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# Optimization of hydrostatic pressure at varied sonication conditions – power density, intensity, very low frequency – for isothermal ultrasonic sludge treatment

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## A B S T R A C T

This work aims at investigating for the first time the key sonication (US) parameters: power density ( $D_{US}$ ), intensity ( $I_{US}$ ), and frequency ( $F_s$ ) – down to audible range, under varied hydrostatic pressure ( $P_h$ ) and low temperature isothermal conditions (to avoid any thermal effect).

The selected application was activated sludge disintegration, a major industrial US process. For a rational approach all comparisons were made at same specific energy input ( $ES$ , US energy per solid weight) which is also the relevant economic criterion.

The decoupling of power density and intensity was obtained by either changing the sludge volume or most often by changing probe diameter, all other characteristics being unchanged. Comprehensive results were obtained by varying the hydrostatic pressure at given power density and intensity. In all cases marked maxima of sludge disintegration appeared at optimum pressures, which values increased at increasing power intensity and density. Such optimum was expected due to opposite effects of increasing hydrostatic pressure: higher cavitation threshold then smaller and fewer bubbles, but higher temperature and pressure at the end of collapse.

In addition the first attempt to lower US frequency down to audible range was very successful: at any operation condition ( $D_{US}$ ,  $I_{US}$ ,  $P_h$ , sludge concentration and type) higher sludge disintegration was obtained at 12 kHz than at 20 kHz. The same values of optimum pressure were observed at 12 and 20 kHz.

At same energy consumption the best conditions – obtained at 12 kHz, maximum power density 720 W/L and 3.25 bar – provided about 100% improvement with respect to usual conditions (1 bar, 20 kHz). Important energy savings and equipment size reduction may then be expected.

### Keywords:

Audible frequency  
Hydrostatic pressure  
Power density  
Power intensity  
Specific energy

## 1. Introduction

### 1.1. Sonochemical engineering issues

Power ultrasound (US) is well known for its outstanding activation of various chemical and physical processes [1]. It has been clearly proved that transient cavitation is the main cause of ultrasound efficiency, either by producing active radicals (chemical activation due to hot spots at bubble cavitation collapse) or very high shear stresses and shock waves due to pressure spots (physical or

mechanical effects). The latter effects are especially efficient to clean, erode and activate solid surfaces or to finely disperse multi-phase media.

Due to extremely complex and coupled phenomena – especially highly nonlinear behavior of cavitation bubbles, inhomogeneity of bubble sizes and locations, inhomogeneity of acoustic field – such ultrasonically assisted processes are far from being conveniently modeled despite tremendous progress in single bubble dynamics and sonoluminescence [2,3].

The development of sonochemical processes mainly depends on two major issues: the convenient knowledge of all relevant parameters at small scale and the rational scaling-up [4]. Concerning the first aspect, the number of possible effective parameters is rather large and still discussed.

There may be divided in 4 groups:

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- Acoustic parameters, i.e. power density (W/L), power intensity (W/cm<sup>2</sup>), frequency, sonication time or better specific US energy (kJ/kg).
- Geometrical parameters, including geometry and relative size of reactor, stirrer and emitter, presence of internals, location of the emitter, liquid filling.
- Standard operation parameters (temperature and pressure).
- Investigated system properties, i.e. physicochemical properties of liquid(s), solid if any, and gas headspace.

Of course, among thousands of works on ultrasound activation there is not a single one, for any US application, having achieved such a large amount of experimental work; most of them being devoted to show some US effect on a selected application.

Concerning ultrasonic parameters, frequency effects have been widely investigated showing a clear difference between high frequencies (>100 kHz), to be selected for radical chemistry, and low frequencies (<100 kHz), much more efficient for mechanical and physical effects [5,6].

Despite a clear advantage of lowering the frequency to improve mechanical effects, extremely few works have tested audible frequencies [7], probably due to a lack of equipment connected to apprehension of noisy conditions.

Ultrasound power is probably the most important parameter, but it should be considered from two sides: the power density or US dose (W/L), always involved in scaling-up studies, and the power intensity (W/cm<sup>2</sup>), better connected to the cavitation threshold. It should be noticed that almost all experimental works on power effects are performed in same equipment where increasing the US power will proportionally increase both the density and the intensity. Consequently, most often, the separate role of these two power characteristics has been ignored. Very few works have indeed investigated density and intensity separately [8–10], by changing either the emitter and/or the reactor geometry. Moreover note that such changes might result in additional effects to that expected and a possible misinterpretation of its individual role. Generally speaking, sonication performances are improved when increasing US power, nevertheless in some works an optimal power has been pointed out [8,11].

Concerning operation conditions, such as temperature, hydrostatic pressure and nature of the gas (either bubbled or lying above the liquid surface), it is well known that they may significantly affect transient cavitation intensity as deduced from single bubble dynamics.

Increasing temperature has a negative effect on cavitation intensity, leading to a severe dampening of the collapse as soon as liquid vapor pressure becomes significant in the cavitation bubbles. Therefore an optimum temperature is generally expected for US-assisted applications, whose value depends on how temperature affects the silent process: for instance, a higher sonication temperature should be applied for extraction or dissolution with respect to applications involving only mechanical effects, like solid disruption, nanoparticle production or emulsification, where room temperature is most often convenient.

Gas content has mostly been investigated for radical production and especially in single bubble dynamics and sonoluminescence, showing best effects with monoatomic gases due to the higher maximum temperature at the end of the quasi adiabatic collapse.

US mechanical effects have been scarcely investigated with regard to many works on reactions enhanced by US radical generation, but an improvement under pressure is usually reported [12–15], and in the most detailed studies an optimum value has been found [1,12,14]. For the specific effect of applied pressure

on sonochemical reactions or sonoluminescence, one can refer to the article of Geng and Thagard [16].

It should be emphasized that all parametric studies are very fragmentary, either due to single parameter exploration or to changes in equipment geometry when changing frequency.

## 1.2. Ultrasonic sludge pretreatment

Sludge processes for wastewater treatment produce large quantity of solid waste, commonly treated by anaerobic digestion. However, this method requires a pretreatment process due to a rate limiting step of hydrolysis. Ultrasonic irradiation is a promising and feasible mechanical disruption technique – mainly based on cavitation phenomenon – for sludge disintegration and microorganism lyses. Excellent reviews have been published on this major application of power ultrasound by Show et al. [17], Carrere et al. [18], Pilli et al. [19] and recently Tyagi et al. [20]. Apart from sludge characteristics and reactor geometry, US parameters (frequency –  $F_s$ , intensity –  $I_{US}$ , density –  $D_{US}$ , etc.) and external conditions (hydrostatic pressure –  $P_h$ , temperature, etc.) should play some roles in the pretreatment efficiency. However, as for other applications, there is lack of researches on the integrated effect of some key US parameters and external conditions on sludge pretreatment.

