the individual chambers.

when it strikes the front of the sonotrodes from the left to the right. Compared to the traditional ULTRAWAVES reactor, this new design prevents the deposition of sludge mass within the individual chambers, suggesting less maintenance. In terms of efficiency, the vibrating structure works effectively on the sludge particles facing the hard cavitation field in the opposite direction.

Hielscher Ultrasound Technology Company (Germany) has developed several lab-scale and industrial ultrasonic devices for different applications (Figure 9). Regarding sludge disintegration, over the last two decades, Hielscher has been supplying US disintegration devices of up to 48 kW individual power to municipal and industrial waste purification worldwide. Hielscher Ultrasound Technology systems can improve biogas production by up to 25 %. To fulfill the requirements for efficient and fast sludge disintegration or water treatment, Hielscher designed very powerful ultrasonic processors, such as the UIP16000 processor, which applies 16000 watts of ultrasonic power as the most powerful processor in the world. It can work effectively for large volume processing and higher energy demand in WWTPs up to 200 m^3/hr using a 62xUIP16000 system.

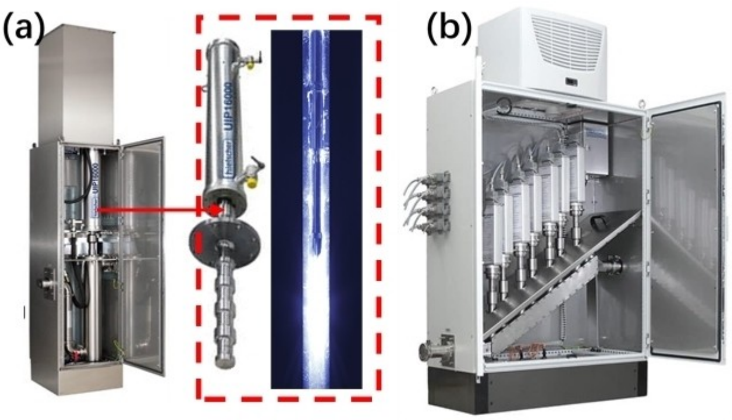

Figure 9. (a): Hielscher industrial ultrasonic device for sludge disintegration equipped with one UIP16000 processor (16000 watts; Flowrate: $>10\ m^3/h$), (b): Hielscher industrial ultrasonic system equipped with 7×UIP1000hd processor (1 kW).

5. Conclusions and Perspectives

This article systematically reviewes the application and process mechanism of the ultrasonic method in municipal sludge treatment, and the typical ultrasonic reactors used in full-scale real-world applications are also summarized. Generally, the use of ultrasonic technology to treat sludge is conducive to sludge reduction, harmlessness, and recycling, which have social, environmental, and economic significance. Particularly, low frequency and low dose ultrasonic treatment of sludge can enhance the sludge activity, change sludge floc structure, release the bound water in the sludge microbial micelles, and improve the sludge dewatering performance. Meanwhile, high-intensity ultrasound can kill bacteria in sludge, eliminate viruses, and decompose substances that generate odor, thus eliminating the source of odor, killing algae, eliminating suspended solids, and improving the solubility of COD. It has also been demonstrated that the shear force generated by ultrasonic cavitation breaks the cells and releases proteins and amino acids. As a result, the content of ammonia and nitrate nitrogen also increased with increasing ultrasonic time. The same phenomenon was also reported phosphorus compounds. Compared with chemical disinfection, the method not only prevents chemical accumulation but also enhances sludge stability for extended periods and effectively prevents the spread of pathogenic bacteria.. Currently, research on ultrasonic sludge reduction mainly focuses on the influence of ultrasonic conditions on promoting anaerobic and aerobic sludge reduction, as well as the influence of ultrasound on the physical, chemical, and biological properties of sludge . Biogas production from sludge can be significantly improved by ultrasonic-assisted treatment because of the enhanced release of proteins and organic matter, as well as boosting the hydrolysis of long-chain fatty acids. The performance of ultrasound-induced sludge disintegration process depends on several factors. Based on the initial nature of the sludge and its composition, the optimization of parameters depends on several factors, including the composition of the sludge and its physicochemical nature, the required industrial treatmentand the geometry of the ultrasonic reactor. Ultrasonicprocesses can be used solely for sludge denitigration or in other systems for advanced treatment and enhanced biogas production. Due to the complexity of sludge composition and the multifaceted factors affecting ultrasonic treatment, ultrasonic treatment of sludge is an extremely complex process, especially since the mechanism remains to be further studied, and sludge cracking technology needs to be further developed and improved. Currently, the main problems in the application of ultrasound to sludge reduction are the optimization of the operating parameters of ultrasound treatment, the improvement of ultrasound efficiency, and the reasonable design of ultrasound reactors. At the same time, we should pay attention toa reasonable combination with the sewage treatment process to give play to the characteristics of ultrasound and lay a foundation for its application in practical projects. Future studies should focus on several important topics. One area of interest is further investigation into the mechanism and influencing factors of ultrasonic sludge treatment, particularly the effects of interactions with different frequencies . Additionally, research can explore the joint application of ultrasonic technology with other technologies to enhance the overall treatment process.

Another key area is to address the critical challenges in transitioning ultrasonic reactors from laboratory settings to practical applications. This includes solving issues related to the materials, design, power consumption, treatment effectiveness, and economic costs. Designing innovative reactors for direct ultrasonic treatment or in combination with other technologies is also crucial. For example, combining ultrasonic processes with chemical agents can lead to faster and more cost-effective treatment. A promising technology in this regard is the homogenous activation of PMS in the presence of ultrasonic irradiation, which can significantly enhance sludge disintegration and detoxification.

Finally, future studies should evaluate the economic and sustainability aspects of ultrasonic sludge pretreatment. This includes assessing biogas production against the expenses and energy required to operate the ultrasonic devices.

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Conflict of Interests

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords: Sludge disintegration · Sono-pretreatment · Sludge anaerobic digestion · Sonochemical reactors · Biogas production

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