OPTIMIZATION OF THE EFFICACY OF SLUDGE ULTRASONIC PRETREATMENT: A REVIEW

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ABSTRACT

Ultrasonication is an advanced technology in sludge pretreatment, almost thanks to hydro-mechanical shear forces created in cavitation. However, there are many factors affecting the efficacy of cavitation and consequently ultrasonic disintegration of sludge. This work aimed at selection, assessment, and indication of important parameters influencing these mentioned processes. Optimization methodologies of related parameters, the differences of optimum values as well as the similarities of effecting trends on cavitation and sludge pretreatment efficiency were specifically pointed out, including ambient conditions (temperature, external pressure), ultrasonic properties (frequency, power input, density, intensity, specific energy input, duration), and sludge characteristics (sludge type, volume of sludge, total solids concentration, pH). The research is a prerequisite for optimization of sludge ultrasonic pretreatment efficiency in lab-scale and practical application.

Keywords: cavitation, Combined pretreatment, Optimization process, Ultrasonic pretreatment, Waste activated sludge

1. INTRODUCTION

The first objective of sludge treatment is to remove organic matters and water, which reduces the volume and mass of sludge and also cut down toxic materials and pathogens. Biological, mechanical, chemical methods and thermal hydrolysis have been listed as popular techniques for sludge pretreatment [1]. Among these techniques, anaerobic digestion (*AD*) is the most traditional one. Nevertheless, this process is limited by long sludge retention time and rather low overall degradation efficiency. Sludge mainly consists of microbial cells which walls limit the biodegradability of intracellular organic matters [2]. Therefore, sludge disintegration pretreatment, which disrupts sludge flocs, breaks cell walls and facilitates the release of intracellular matters into the aqueous phase, can be considered as a simple approach for improving rate and/or extent of degradation.

Ultrasonication (*US*) is a promisingly applicable mechanical disruption technique for sludge disintegration and microorganism lyses. Nevertheless, *US* requires high energy input and causes great discussions due to economic issues in practical application. This high cost could be reduced by the combination with other pretreatment methods, the adjustment of sludge properties such as total solid content (*TS*), pH and volume of sludge, and/or the optimisation of ultrasonic parameters such as frequency, intensity, density, specific energy input (*ES*), temperature *(T),* and external pressure, etc.

This work aimed at presenting main factors affecting cavitation, subsequently the efficacy of sludge ultrasonic pretreatment. Moreover, methodologies of optimizing these parameters carried out by recent researches were collected, analyzed, and found their relations serving the optimization of whole sludge ultrasonic pretreatment process in lab scale as well as in actual application.

2. BRIEF BACKGROUND OF ULTRASONIC PRETREATMENT OF SLUDGE

The piezoelectric generator is one of the most common techniques for generating ultrasound. This apparatus is comprised of three major parts: converter, booster, and horn (or probe). In *the converter* (transducer), the piezoelectric ceramics is put in the electric fields with varying polarity which causes changes in its dimension. These repeated changes create ultrasound of a specific frequency. *The booster* is designed to control (increase or decrease) the amplitude of the ultrasonic energy before it is delivered to the liquid through the horn (sonotrode). These three parts are stacked by clamping at the nodal points of either the converter or the booster. *The horn,* like the booster, also contributes to the amplification of the *US*; therefore the half or full wavelength design of the horn depends on the application of this apparatus. Furthermore, the design of the horn, enhanced by the power input levels, impacts on the intensity of the sonication, which indicates the magnitude of the ultrasonic motion, in other words, the amplitude of the vibration. An example of *US* set-up and the diagram of sonication range are presented in Fig. 1 and Fig. 2, respectively.

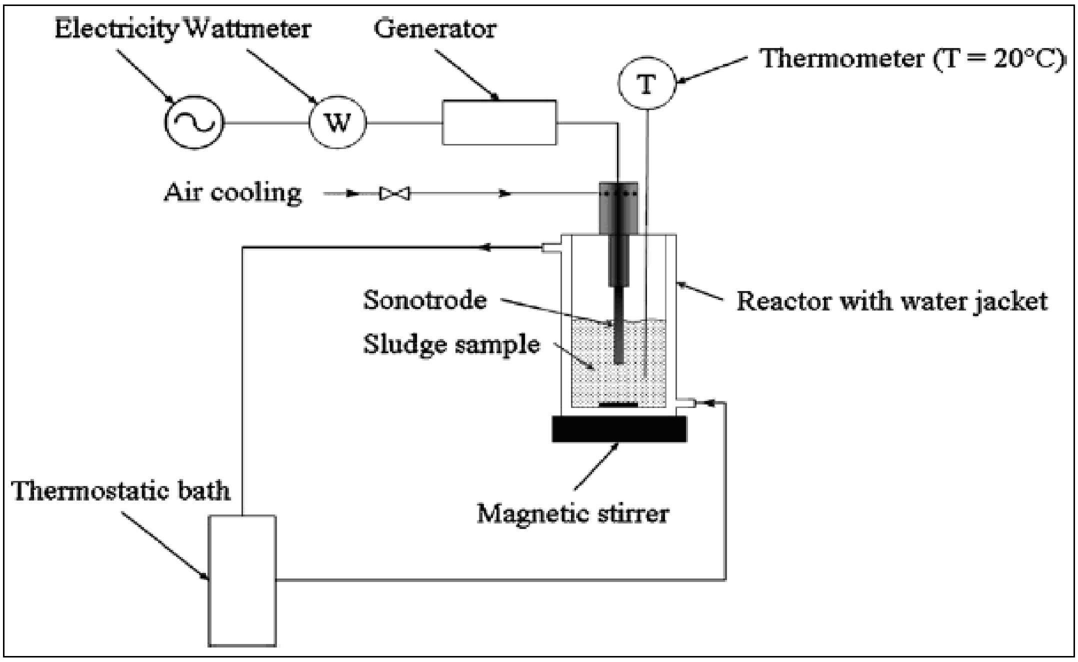

Figure 1. Ultrasonic set-up [3].