Increasing US frequency in the 25–1100 kHz range has been reported to lower the degree of sludge disintegration [19,21,22]. Lowering  $F_s$  below 20 kHz could then be interesting and needs specific investigation.

Most often unit power (power per unit volume or power density) and intensity (power per unit surface of emitter) have been varied simultaneously and proportionally by changing US power in the same equipment [17,23–26]. As expected, higher mechanical shear forces produced at higher  $I_{US}$  rupture microorganism cell walls, leading to an increased solubilization of organic matter. For instance, Neis et al. [27] found that the degree of sludge disintegration more than doubled by increasing  $I_{US}$  from 6 to 18 W/cm<sup>2</sup>.

Note that generally acousticians refer to intensity (or acoustic pressure) while process engineers prefer unit power for scaling-up purposes.

A preliminary study [28], more sludge oriented, was recently published comparing several types of sludge under both isothermal and adiabatic sonication, and for the first time varying hydrostatic pressure (1–16 bar) but at same input power (150 W), same frequency (20 kHz) and same probe diameter (35 mm). All comparisons were made at equal specific energy input  $ES$ , i.e. energy per solid weight, which is the only rational way for parametric investigations. It was first verified that, at same specific energy, high power – then short sonication time – was more efficient. An optimum value of hydrostatic pressure was observed at about 2 bar with significant improvement as compared to atmospheric pressure.

The present work aims at providing an extensive parameter investigation including three main aspects: (i) the separate roles of power density and power intensity, (ii) the effect of very low frequency, down to the audible range (12 kHz) as compared to 20 kHz the usual low frequency, (iii) the role of hydrostatic pressure under the complete range of ultrasonic parameters (power density, intensity and frequency).

Such unprecedented parameter exploration in the same equipment concerns the special application of sludge solubilization (or disintegration) which represents the largest scale of US-assisted industrial processes. In isothermal low temperature conditions, transient cavitation being the only cause of sludge disintegration, the main trends of this work are likely to be found in other cavitation controlled applications.

## 2. Materials and methods

Most of the useful information was given in Le et al. [29]. This section mainly details new improved ultrasound equipment: two frequencies, two probe sizes at each frequency, larger US power range, which have been used here on a single type of activated sludge.

### 2.1. Sludge samples

Waste activated sludge (WAS) was collected from Ginestous wastewater treatment plants (Toulouse, France). Note that sludge sampling was performed at different periods in relation with the changes in US equipment along this work. Synthetic sludge sample, whose properties are given in Table 1a, was used for investigating  $P_{US}$  and  $I_{US}$  effects. The effect of  $F_S$  was then looked into using the synthetic WAS characterized in Table 1b.

Sludge was stored in a freezer, which might change some physical characteristics of the sludge, but should not significantly affect COD solubilization results [30]. In the present study, less than 8% difference in sludge disintegration results have been observed between fresh sludge (without freezing) and frozen sludge for the  $ES$  range of 7000–50,000 kJ/kg<sub>TS</sub>. The sludge was defrosted and diluted with distilled water before experiments to make synthetic sludge samples with 28 g/L of total solid (TS), the optimum concentration for sludge US disintegration [28].

### 2.2. Ultrasound application

Ultrasonic irradiation was emitted by a cup-horn ultrasound unit included in an autoclave reactor which was connected to a pressurized N<sub>2</sub> bottle (see Fig. 1).

The reactor and its internals were made of 316 L stainless steel. The reactor had an internal diameter of 9 cm and a depth of 18 cm, for a maximum capacity of 1 L. A cooling water stream (15 °C) was continuously circulated in an internal coil to control temperature (T) of the solution at 28 ± 2 °C during sonication. The solution was stirred by a Rushton type turbine (32 mm diameter) at 500 rpm according to our previous work [28]. A constant volume (V) of synthetic sludge sample (0.5 L) was used for all experiments, excepting that exploring intensity variations at constant density through proportional changes in power and volume (cf. Fig. 5).

The US equipment was especially built up by Sinaptec for this work in order to access to audible frequency. It included two generators working at 12 and 20 kHz, and for each two associated probes of 13 and 35 mm diameter, labeled as *SP* and *BP*, respectively. The probes being mounted flush at the bottom of this cup-horn type reactor, the exact sonication area was exactly known, better than with a dipping horn, allowing accurate analysis of

intensity effects. The maximum  $P_{US}$  (transferred from the generator to the transducer) was 100 W and 400 W for *SP* and *BP*, respectively. The 20 kHz device was composed of four elements: a piezoelectric transducer, a titanium booster, an aluminum flange ensuring a good mechanical connection, and the cup-horn emitter. There was no booster for the 12 kHz device. During operation, the transducer was cooled by compressed air.

Due to the technical limitations of the US systems,  $I_{US}$  ranged between 5.2 and 75 W/cm<sup>2</sup> (Table 2). It is probably well above the cavitation threshold for WAS at atmospheric pressure (according to the range for water with many impurities) even though it was suggested to be at about 20 W/cm<sup>2</sup> by Zhang et al. [22]. Note that the maximum power ratio of 360/50 between *BP* and *SP* corresponds to the surface ratio of the probes, allowing comparison at same  $I_{US}$ .

Different sonication durations corresponding to four values of  $ES$  (7000, 12,000, 35,000, and 50,000 kJ/kg<sub>TS</sub>) were tested:

$$ES = (P_{US} * t) / (V * TS)$$

where  $ES$  is the specific energy input or energy per total solid weight (kJ/kg<sub>TS</sub>),  $P_{US}$  is the US power input (W),  $t$  is the sonication duration (s),  $V$  is the sludge volume (L), and  $TS$  is the total solid concentration (g/L).

The equivalent amplitude of acoustic pressure corresponding to each  $P_{US}$  and probe size, calculated from the following equation, is also given in Table 2:

$$P_A = (2 * I_{US} * c * \rho)^{1/2}$$

where  $P_A$  is the amplitude of acoustic pressure (Pa),  $I_{US}$  is the ultrasonic intensity (W/m<sup>2</sup>),  $c$  is the sound speed (m/s), and  $\rho$  is the density of the sludge suspension (kg/m<sup>3</sup>).

The density of the sludge suspension at  $TS = 28$  g/L was measured at 25 °C and found almost equal to that of water: 996.7 kg/m<sup>3</sup>. However, the speed of sound in sludge suspensions was neither measured in this work nor found in others. Values in different suspensions, e.g. kaolin clay slurries [31], clay sediments [32,33], cornstarch [34], glass [35], were therefore examined to find a convenient one. In the weight range of 1–5%, the differences in sound speed may be ignored and the values are almost equal to that in water (1496 m/s at 25 °C) which was consequently used for the calculation.

The conditions listed in Table 2 allowed to investigate the effect of  $P_{US}$  first, then the effect of  $I_{US}$  resulting either from  $P_{US}$  or emitter surface variation at atmospheric pressure. The effect of  $I_{US}$  was also studied by varying sludge volume and  $P_{US}$  proportionally with the same *BP* to keep the same  $D_{US}$ ,  $F_S$  effect (12 and 20 kHz) was then looked into using *BP* in a  $P_{US}$  range of 50–360 W. Finally, hydrostatic pressure was varied (1–6 bar) for each combination of  $P_{US}$ , probe size ( $I_{US}$ ), and  $F_S$  to approach the corresponding optimal pressure. Many experiments were duplicated and the coefficients of variation of sludge disintegration results were about 5%.

### 2.3. Analytical methods

Total and volatile solids contents (TS and VS, respectively) were measured according to the following procedure [36]. TS was determined by drying a well-mixed sample to constant weight at 105 °C. VS was obtained from the weight loss on ignition (550 °C) of the residue.

The degree of sludge disintegration ( $DD_{COD}$ ) was calculated by determining the soluble chemical oxygen demand after strong alkaline disintegration of sludge ( $SCOD_{NaOH}$ ) [28] and the chemical oxygen demand in the filtered (0.2 μm) supernatant before and after treatment ( $SCOD_0$  and  $SCOD$  respectively) using Hach spectrophotometric method [37]:

**Table 1**  
Characteristics of the sludge samples.

Parameter	Value	
	a	b
<i>Raw sludge sample</i>		
pH	6.3	6.3
Total solids (TS)	g/L 31.9	34.2
Volatile solids (VS)	g/L 26.4	30.2
VS/TS	% 82.8	88.3
<i>Synthetic sludge sample</i>		
Total solids (TS)	g/L 28.0	28.0
Mean $SCOD_0$	g/L 2.8	4.1
$SCOD_{NaOH\ 0.5M}$	g/L 22.7	22.1
TCOD	g/L 36.3	39.1
$SCOD_{NaOH}/TCOD$	% 62.5	56.5

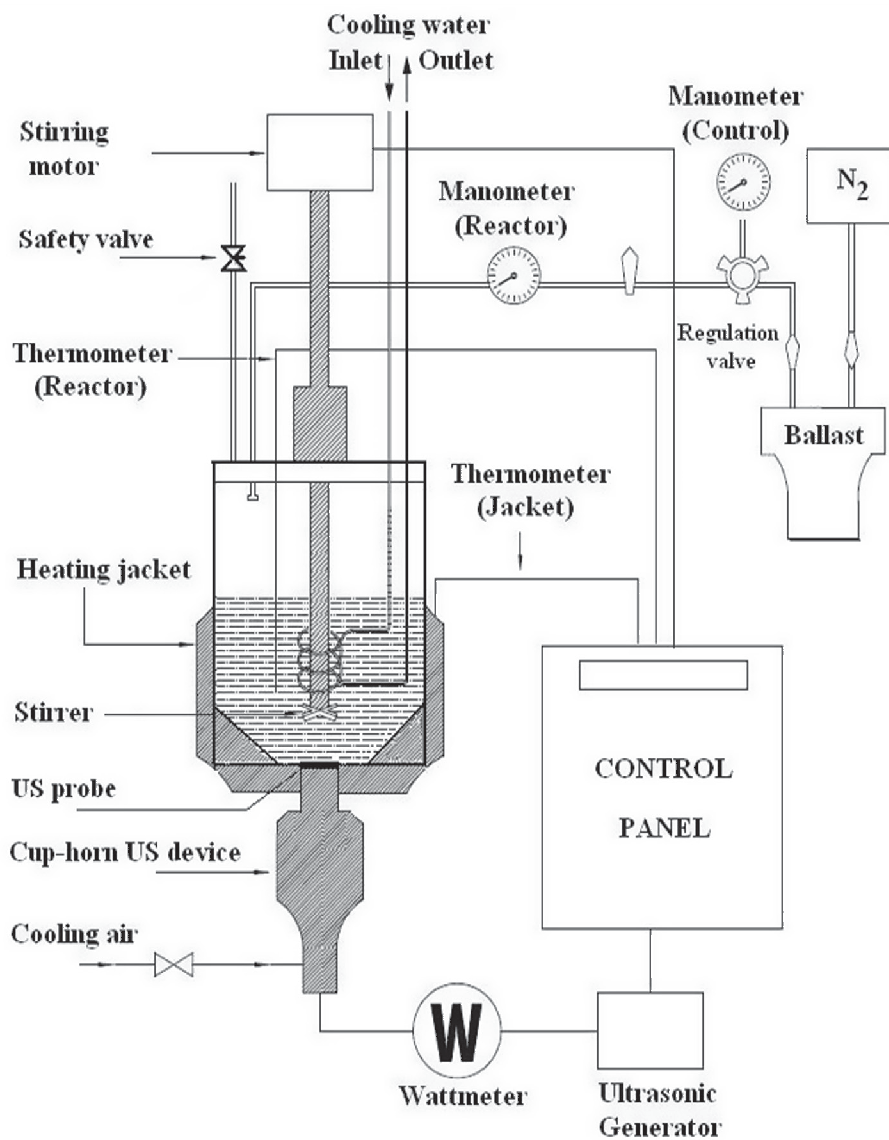


Fig. 1. Ultrasonic autoclave set-up.

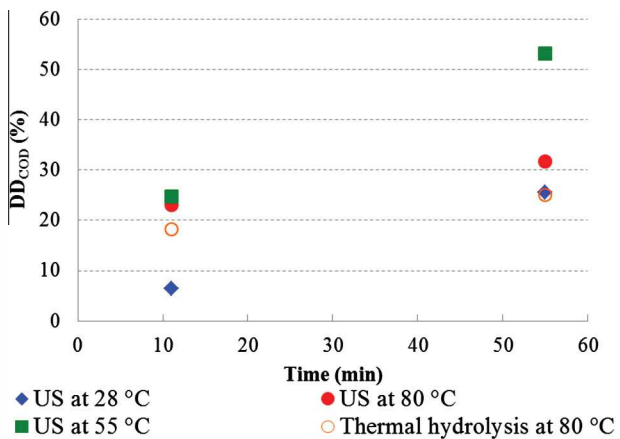


Fig. 2. Effect of temperature on  $DD_{COD}$  by isothermal US (20 kHz, atm. pressure,  $P_{US} = 150$  W, BP, WAS presented in Table 1a) and thermal hydrolysis.

$$DD_{COD} = (SCOD - SCOD_0) / (SCOD_{NaOH} - SCOD_0) * 100(\%)$$

$DD_{COD}$  represents the normalized quantity of organic carbon that has been transferred from the cell content (disruption) and solid materials (solubilization) into the external liquid phase of sludge.