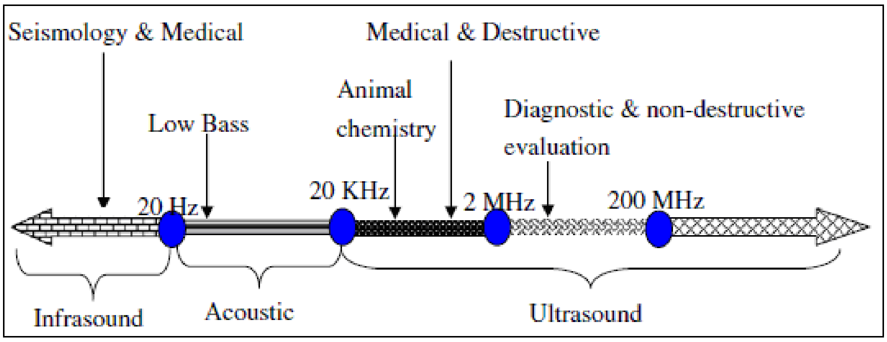

Figure 2. Diagram of ultrasound range [4].

When propagating in a solution, ultrasound waves generate compressions (they cause a positive pressure on the liquid by pushing molecules together) and rarefactions (they cause a negative pressure by pulling molecules one from each other). If a sufficiently large negative pressure is applied during rarefaction, acoustic cavitation will take place [5].

It is now clearly stated that most of ultrasound outstanding effects are due to acoustic cavitation. Acoustic cavitation is a very complex highly non-linear phenomenon which occurs at given acoustic pressure conditions (needing rather high ultrasound intensity, > 1 W/cm^2 in water at room conditions). Micro-bubbles are generated from nuclei -favored by dissolved gas, wall defects, and liquid impurities- during the low pressure half periods (bubble formation and expansion). They may oscillate a few periods, undergoing a slow average growth due to the so called “rectified diffusion” process (up to several μm) and suddenly, reaching a critical size, they dramatically grow during the low pressure half period and collapse violently in a very short fraction of the high pressure half period. Most often the bubble breaks up after the collapse point, giving smaller bubbles ready to reproduce the same scenario: oscillatory growth, driven by rectified diffusion, then sudden collapse (as schematized on Fig. 3).

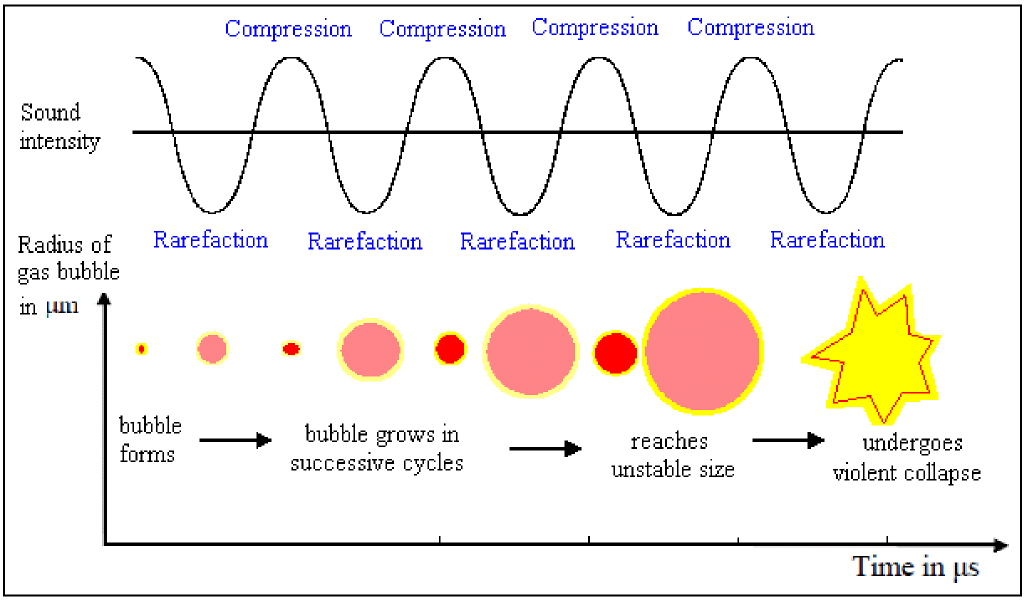

Figure 3. Formation and collapse process of a cavity.

There are two different bubble behaviors known as stable bubbles and transient bubbles. *Stable bubbles*, generated when the highest sound pressure in the rarefaction cycle is not strong enough to cause the bubble collapse, normally oscillate about thousands of acoustic cycles. *Transient bubbles* mention the conditions where the acoustic pressure can make the cavities expand their sizes of at least twice their initial sizes before collapsing violently and rapidly only after a half or several acoustic cycles on compression [6]. The final collapse leads to a temperature as high as 5000 K at the bubble center, a pressure of 500 bar, and a high radial velocity -up to the sound speed- then shock waves at the bubble rebound [7].

When applied to solid suspension and especially for sludge treatment the power/energy may be expressed in many ways as given in table 1: specific energy input *ES*, *US* dose, *US* density, and *US* intensity.

Table 1. Expressions of US energy for sludge disintegration.