### 3. Results and discussion

#### 3.1. Preliminary experiments: selection of temperature conditions

It is well known that cell lysis and then COD solubilization may occur at moderate temperature without any other activation. It was first verified that only negligible COD changes occurred within 2 h at 28 °C under the selected stirring speed (500 rpm).

US isothermal experiments achieved at three different temperatures clearly showed better solubilization at 55 °C than at 28 °C and 80 °C (Fig. 2). Indeed results of solubilization at 80 °C gave very similar performances with or without US (round symbols in Fig. 2),

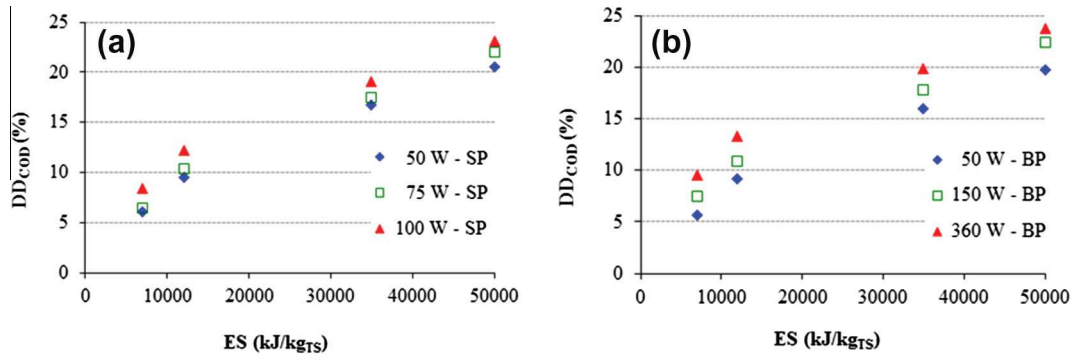


Fig. 3. Effect of ES and  $P_{US}$  on  $DD_{COD}$  (20 kHz, atm. pressure, WAS presented in Table 1a): (a) SP (b) BP.

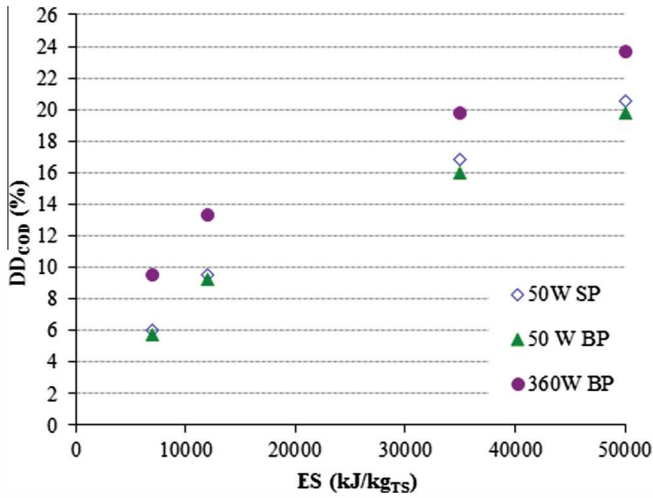


Fig. 4. Comparison of  $I_{US}$  (same  $D_{US} = 100$  W/L) and  $D_{US}$  (same  $I_{US} = 37.5$  W/cm<sup>2</sup>) effects on  $DD_{COD}$  at different ES (20 kHz, atm. pressure, WAS from Table 1a).

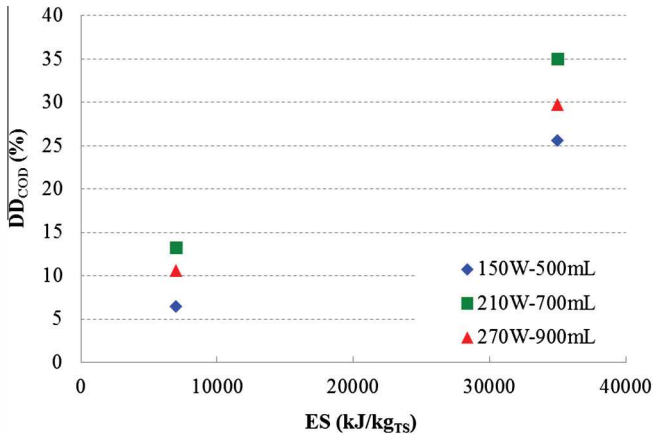


Fig. 5. Effect of  $I_{US}$  (at same  $D_{US} = 300$  W/L by changing  $P_{US}$  and sludge volume proportionally with the same probe) on  $DD_{COD}$  (20 kHz, atm. pressure, WAS from Table 1b, BP).

suggesting that, at such high temperature, cavitation is much less efficient than at lower temperatures (due to high vapor content in collapsing bubbles) and might be ignored as regard to pure thermal effects.

Even though 55 °C is much better for sludge disintegration, a temperature of 28 °C has been selected in this work devoted to parameter effects on US efficiency, in order to avoid any mixed

**Table 2**  
Test parameters and levels.

Combination	$I_{US}$ (W/cm <sup>2</sup> )	$D_{US}$ (W/L)	Equivalent acoustic pressure (bar)
SP 50 W	37.5	100	10.6
SP 75 W	56.3	150	13.0
SP 100 W	75.	200	15.0
BP 50 W	5.2	100	3.9
BP 150 W	15.6	300	6.8
BP 360 W	37.5	720	10.6

thermal and US effects. In addition it should be mentioned that preliminary heating up to 55 °C yet produced significant solubilization before starting isothermal sonication, which would result in less accurate data analysis.

### 3.2. Effect of $P_{US}$ on sludge disintegration

The effect of US power  $P_{US}$  on WAS disintegration was studied at 20 kHz for the two probes. As explained before, changing  $P_{US}$  in the same equipment will result in similar changes in density  $D_{US}$  and intensity  $I_{US}$ . Results are given in Fig. 3. The same conclusion was deduced from the experiments conducted on different  $P_{US}$  ranges and with both probe sizes. The higher  $P_{US}$ , the higher  $DD_{COD}$  was achieved at same ES due to the increase in cavitation intensity. Despite higher uncertainty at low  $DD_{COD}$  (then low ES), the main effect was clearly observed at the lowest ES value (7000 kJ/kg<sub>TS</sub>) where  $DD_{COD}$  was improved by 40% and 67% when increasing  $P_{US}$  from 50 to 100 W for SP (Fig. 3a) and from 50 to 360 W for BP (Fig. 3b), respectively.

This limited but always positive effect of  $P_{US}$  on WAS disintegration proves that in the investigated range of  $I_{US}$  (<75 W/cm<sup>2</sup>), there is no significant “saturation effect” due to a bubble cloud formation near the probe and then no severe damping of the US wave, as observed in some previous works [38–40].

In agreement with other researchers [24,30,41–43], the highest  $P_{US}$  – shortest sonication time mode was the most effective protocol for sludge pretreatment in these conditions (low temperature, atmospheric pressure and  $I_{US} < 75$  W/cm<sup>2</sup>).

### 3.3. Effect of $I_{US}$ and $D_{US}$ on sludge disintegration

In order to separately investigate power density and intensity, two probe sizes were used. Comparisons were made at same specific energy.

Effects of  $I_{US}$  on sludge disintegration were investigated at same  $P_{US}$  (50 W) then same  $D_{US}$ , by changing the probe: SP ( $I_{US} = 37.5$  W/cm<sup>2</sup>) vs. BP ( $I_{US} = 5.2$  W/cm<sup>2</sup>). These experiments were conducted at 20 kHz. Results are shown in Fig. 4, along with those of a complementary experiment conducted with the big probe at the same

$I_{US}$  value of 37.5 W/cm<sup>2</sup> as the small one, but at a higher  $D_{US}$  (360 W-BP, 720 W/L), for comparison of both effects.

First, experiments at the same  $P_{US}$  of 50 W showed only very little improvements of  $DD_{COD}$  (less than 5%) when increasing  $I_{US}$  from 5.2 to 37.5 W/cm<sup>2</sup>. A similar but less significant observation could be deduced from Fig. 3 for a higher  $P_{US}$  level: only about 10% of  $DD_{COD}$  improvement was achieved when increasing  $I_{US}$  by approximately 5 times, from 150 W-BP (15.6 W/cm<sup>2</sup>) to 100 W-SP (75 W/cm<sup>2</sup>) combination. Conversely, increasing  $D_{US}$  from 100 to 720 W/L by using the big probe instead of the small one and keeping same  $I_{US}$  (37.5 W/cm<sup>2</sup>) provided much better disintegration (up to 60% at the lowest  $ES$ ).

The apparently poor effect of  $I_{US}$  may be surprising as it is claimed to be a significant parameter in some of the previous works [19,27,44]. It should be recalled that in our experiments  $I_{US}$  was varied by varying probe size in the same reactor, which involves an important modification of the ultrasonic field with a reduced irradiated volume. It could be therefore suggested that the expected gain due to higher cavitation at higher  $I_{US}$  would approximately be balanced by the reduced volume of the cavitation zone. Of course this approximate balance should no longer be expected when  $I_{US}$  is reduced down to the cavitation threshold where  $US$  has no more effects.

Another way of checking this parameter would consist of changing both the reactor volume and  $P_{US}$  proportionally with the same probe. Obviously, changing reactor size would be much more complex to achieve, especially under pressure. Thus additional experiments were carried out via changing  $P_{US}$  and sludge volume proportionally, with the same  $BP$  to keep the same  $D_{US}$  (300 W/L). They are labelled in Fig. 5: 150 W–0.5 L, 210 W–0.7 L, and 270 W–0.9 L. Note that a sludge volume between 0.5 and 0.9 L corresponds to the maximum possible range for the reactor configuration used in this work. Despite a rather restricted intensity range (16–28 W/cm<sup>2</sup>)  $DD_{COD}$  variations were clearly higher than in the previous experiments with same sonicated volume and different probe sizes. As shown in Fig. 5, the best conditions for  $DD_{COD}$  were the intermediate ones which might be intuitively explained by opposite effects of increasing intensity (thus cavitation strength) and increasing volume (thus damping  $US$  wave far from the emitter surface and reducing the active volume fraction). In addition, the stirring speed might be unable to well homogenize the 0.9 L suspension (as it was optimized at 0.5 L). Therefore, with this approach of variable sludge volume, a clear optimum of  $I_{US}$  could be found, *i.e.* 22 W/cm<sup>2</sup>, contrary to probe size variation.

These two approaches of intensity effects leading to very different results confirm the complexity of experimental analysis of ultrasound intensification of any process due to the additional effects of reactor and probe design on the acoustic field, especially in transient cavitation conditions. The clear variation of  $US$  efficiency when changing slightly the sonicated sludge volume might be more significant than the poor effect observed when changing much more the intensity but through probe size.

### 3.4. Effect of frequency on the efficacy of sludge sonication

As mentioned earlier, even though most applications using mechanical effects of  $US$  power are improved when reducing  $F_s$ , nearly no information is available under 20 kHz – the usual limit of commercial equipment corresponding also to the limit of human hearing. Sludge sonication at 12 kHz was investigated for the first time, and assessed through  $DD_{COD}$  and particle size reduction with respect to the standard 20 kHz treatment. According to the availability of the equipment, experiments were successively carried out at 50 and 150 W, using sludge sample given in Table 1a, then at 360 W using  $WAS$  sample given in Table 1b. Results are shown on Fig. 6.

Fig. 6 shows that with the two different sludge samples, the lower the frequency, the more the sludge was disintegrated due to more violent cavitation. As previously found at 20 kHz, more sludge disintegration was achieved at higher  $P_{US}$  and again the largest differences were noticed at low  $ES$ . It is interesting to note that the beneficial effect of lowering frequency is clearly enhanced at increasing power, for example, at the lowest  $ES$  (7000 kJ/kg<sub>TS</sub>) by 21%, 45% and 64% for  $P_{US}$  of 50, 150 and 360 W, respectively. At lower frequencies, shock waves are stronger and mechanical effects are favoured due to the resonance bubble size being inversely proportional to the acoustic frequency [45]. However, at low  $F_s$ , the maximum collapse time and the maximum size of the expanded cavity are increased, thus the optimum cavitation effect should occur at higher  $P_{US}$  [40].

Although it was shown that particle size reduction had much faster dynamics than  $COD$  solubilization [28], it was interesting to compare particle size reduction under 12 and 20 kHz sonication. As shown in Fig. 7, the lower the frequency, the faster the sludge particle size was reduced during the first two minutes after which the differences in size were very small.

### 3.5. Effect of $P_{US}$ , $I_{US}$ , and $F_s$ on the optimum pressure and subsequent $DD_{COD}$

According to our previous research [28], performed at constant power density and intensity (300 W/L and 15.6 W/cm<sup>2</sup>, respectively), the same optimum of pressure was found (about 2 bar) whatever  $ES$  and sludge type. This section presents the dependence of this optimal pressure on  $D_{US}$  and  $I_{US}$  by changing the probe size, as well as on  $F_s$ . As previously, this optimum is related to  $US$  solubilization of organic matter quantified through  $DD_{COD}$ .

#### 3.5.1. Effect of power density and intensity on optimum pressure

Sonication at 20 kHz was applied on secondary sludge (Table 1a) at an  $ES$  value of 50,000 kJ/kg<sub>TS</sub>, varying hydrostatic pressure between 1 and 6 bar with 0.5 bar intervals to look for the optimum. Results are presented in Fig. 8. Note that due to limitation of the apparatus, the  $US$  system could not work at pressures higher than 2 bar for  $SP$  at 50 W.