No.	Parameter	Expression	Unit	Reference
1	Specific energy input	$ES = (P_{US} * t) / (V * TS)$	kJ/kg_{TS}	[8]
2	Ultrasonic dose	$DO_{US} = P_{US} * t / V$	J/L	[9]
3	Ultrasonic density	$D_{US} = P_{US} / V$	W/L	[9]
4	Ultrasonic intensity	$I_{US} = P_{US} / A$	W/cm^2	[10]

P_{US}: power input (kW), t: sonication duration (s), V: volume of sludge (L), TS: total solids concentration (kg/L), A: surface area of the probe (cm^2)

Wang *et al.* 2005 cited by Pilli *et al.* [4] indicated that the mechanisms implied in *US* sludge disintegration are hydro-mechanical shear forces, oxidizing effect of OH , H , N , and O produced under *US*, and thermal decomposition of volatile hydrophobic substances in the sludge due to the increase in temperature during sonication. The effect of hydro-mechanical shear forces is nevertheless much higher than that of radicals.

3. OPTIMIZATION OF ULTRASONIC PRETREATMENT OF SLUDGE

As mentioned, sludge pretreatment aims at disrupting sludge flocs, rupturing cell walls, and facilitating the release of intracellular matters into the aqueous phase. *Ultrasonic irradiation (US)* is known as a feasible and promising mechanical disruption technique for sludge disintegration and microorganism lyses according to the treatment time and power, equating to specific energy input. Several positive characteristics of this method are efficient sludge disintegration, improvement in biodegradability and bio-solids quality, increase in biogas/methane production, no chemical additives, less sludge retention time, and sludge reduction.

The ambient conditions of the reaction system can significantly affect the intensity of cavitation; consequently affect the efficiency (rate and/or yield) of reaction. Different conditions resulted in different effectiveness of sludge ultrasonic pretreatment. The cavitation process is influenced by many factors: gas and particulate matter, solvent, field type, types of ultrasound cavitation, applied frequency, sonication density, acoustic intensity, attenuation, temperature, external applied pressure, and sample preparation, *etc.* [4, 5, 11]. This work aims at presenting main parameters significantly affecting the cavitation in order to contribute optimization of sludge ultrasonic pretreatment efficacy.

3.1. Ultrasonic frequency

The US frequency has a significant effect on the cavitation process because it alters the critical size of the cavitation bubble [11]. According to Minneart (1933) cited by Lorimer and Timothy [5], not all bubbles are capable of producing significant cavitation effects. The greatest coupling of the US energy will occur when the applied US frequency is equal to the natural resonance frequency of a bubble; for greater applied frequencies, oscillations will be complex; but for less applied frequencies, collapse can occur.

When US frequency is increased, the production of cavitation decreases. In qualitative terms, it may be argued that at very high frequency, where the rarefaction (and compression) cycles are very short, the finite time required for the rarefaction cycle is too short to permit a bubble to grow to a size sufficient to cause disruption [5]. On the other hand, if a bubble was produced during rarefaction, the compression cycle occurs faster than the time required for the bubble to collapse [11].

Moreover, higher frequencies require more power for an equivalent amount of chemical work, since the higher rates of molecular motion at higher frequencies result in greater power losses. Power required to make water cavitate at 400 kHz was ten times as many as that at 10 kHz. 20 - 50 kHz of frequency was generally chosen for cleaning purposes and had subsequently been found to be suitable values in sonochemistry [5]. In addition, according to Entezari et al. (1997) cited by Thompson and Doraiswamy [11], this range was chosen because the alteration of frequency had no apparent effect in several reactions, such as in the dissociation of carbon disulfide.

Effects of US frequency were investigated by Yoshiyuki *et al*. [12] at values of 45, 129, 231, and 490 kHz as well as by Rooze *et al.* [13] at values of 20, 41, and 62 kHz. The findings of Zhang *et al.* (2007, 2008) cited by Pham *et al*. [14] showed that low frequency (25 kHz) was more effective than higher ones (80 and 150 kHz), or in another aspect, higher US energy was more efficient than lower US energy for sludge treatment, indicating mechanical effects, instead of free radicals, to be responsible for the bioactivity enhancement.

Owing to the increase in frequency, the *degree of sludge disintegration* (DD_{COD}) as well as the VS reduction decreased. Corresponding to the US frequency of 41, 207, 360, and 1068 kHz (with identical US densities for 60min), the values of DD_{COD} were 13.9, 3.6, 3.1, and 1.0%, respectively; and VS reduced by 32.2, 28.9, 26.3, and 25.2 %, respectively [9].

In summary, lower US frequency produces more violent cavitation, leading to higher localized temperatures and pressures at the cavitation site. However, higher frequencies may actually increase the number of free radicals in the system, subsequently facilitate the bulk reaction.

3.2. Temperature

Theory-based, increasing temperature will decrease surface tension and raise the equilibrium vapour pressure of the medium (and so lower both T_{max} and P_{max}), leading to easier bubble formation (due to the decrease of the cavitation threshold). However, these kinds of cavitation bubbles contain more vapors which reduce the US energy produced by cavitation because they cushion the implosion. Besides, great numbers of cavitation bubbles generated simultaneously will be the attenuation or dampening effect on the propagation of US energy from the emitter through the system. Increasing the temperature was also simultaneously decreasing the intensity of cavitation, thus reducing the amount of free radicals produced within the bubble [5].

Nevertheless, in terms of sludge disintegration, it is important to note that there is an opposite trend: sludge ultrasonic pretreatment efficacy increases following an increase in the bulk temperature.