Fig. 8a dispatches data obtained with  $BP$  at various pressures for three power inputs (50, 150 and 360 W), then three proportional values of density and intensity. A marked optimum appeared in any case. This optimal pressure shifts toward higher values when increasing  $P_{US}$ : 1 bar (or even lower) at 50 W, 2 bar at 150 W, and 3.5 bar at 360 W. In addition the corresponding maximum of  $DD_{COD}$  is also significantly higher at higher power, showing the great advantage of sonication under convenient pressure: 56% improvement at 360 W with respect to atmospheric pressure. Even more spectacular results are shown on Fig. 8b indicating that the optimum pressure and the corresponding maximum of  $DD_{COD}$  are very sensitive to the probe size. The comparison of  $DD_{COD}$  variations at 50 W shows very different behaviors at increasing pressure: continuous decrease with  $BP$  and marked maximum with  $SP$ . It should be noted that at 1.5 bar and 50 W, the efficiency of sludge disintegration is more than double at high intensity ( $SP$  vs.  $BP$ ). It should be recalled that conversely no significant effect of intensity through probe size variation was previously observed at atmospheric pressure (cf. Fig. 4). A last and unexpected information is given by these figure when comparing  $DD_{COD}$ /pressure profiles: in some cases lower power may be more efficient if it is used near the corresponding optimum pressure. For example, on Fig. 8a at 2 bar:  $DD_{COD}$  is about 20% higher at 150 W than at 360 W; on Fig. 8b at 1.5 bar: 50 W is better than 100 W (by about 30%). This is clearly in contradiction to the general rule of *higher power – shorter sonication time*, which



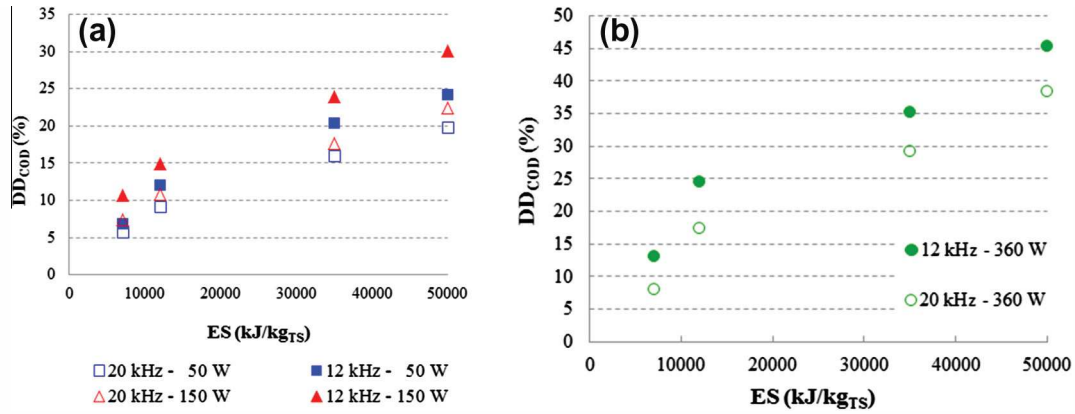


Fig. 6. Effect of ES and sound frequency on sludge disintegration (atm. pressure, BP): (a) WAS given in Table 1a. (b) WAS given in Table 1b.

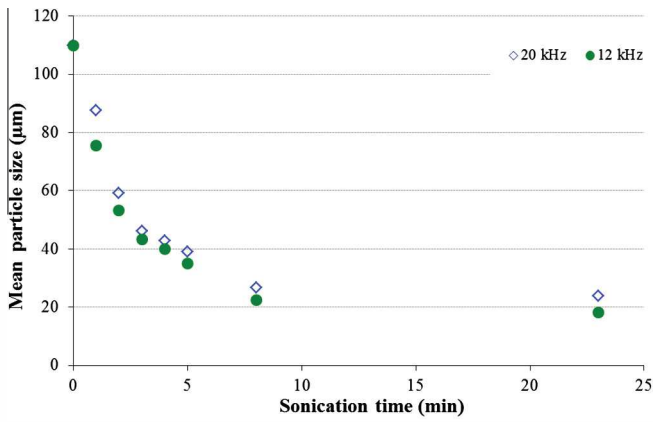


Fig. 7. Mean particle size reduction under US at different  $F_s$  ( $P_{US} = 360$  W, atm. pressure, WAS from Table 1b, BP).

is nevertheless always verified (and even more marked) near optimum pressures.

The existence of an optimum pressure was yet shown in our previous work [28] and may easily be explained from the simplified cavitation bubble collapse model proposed by Neppiras [46]. This model assumes an isothermal bubble growth up to the maximum radius where the bubble is mainly filled by vapor at equilibrium at the ambient temperature  $T_o$ , then a very fast adiabatic collapse leading to a hot spot at  $T_{max}$  and  $P_{max}$  within the bubble:

$$T_{max} = T_o \left\{ \frac{P_m(\gamma - 1)}{P} \right\}$$

$$P_{max} = P \left\{ \frac{P_m(\gamma - 1)}{P} \right\}^{\left( \frac{\gamma}{\gamma - 1} \right)}$$

where  $T_o$  is temperature of the bulk solution,  $\gamma$  is the ratio of specific heats,  $P$  is the pressure in the bubble at its maximum size and usually assumed to be the vapor pressure of the liquid,  $P_m$  is the total solution pressure at the moment of final collapse ( $P_m \sim P_h + P_a$ , with  $P_h$  the hydrostatic pressure and  $P_a$  the acoustic pressure).

Note that calculations of single bubble dynamics often show the hot spot before the maximum acoustic amplitude is reached, then hydrostatic pressure could have even more influence than acoustic pressure. Thereby, increasing hydrostatic pressure leads to an increase of  $P_{max}$  and  $T_{max}$ , i.e. of cavitation intensity. For US mechanical effects only the pressure peak appears relevant.

On the other hand, as abovementioned, increasing hydrostatic pressure also results in an increase in the cavitation threshold, thus the amplitude of acoustic pressure ( $P_A$  depending on  $I_{US}$ ) should be in excess as compared to hydrostatic pressure for cavitation bubbles to be generated: indeed it can be roughly assumed that if  $P_h - P_A > 0$ , there is no resultant negative pressure and no cavitation. In addition it is generally accepted that less and smaller cavitation bubbles are formed when increasing hydrostatic pressure.

These two considerations qualitatively explain the existence of an optimum for hydrostatic pressure which should increase when

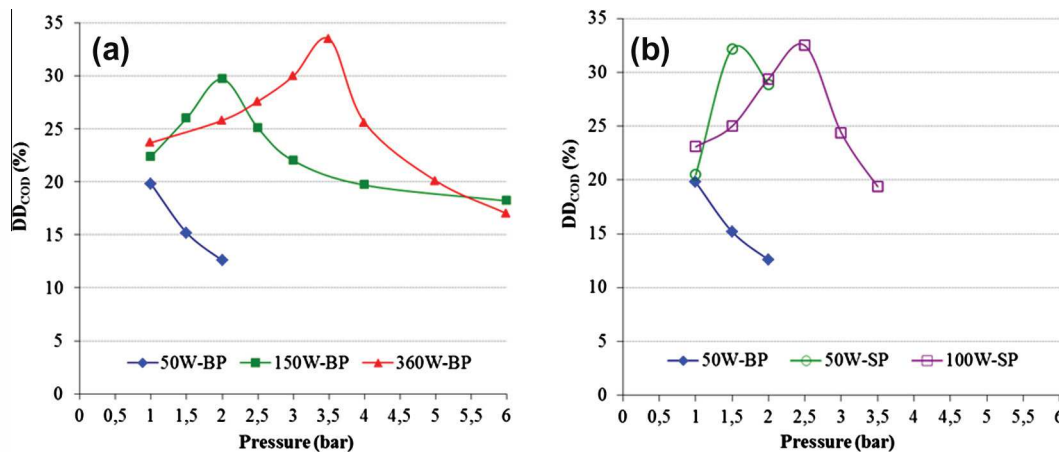


Fig. 8. Effect of hydrostatic pressure on  $DD_{COD}$  of WAS (Table 1a) for different  $P_{US}$  and probe sizes ( $F_s = 20$  kHz,  $ES = 50,000$  kJ/kg<sub>TS</sub>): (a) BP, (b) SP and comparison SP/BP at same  $P_{US}$ .

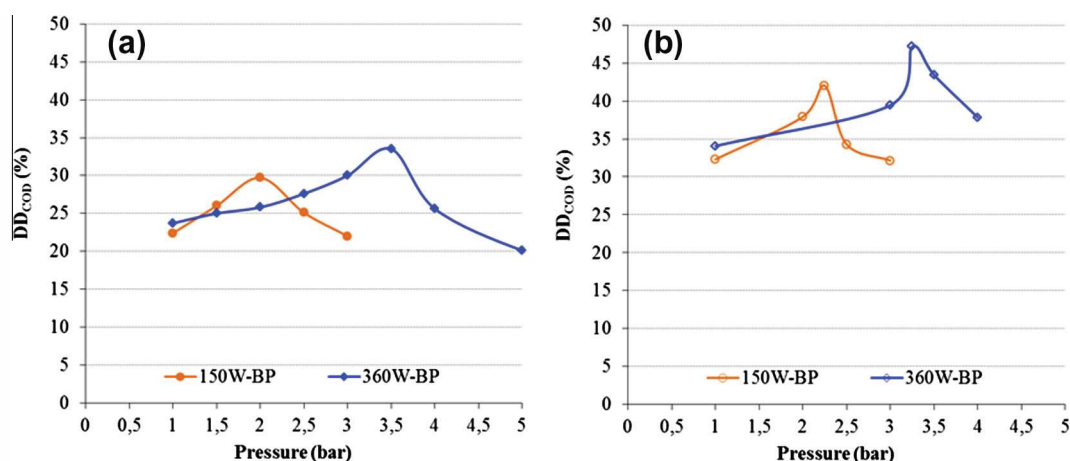


Fig. 9. Effect of hydrostatic pressure on  $DD_{COD}$  of WAS (Table 1b) for different  $P_{US}$  (BP,  $ES = 35,000$  kJ/kg<sub>TS</sub>): (a)  $F_s = 20$  kHz, (b)  $F_s = 12$  kHz.

increasing the power intensity. This is the reason of such difference at 50 W between BP and SP under pressure, which could be even more important at lower power where the cavitation threshold would be hardly reached with BP.

### 3.5.2. Effect of frequency on optimum pressure

Synthetic WAS samples given in Table 1b were used to investigate the effect of very low  $F_s$  on the optimum pressure and subsequent  $DD_{COD}$ . An  $ES$  value of 35,000 kJ/kg<sub>TS</sub> was applied using the 12 kHz sonicator with  $P_{US}$  of 150 and 360 W through BP. Based on the results observed at 20 kHz, the pressure range 1–4 bar was more carefully studied with closer pressure intervals: 0.25 bar. Results are presented in Fig. 9a showing very similar  $DD_{COD}$  variations with pressure for each power. As found at 20 kHz (Fig. 9b), the optimum pressure shifted when increasing  $P_{US}$ . Besides, the location of this optimum seemed to be independent from  $F_s$  in the restricted investigated range: 2 bar at 150 W and 3.5 bar at 360 W (using 0.5 bar intervals) for 20 kHz as compared to 2.25 bar at 150 W and 3.25 bar at 360 W (0.25 bar intervals) for 12 kHz.

It was also verified by comparing Fig. 9a and b that higher  $DD_{COD}$  was achieved at 12 kHz with 30–40% improvement in the whole pressure range, including the optimum pressure. Contrary to power density and intensity the positive effect of very low frequency is very straightforward.

This continuous improvement of sludge solubilization when lowering frequency, well known also for any US mechanical effect in the usual frequency range (20–100 kHz), has been demonstrated and quantified here down to audible range. As it is expected to be also verified in most US applications it would deserve future work towards even lower frequencies.

## 4. Conclusions

An extensive parameter investigation has been performed based on sludge disintegration assessed by COD solubilization and the specific energy (US energy per solid weight) as the relevant criterion for rational comparisons.

The main results concern the importance of hydrostatic pressure which highlights the separate roles of power density and power intensity. At any investigated condition ( $P_{US}$ ,  $I_{US}$ ,  $F_s$ ) a clear optimal pressure was observed due to opposite effects of pressurization: a negative one on the bubble number and size connected to enhanced cavitation threshold, but a positive one on bubble collapse characteristics ( $P_{max}$ ,  $T_{max}$ ). The higher the power intensity (and then the higher acoustic pressure  $P_A$ ) and power density, the higher is the optimum hydrostatic pressure – since much lower than  $P_A$  – providing also higher disintegration. In given equipment,

at same specific energy, US performance might be more than doubled by selecting high power and optimum pressure. Nevertheless at a fixed pressure the usual recommendation “high power-short sonication time” might fail: a lower power but closer to its optimum pressure could perform better.

In addition, audible frequency was successfully tested: in any case with same conditions 12 kHz outperforms 20 kHz. This opens the way to even lower frequencies and to find a possible optimal frequency as observed at high frequency for US radical sonochemistry.

This work should be extended to other applications of US mechanical effects to verify and quantify the very positive role of hydrostatic pressure. Using high power and the corresponding optimal hydrostatic pressure would allow very important energy savings, by partly replacing acoustic pressure by hydrostatic pressure, and in addition much shorter treatment time, then smaller equipment.