It was proved that the US treatment has two simultaneous effects: (i) vigorous agitation caused by the formation and explosion of tiny bubbles, and (ii) the increase in the bulk temperature. To separately evaluate their effects, Le *et al*. [15] carried out four operating procedures: (1) without US + controlled T (28^{o}C), (2) US + controlled T (28^{o}C), (3) US + uncontrolled T, (4) without US + progressive increase of T up to 77^{o}C (with same T rise profile as found with US in (3) to see the effect of thermal hydrolysis). After about 2 hours of stirring at a constant temperature and without US, DD_{COD} was very low (0.3%), which indicated that the stirrer played a main role in making a homogeneous solution, but did not significantly affect the release of COD. At all observed time ranges, DD_{COD} values under adiabatic sonication were the highest, followed by those obtained under low temperature sonication and thermal hydrolysis, which was also found in recent researches [3, 16, 17]**.** The authors concluded that the higher the temperature of sludge samples was, the more efficient the US disintegration was. This is opposite to most power US applications as cavitation intensity is higher at low temperature. It is then clear that US disintegration of sludge is the result of two different effects: the specific cavitation effect and the thermal effect.

It was also noted that there was a rapid increase in temperature of the bath during US: at 0.44 W/ml, the batch temperature increased over 50°C within 2 min. The same experiments (with D_{US} of 0.11W/mL and 0.33W/mL) were carried out on two sludge samples: with (15°C) and without temperature control. After certain duration of US, the SCOD/TCOD ratio in the T-controlled sample (15^{o}C) was lower than that in the T-uncontrolled one. The COD continued being produced into the supernatant up to 120 min when the bulk temperature went up to 60°C. Cavitation explosion and bulk temperature increase have equal influence on sludge floc disintegration and cell lysis [16].

Li et al. [18] also indicated that the higher the temperature caused by the increase in US duration was, the higher the efficiency of sludge US disintegration was: at 4 W/mL-1 min of sonication, DD_{COD} was 9% for both samples with (20^{o}C) and without temperature control; but at 0.8 W/mL-5 min of sonication, DD_{COD} was 27 % for the uncontrolled sample versus 23% of DD for the control one. However, the temperature effect was limited when US duration was short.

It could be suggested that for any scale up operation, in one hand, the process should be carried out without cooling to reduce the expenses of the cooling system; in the other hand, the extreme temperature must be controlled not to damage the mechanical equipments. In other words, the US system should be controlled and cooled down to the possible highest temperature in order to both take advantage of US (cavitation and temperature effects) and to maintain the effectiveness of mechanical equipments [3].

3.3. External Pressure

Changing the hydrostatic pressure can alter (*i*) the resonance frequency and (ii) equilibrium radius of the bubble and drive the system toward resonance conditions [11].

$$R\ddot{R} + \frac{3}{2}(\dot{R})^2 = \frac{1}{\rho_0}\left[(P_h + 2\sigma/R_0)(R_0/R)^{3\gamma} - \frac{2\sigma}{R} - (P_h - P_A \sin 2\pi f t)\right] \quad \text{(i)}$$

where R is the bubble radius at some time t, R_0 is the equilibrium bubble radius, P_h is the hydrostatic (ambient) pressure, $\dot{R}$ is the bubble wall velocity, and $\ddot{R}$ is the bubble wall acceleration.

Bubbles with an equilibrium radius R_0 in a liquid system with a fixed temperature T_0 and negligible viscosity will pulsate with a resonance frequency ωr, as defined by

$$\omega_r^{\,2} = \frac{1}{\rho R_0^{\,2}}\left[3\gamma\left(P_h + \frac{2\sigma}{R_0}\right) - \frac{2\sigma}{R_0}\right] \quad \text{(ii)}$$

As mentioned, when the angular frequency of the ultrasound is equal to the resonant frequency of the bubble (f=ωr), resonant cavitation occurs. Operating at resonant conditions will increase the rate and yield of reactions [19 - 21]. To math the resonant conditions while many US transducers have been designed with a set frequency, the resonant frequency of the bubble needed adjusting by varying the hydrostatic pressure [19] or the system temperature [20] to adjust. Other factors affecting the resonant frequency of the bubble were solution characteristics (density and surface tension). Conversely, varying the US frequency in order to drive the bubble dynamics toward transient cavitation was also investigated [21].

In short, increasing the external pressure *(P_h)* leads to both an increase in the cavitation threshold and the intensity of cavity collapse [5]. Qualitatively, in case P_a - P_h < 0, there is no longer a negative phase of the sound, and consequently cavitation cannot occur. Clearly, a sufficiently large increase in US intensity *(I)* can produce cavitation even at high overpressures due to generating larger values of *Pa (I § P_A^2; $P_a = P_A$ sin $2\pi ft$),* making *Pa* - *Ph* > 0. As mentioned, P_m is approximately the total of P_h and P_a, increasing the value of P_h result in more rapid and violent collapses [5].

Most US experiments have been carried out at atmospheric pressure, and only a few studies have been focusing on how increasing static pressure affects cavitation, but almost concern sonoluminescence. The findings by Finch (1955) cited by Chendke and Fogler [22] indicated that the greatest sonoluminescence intensity was observed in water at a static pressure of about 1.5 atm when varying in the range of 1-8 atm. Chendke and Fogler [22] recommended a value of 6 atm to promote sonoluminescence in nitrogen-saturated water. In aqueous carbon tetrachloride solutions, the intensity of the sonoluminescence did not show any monotonous behavior over the range of 1-20 atm: it first hiked up to 6 atm, then reached a minimum at 8 atm, got a new maximum value at 12 atm, and was finally almost inhibited above 18 atm [23]. Brett and Jellinek (1956) cited by Chendke and Fogler [22] reported the effects of pressure on cavitation bubbles; thereby cavitation bubbles could be visible for gas-applied pressure as high as 16 atm. Whillock and Harvey [24] investigated the effects of hydrostatic pressure on the corrosion of 304L stainless steel in an ultrasonic field. An increase in hydrostatic pressure up to 4 bar at constant temperature caused a strong increase in corrosion rate. Neppiras and Hughes [25] investigated the influence of pressure (up to 5.8 atm) on the disintegration of yeast cells and found an optimum value of 4 atm.

To our knowledge, the effect of pressure on sludge pretreatment has hardly been investigated. The first work in this field was conducted by Le *et al*. [15] with external pressure range of 1-16 bar. Different ES values show the same trends of DD_{COD}: initially increase up to 2 bar and decrease afterwards, noticeably at pressures over 4 bar. Compared with experiments at atmospheric pressure, sludge disintegration efficacy was significantly improved at the optimum pressure of 2 bar; e.g. the increase in DD_{COD} was by 67%, 36%, 27%, 23%, and 22% with ES values of 7000, 12000, 35000, 50000, and 75000 kJ/kg_{TS}, respectively. This approach might lead to energy savings in sludge pretreatment applications with ultrasound. Further experiments were performed to examine the effect of pressure (1-16 bar) along with temperature during US duration. Once again, the optimum pressure was found in this work to be about 2 bar regardless of temperature conditions. In addition, the authors indicated that the effect of sole pressure was less than that of sole adiabatic condition in association with US.

This phenomenon, much more important at ambient temperature (controlled T), could be explained by the decrease in vapour pressure of the mixture, the increase in cavitation threshold, and the limitation of bubble formations. Therefore, to produce cavitation at higher static pressures, the acoustic pressure must be increased via an increase in US intensity. However, at a given US intensity, too high static pressure prevents bubble formations, cavitation, and then sludge ultrasonic disintegration.

3.4. Ultrasonic power and density

Gutierrez and Henglein (1990) cited by Thompson and Doraiswamy [11] indicated that the reaction rate increased to a maximum following the increase in P_{US}, and then decreased with further increase in P_{US}. Ratoarinoro *et al.* (1995a) and Contamine *et al.* (1994) cited by Thompson and Doraiswamy [11] explained that at high P_{US}, the formation of a dense cloud of cavitation bubbles around the probe acts to block the energy transmitted from emitter to the bunk solution. The optimum P_{US} also depends on US frequency: different optimum values of P_{US} were found at different US frequencies when investigating the corrosion rate of 304L stainless steel; no optimum value was observed 20 kHz of US frequency [24].

Comparing to untreated sludge, after 60 min of US, DD_{COD} reached 56%, 64% and 80% corresponding to 50, 100 and 200W of sonication, respectively. It was clear that the solubilisation of organics increased owing to the elevated applied P_{US} [3].

According to Kidak *et al.* [3], "*high P_{US} - short US duration"* would be effective in *non-homogeneous (heterogeneous) sludge* like municipal sludge while "*low P_{US} and long US duration"* works in *homogenous sludge* like industrial sludge. The reason could be attributed to the fact that particles in municipal sludge (the fibrous particles coming from toilet papers) were resistant to US disruption; thus P_{US} should be increased to break these particles. Whereas the settled bacteria - the major components in industrial sludge - were broken to soluble materials even at low P_{US}; more solubilisation consequently could be obtained after increasing the retention time.

The results from *the transmutative power function model* indicated that "*low P_{US} - long US duration*" was more efficient than "*high P_{US} - short US duration*" [18]. However, other researches showed a reverse result: the latter model was more efficient than the former [6, 15, 17, 26]. SCOD in the supernatant increased owing to the increase in D_{US} [6, 27]. At ES of 40 kWh/kg_{DS}, an increase in SCOD was 1.2, 1.4, and 1.9-fold corresponding to D_{US} of 0.18, 0.33, and 0.52 W/mL, respectively [6]. With an increase in D_{US} from 0.11 to 0.33 W/mL (120 min sonication), the total solubilised COD (SCOD/TCOD) was 2% and 20% in the supernatant, respectively [16]. Another research showed that at the same level of ES, SCOD increased by 1.2, 2.3 and 4.8-fold at 2, 3, and 4 W/mL, respectively [27].

It is clear that P_{US} and D_{US} are important parameters in WAS disintegration which need considering in terms of cost-benefit purpose in full-scale application and the practical results are supposed to be inclusive of P_{US} and D_{US} at all times.

3.5. Ultrasonic intensity

In general, an increase in intensity *(I)* will increase the sonochemical effects. Since I § P_A^2, the maximum pressures and temperatures within a transient collapse will increase $(P_m \sim P_h + P_A)$. However, it must be noted that intensity cannot be increased indefinitely. With an increase in pressure amplitude (P_A), the bubble may grow so large on rarefaction (R_{max}) that the time available for collapse is insufficient.

Apart from bubbles formation, *bubbles behavior* is also associated with US intensity (I_{US}). As discussed, the disruptive effect of transient bubbles in a short US duration is more noticeable than that of stable bubbles with long US duration. Thus, I_{US} may be considered as a predominant parameter than US duration in terms of bubble behavior, complying with Gronroos *et al*. [6].

Quarmby *et al*. (1999) cited by Pilli *et al*. [4] indicated that the higher mechanical shear forces produced at higher intensities ruptured microorganism cell walls, leading to the increase in SCOD. The DD_{COD} was more than double by increasing the intensity from 6 to 18 W/cm^2 [10].

It is proposed that the ultrasonic process can be optimized by increasing I_{US} (for the higher disruption capability within the shortest possible US duration) to minimize energy use [6].

3.6. Specific energy input

Different researches showed different results of the optimum ES: (i) at 12000 kJ/kg$_{DS}$ (with 34.4 g/kg dry solids), a maximum increment in DD_{COD} was 32% [10]; (ii). 35000 kJ/kg$_{TS}$ (with 3 % TS content) was the optimum ES for the highest SCOD release (Khanal *et al*. (2006) cited by Pilli *et al*. [4]); (iii) ES of 50000 kJ/kg$_{TS}$ was the optimum for sludge disintegration, higher value (>50 kJ/g$_{TS}$) maybe slow down the increment rate of protein, polysaccharides and DNA [28], etc.

On the contrary, with ES from below 1000 to 26000 kJ/kg$_{TS}$, the increase in SCOD (I_{SCOD}) and the soluble COD ratio (SCOD/TCOD) was from 120 % and 4 % to 1233 % and 26 %, respectively. This indicated a positive correlation ($R = 0.993$, $P < 0.01$) between SCOD and ES, but the optimum value for complete disintegration was not found (because the SCOD kept on going up with increase in ES, even at 26000 kJ/kg$_{TS}$ [8]), in agreement with Le *et al*. [15] who investigated the ES range of 0-75000 kJ/kg$_{TS}$.

3.7. Ultrasonic duration

The ratio of sludge solubilisation is defined as the concentration of the organic substances (protein, carbohydrates and COD) in the supernatant after pretreatment to the total organic substances before pretreatment multiplied by 100 [29], which is different from the definition of Bougrier *et al.* [30].

Another approach was the increase in solubilisation (I_x), calculated by the difference between the sonicated X and the initial one (X_0): $\mathbf{I_x (\%) = [(X - X_0) / X_0] \times 100}$. (X could be TDS, $NH4^+$–N, $NO3^-$–N, the protein, and polysaccharide content of the supernatant) [8].

It was proved that the solubilisation of WAS increased gradually with the increase in US duration [5, 6, 29]. Shimizu *et al.* (1993) cited by Pilli *et al.* [4] showed that to get 50 % and 75–80 % increase in solubilisation, it required at least 30–40 min and 90 min of sonication, respectively. The release of SCOD had a linear correlation with US duration [6, 15, 27]. Apart from SCOD, protein and carbohydrate in the sludge/supernatant had the similar trend because US broke down flocs, ruptured bacteria cell walls and then released extracellular organic compounds inside the bacterial flocs [29].

In addition, the VS reduction in AD digester increased gradually following the increase in US duration. VS reduction were 21.5 %, 27.3 % and 33.7 % in the control sample, 30min-sonicated sample, and 150 min-sonicated sample (P_{US} = 45 W), respectively, corresponding to an increment of 27 % and 56.7 % compared to the control after 30 and 150 min of sonication, respectively. Besides, the biogas production in the sonicated sludge had a similar trend. Moreover, the methane percentage in the biogas increased simultaneously corresponding to an increment of 9.7 % compared to the control after 150 min of sonication [9].

3.8. Sludge type and volume of sludge

The SCOD was higher in the secondary sludge: increased 4 and 7.7-fold in the primary and secondary sludge, respectively [27].

The comparison of two different volumes of municipal sludge showed that higher sludge volume resulted in a decrease in sludge disintegration due to the difficulties in creating homogeneous agitation produced by both magnetic stirring and the sonication waves. In other words, with high volume of sludge, the solution could not be as effectively-mixed as with a smaller one; thus, the particles could not move to the tip of the US probe considered as the most active sonication zone, the degree of sludge disintegration consequently decreased [3].

3.9. Total solids concentration of sludge

Considering different sludge concentrations represented by SS (3.57, 7.13, and 14.26 g/L), Li *et al.* [18] indicated that the sludge of low concentration was easier to be disintegrated because its particles can utilize more US energy.

However, other researches showed that higher solids in the liquid made more cavitation sites and more hydro-mechanical shear forces. In case beyond the optimum concentration, absorption effects (or the attenuation effect) disrupted the homogeneous distribution of the acoustic waves [4, 15, 27, 31]: SCOD increased from 1000 mg/L to 1800, 4000, 5800, and 3200 mg/L with the TS content of 0.98, 1.7, 2.6, and 3.6 % w/v, respectively [31].

According to Kidak *et al.* [3], DD_{COD} hiked up with the increase in TS from 4, 8 to 12 g/L. This could be explained that the more solids were around the US probe, the more they were disintegrated due to an active zone formed just around the probe tip. Moreover, the particles could find others surrounding more easily, which also favored the possibility of the particle crush effect. However, there was an opposite trend, the severe decrease in the DD_{COD}, when TS was beyond a certain point because US waves could not be evenly propagated into the medium: DD_{COD} for TS of 24 g/L-solution was lower than of 12 g/L.

The ultrasound disruption index D (a relationship between the disruption efficiency and the solids content) was proposed to evaluate the optimal solid content range:

$$\mathbf{D = \delta\ (S/E)} \quad [6]$$

where *D* is the US disruption index, *S* (mg/L) is the SCOD released in the supernatant by disruption, *E* (kWh/kg_{DS}) is specific energy to sonicate 1 kg dry solids of the sludge, *S/E* is the slope of the trend line for SCOD versus specific energy, and δ is the correlation coefficient relative to US density, which is regarded as 1. With the constant energy input, the optimum range of TS lies between 2.3 % and 3.2 % [6].

In addition, the more dry solids (DS) of sludge, the more DD_{COD} increased because higher concentration of microbes could be disrupted. However, the optimum DS would change and depended on many factors such as reactor configuration (reactor size, transducer type), sludge viscosity, temperature, and polymer concentration (if any – in flocculation) [17].

Five synthetic sludge samples corresponding to 12, 24, 28, 32, and 36 g/L of TS were investigated by Le *et al.* [15]. In agreement with other researchers [3, 4, 6, 27, 31, 32], an optimum value of TS for efficient sludge disintegration by US was observed, 28 g/L. This could be explained by opposite effects. The increase in TS provides more cells and aggregates to be in contact with cavitation bubbles; thereby, the P_{US}, which is required to generate cavitation, is more efficiently consumed. Nevertheless, at higher sludge loading, the acoustic pressure field will decrease faster from the emitter due to the degraded propagation of US waves in a denser suspension. Consequently, acoustic cavitation intensity will be reduced. These two opposite effects lead to an optimum TS concentration which could slightly depend on sludge characteristics, reactor design, and P_{US}. In other words, according to Show *et al.* [6], sufficient quantity of liquid for vaporization and then formation of micro-bubbles is needed in a liquid–solid system. In case of too high TS content, liquid vaporization may be prevented, subsequently affects cavitation bubble formations. Moreover, due to a lack of liquid channels, the generated bubbles may not be well propagated and the cavitation only occurs around the US probe, leading to poor efficiency of sludge disintegration and US probe erosion.

3.10. pH of sludge

According to Wang *et al*. (2005) cited by Pilli *et al.* [4], the effects of sonication parameters and sludge properties on solubilisation of the chemical oxygen demand (*COD*) can be rated as follows: sludge pH > sludge concentration > ultrasonic intensity > ultrasonic density. This suggests that pH adjustment to a suitable value prior to *US* pretreatment is an important step.

3.10.1. Chemical/Alkaline – US pretreatment

Chu *et al*. [16] showed that EPS and gels surrounding cells limit the efficiency of US treatment on sludge disintegration. Adjustment pH of sludge to alkali medium, known as *chemical/alkaline disintegration of sludge*, promotes the EPS hydrolysis and gel solubilisation. After that, cell walls cannot maintain an appropriate turgor pressure [33] and easily disrupts. Therefore, the combination of chemical and ultrasonic treatments, which are based on different mechanisms of sludge disintegration (modification of structural properties for the first, intense mechanical shear force for the second), is expected to take advantage of both and achieve a better efficiency of sludge pretreatment.

The chemicals used for increasing the pH of sludge also affect WAS solubilisation and their efficacy is as follows: NaOH > KOH > $Mg(OH)_2$ and $Ca(OH)_2$ [33, 34]. Ca^{2+} and Mg^{2+} are key substances connecting cells with extra-cellular polymeric substances (EPS). As a result, their presence may enhance the reflocculation of dissolved organic polymers [33], which leads to a decrease in soluble *COD*. In the other hand, overconcentration of Na^+ (or K^+) was reported to cause subsequent inhibition of *AD* [1].

Chiu et al. (1997) cited by Bunrith [35] investigated the hydrolysis rate of chemical, ultrasonic, chemical-ultrasonic, and simultaneous ultrasonic and chemical pretreatment on WAS (1% of TS at ambient temperature). Three set of experiments were designed and conducted; (i) pretreated with 40 meq/L NaOH for 24 h, (ii) pretreated with 40 meq/L NaOH for 24 h followed by US for 24 sec/mL, and (iii) simultaneous ultrasonic (14.4 sec/mL) and chemical (40 meq/L NaOH) pretreatment. The authors indicated the initial hydrolysis rate of the third approach was the highest. Moreover, this approach could shorten the WAS pretreatment time and resulted in a prolific production of SCOD. The second approach was more effective in SCOD and soluble organic nitrogen compared to the first approach but to be closed to the third one.

Bunrith [35] also compared effects of different pretreatment methods (chemical, US, and chemical-US pretreatment) on WAS disintegration and subsequent AD (10, 15, and 25 days of sludge retention time SRT). Chemical-US pretreatment released more SCOD at high chemical dose and high ES. However, it was found that operating with 10 mg/g_{TS} chemical dose at ES of 3.8 kJ/g TS was effective on sludge disintegration. Chemical-ultrasonic was the most effective technique on sludge disintegration. %SCOD *(*$SCOD_{treated}$*/TCOD*100 %)* obtained from chemical-US, chemical, and ultrasonic pretreatment were by 18, 13.5, and 13 %, respectively, indicating the combination effects of hydro-mechanical shear force and OH- radical reaction. According to kinetic study results, the Rate Constant of The Hydrolysis Step of chemical-ultrasonicated sludge was higher than that of ultrasonicated and non-pretreated sludge, proving that chemical-US pretreatment made more organic mass available for biological digestion. Hence, the degradation rate of this pretreated sludge was faster than others, which eventually reduce the digester volume for same digestion efficiency.

Jin *et al.* [33] investigated the effects of combined alkaline and ultrasonic pretreatment of sludge on AD. SCOD values for combined method were higher than those for sole ultrasonic and sole alkaline pretreatment. SCOD levels in different options of combined NaOH and US pretreatments were in descending order as follows: simultaneous pretreatment > NaOH-US pretreatment > US-NaOH pretreatment. Low NaOH dosage (100 g/kg_{DS}), short duration of NaOH treatment (30 min), and low ES (7500 kJ/kg_{DS}) were suitable for sludge disintegration. In the subsequent AD, the degradation efficiency of organic matter was increased from 38.0 % to 50.7 %, which was much higher than that with ultrasonic (42.5 %) and with NaOH pretreatment (43.5 %) at the same SRT.

Kim *et al.* [2] investigated the effects of combined (alkaline + ultrasonic) pretreatment on sewage sludge disintegration. At first, the individual effect of alkaline (pH 8-13) and ultrasonic (3750–45000 kJ/kg_{TS}) pretreatments on sludge disintegration were separately tested. The effect of combined method (where US pretreatment was applied to the alkali-pretreated sludge) was then investigated at different I_{US} by Response Surface Methodology. It was found that the solubilisation (SCOD/TCOD) increase was limited (50 %) in individual pretreatments; however, it reached 70% in combined method, indicating that high pH levels of sludge played a critical role in enhancing the subsequent US pretreatment efficiency. Besides, DD_{COD} proportionally increased following the increase in pH (from 8 to 13), but decreased gradually when ES values were more than 20000 kJ/kg_{TS}. Finally, in order to assess the effect of combined pretreatment on AD, a pretreated sludge (pH 9 + ES of 7500 kJ/kg_{TS}) was fed to a 3 L of anaerobic sequencing batch reactor after 70 days of control operation. CH_4 production yield significantly increased from 81.9 ± 4.5 $mLCH_4/gCOD_{added}$ to 127.3 ± 5.0 $mLCH_4/gCOD_{added}$ by pretreatment. However, about 20% higher soluble N concentration found in the reactor after AD would be an additional burden in the subsequent nitrogen removal system.

3.10.2. Acidic – US pretreatment

Acidic pretreatment is a rare chemical pretreatment method and is applied by the addition of acid to lower the pH of sludge. Acidic pretreatment was thought to accelerate the hydrolysis step by breaking up the cell walls and mineralization of microbial cells, to improve dewaterability, and to improve the overall performance of subsequent AD.

According to Neyens *et al.* [36], the net negative charges on the surface of sludge particles kept them apart. When the pH was decreased down to 2.6 – 3.6, the negative charge on the surface became neutral and at that point, the repulsive force between particles decreased down to minimum and physical stability (such as easy dewatering and flocculation) could be observed.

Sludge cells were proved to be disintegrated and dissolved by acidic treatment at ambient or low temperatures. Only the acid dose significantly affected the solubilisation of sludge [37], and the optimal pH values for the reduction of volatile suspended solids and of excess sludge varied between 1.5 [37], and 3 [36].

Apul [38] indicated that acidic pretreatment (pH 1.5, 2.5, and 4.5 with 20 min of contact time) had a very low performance compared to US pretreatment for enhancing the solubility of sludge. Primary requirement of a pretreatment method is the effectiveness of solubilisation prior to digestion; however, acidic pretreatment was not capable of dissolving organic matters effectively. Combining acidic and mild-sonication pretreatment (acidic-US pretreatment) was expected to disturb the floc structures and to release organic matters into liquid phase and consequently, decrease the overall consumption of energy and chemical. Additionally, the physical characteristics (such as dewaterability and turbidity) of this pretreated sludge were expected to be much better than those of sole US pretreatment. However, the lower the pH value, the worse the solubilisation was due to the antagonistic effect of acid on US pretreatment. Briefly, the efficacy (in terms of solubilisation of organics) of combination of these both methods was better than that of sole acidic pretreatment but worse than that of sole mild-US pretreatment.

4. CONCLUSIONS

Ultrasonication is known as an advanced technology which has been studying to develop and widely apply in sludge pretreatment thanks to the following remarkable properties: mass reduction, odor removal, pathogen decrease, less energy use, and energy recovery in form of methane. It was proved that sludge disintegration is mainly caused by hydro-mechanical shear forces created in cavitation. However, there are many factors affecting the efficacy of cavitation and consequently ultrasonic pretreatment of sludge.

This work aimed at selection, assessment, and indication of important parameters influencing these mentioned processes. Optimization methodology of related parameters, the differences of optimum values of recent researches as well as the similarities of effecting trends of these parameters on cavitation and sludge pretreatment efficiency were specifically pointed out, including ambient conditions (temperature, external pressure), ultrasonic properties (frequency, power input, US density, US intensity, specific energy input, US duration), and sludge characteristics (sludge type, volume of sludge, TS concentration, pH). The research is a prerequisite for the determination of related important parameters to optimize sludge ultrasonic pretreatment efficiency in lab-scale and practical application.

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TÓM TẮT

TỔNG QUAN VỀ TỐI ƯU HÓA HIỆU QUẢ TIỀN XỬ LÍ BÙN THẢI BẰNG CÔNG NGHỆ SIÊU ÂM

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Siêu âm (Ultrasonication) là một công nghệ tiên tiến trong lĩnh vực tiền xử lí bùn thải, chủ yếu nhờ vào lực cắt thủy cơ (hydro-mechanical shear forces) từ quá trình tạo bọt (cavitation). Có nhiều yếu tố ảnh hưởng đến hiệu quả cavitation, theo đó là hiệu quả phân rã bùn thải. Bài báo nhằm mục đích lựa chọn, đánh giá và xác định các thông số quan trọng có ảnh hưởng đến các quá trình nêu trên. Phương pháp tối ưu hóa các thông số liên quan, sự khác biệt giữa các giá trị tối ưu cũng như những điểm tương đồng về xu hướng tác động đến hiệu quả cavitation và tiền xử lí bùn thải sẽ được phân tích cụ thể, bao gồm các điều kiện môi trường (nhiệt độ, áp suất), tính chất siêu âm (tần số, công suất, mật độ, cường độ, năng lượng riêng, thời gian) và đặc tính bùn thải (loại bùn thải, thể tích, nồng độ chất rắn, độ pH). Nghiên cứu này là tiền đề cho việc tối ưu hóa hiệu quả tiền xử lí bùn thải bằng công nghệ siêu âm - quy mô phòng thí nghiệm hoặc ứng dụng thực tế.

Từ khóa: cavitation, tiền xử lí kết hợp, tối ưu hóa quá trình, tiền xử lí siêu âm, bùn thải hoạt tính.