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## References

- [1] T.J. Mason, J.P. Lorimer, *Applied Sonochemistry: Uses of Power Ultrasound in Chemistry and Processing*, Wiley, 2003.
- [2] M.P. Brenner, S. Hilgenfeldt, D. Lohse, Single bubble sonoluminescence, *Rev. Mod. Phys.* 74 (2002) 425–484.
- [3] K.S. Suslick, D.J. Flannigan, Inside a collapsing bubble: sonoluminescence and the conditions during cavitation, *Annu. Rev. Phys. Chem.* 59 (2008) 659–683.
- [4] P.R. Gogate, V.S. Sutkar, A.B. Pandit, Sonochemical reactors: important design and scale up considerations with a special emphasis on heterogeneous systems, *Chem. Eng. J.* 166 (2011) 1066–1082.
- [5] T.J. Mason, A.J. Cobley, J.E. Graves, D. Morgan, New evidence for the inverse dependence of mechanical and chemical effects on the frequency of ultrasound, *Ultrason. Sonochem.* 18 (2011) 226–230.
- [6] K.V.B. Tran, T. Kimura, T. Kondo, S. Koda, Quantification of frequency dependence of mechanical effects induced by ultrasound, *Ultrason. Sonochem.* 21 (2014) 716–721.
- [7] L. Zeng, J.W. McKinley, Degradation of pentachlorophenol in aqueous solution by audible-frequency sonolytic ozonation, *J. Hazard. Mater.* 135 (2006) 218–225.
- [8] M. Sivakumar, A.B. Pandit, Ultrasound enhanced degradation of Rhodamine B: optimization with power density, *Ultrason. Sonochem.* 8 (2001) 233–240.
- [9] T. Bosiljkov, B. Tripalo, M. Brnčić, D. Ježek, S. Karlović, I. Jagaš, Influence of high intensity ultrasound with different probe diameter on the degree of

- homogenization (variance) and physical properties of cow milk, *Afr. J. Biotechnol.* 10 (2011) 34–41.
- [10] G. Andaluri, E.V. Rokhina, R.P.S. Suri, Evaluation of relative importance of ultrasound reactor parameters for the removal of estrogen hormones in water, *Ultrason. Sonochem.* 19 (2012) 953–958.
- [11] E.C. Couppis, G.E. Klinzing, Effect of cavitation on reacting systems, *AIChE J.* 20 (1974) 485–491.
- [12] E.A. Neppiras, D.E. Hughes, Some experiments on the disintegration of yeast by high intensity ultrasound, *Biotechnol. Bioeng.* 6 (1964) 247–270.
- [13] O. Behrend, H. Schubert, Influence of hydrostatic pressure and gas content on continuous ultrasound emulsification, *Ultrason. Sonochem.* 8 (2001) 271–276.
- [14] C. Sauter, M.A. Emin, H.P. Schuchmann, S. Tavman, Influence of hydrostatic pressure and sound amplitude on the ultrasound induced dispersion and deagglomeration of nanoparticles, *Ultrason. Sonochem.* 15 (2008) 517–523.
- [15] T. Leong, M. Ashokkumar, S. Kentish, The fundamentals of power ultrasound – a review, *Acoust. Aust.* 39 (2011) 54–63.
- [16] M. Geng, S.M. Thagard, The effects of externally applied pressure on the ultrasonic degradation of Rhodamine B, *Ultrason. Sonochem.* 20 (2013) 618–625.
- [17] K.Y. Show, T. Mao, D.J. Lee, Optimization of sludge disruption by sonication, *Water Res.* 41 (2007) 4741–4747.
- [18] H. Carrère, C. Dumas, A. Battimelli, D.J. Batstone, J.P. Delgenès, J.P. Steyer, I. Ferrer, Pretreatment methods to improve sludge anaerobic degradability: a review, *J. Hazard. Mater.* 183 (2010) 1–15.
- [19] S. Pilli, P. Bhunia, S. Yan, R.J. LeBlanc, R.D. Tyagi, R.Y. Surampalli, Ultrasonic pretreatment of sludge: a review, *Ultrason. Sonochem.* 18 (2011) 1–18.
- [20] V.K. Tyagi, S.L. Lo, L. Appels, R. Dewil, Ultrasonic treatment of waste sludge: a review on mechanisms and applications, *Crit. Rev. Environ. Sci. Technol.* 44 (2014) 1220–1288.
- [21] A. Tiehm, K. Nickel, M.M. Zellhorn, U. Neis, Ultrasound waste activated sludge disintegration for improving anaerobic stabilization, *Water Res.* 35 (2001) 2003–2009.
- [22] G. Zhang, P. Zhang, J. Gao, Y. Chen, Using acoustic cavitation to improve the bio-activity of activated sludge, *Bioresour. Technol.* 99 (2008) 1497–1502.
- [23] F. Wang, Y. Wang, M. Ji, Mechanisms and kinetics models for ultrasonic waste activated sludge disintegration, *J. Hazard. Mater.* B123 (2005) 145–150.
- [24] G. Zhang, P. Zhang, J. Yang, H. Liu, Energy-efficient sludge sonication: power and sludge characteristics, *Bioresour. Technol.* 99 (2008) 9029–9031.
- [25] C. Liu, B. Xiao, A. Dauta, G. Peng, S. Liu, Z. Hu, Effect of low power ultrasonic radiation on anaerobic biodegradability of sewage sludge, *Bioresour. Technol.* 100 (2009) 6217–6222.
- [26] C. Li, G. Liu, R. Jin, J. Zhou, J. Wang, Kinetics model for combined (alkaline + ultrasonic) sludge disintegration, *Bioresour. Technol.* 101 (2010) 8555–8557.
- [27] U. Neis, K. Nickel, A. Tiehm, Enhancement of anaerobic sludge digestion by ultrasonic disintegration, *Water Sci. Technol.* 42 (2000) 73–80.
- [28] N.T. Le, C. Julcour-Lebigue, H. Delmas, Ultrasonic sludge pretreatment under pressure, *Ultrason. Sonochem.* 20 (2013) 1203–1210.
- [29] N.T. Le, C. Julcour, B. Ratsimba, H. Delmas, Improving sewage sludge ultrasonic pretreatment under pressure by changing initial pH, *J. Environ. Manage.* 128 (2013) 548–554.
- [30] R. Kidak, A.M. Wilhelm, H. Delmas, Effect of process parameters on the energy requirement in ultrasonical treatment of waste sludge, *Chem. Eng. Process.* 48 (2009) 1346–1352.
- [31] J.A. Bamberger, M.S. Greenwood, Measuring fluid and slurry density and solids concentration non-invasively, *Ultrasonics* 42 (2004) 563–567.
- [32] J.P.Y. Maa, K.J. Sun, Q. He, Ultrasonic characterization of marine sediments: a preliminary study, *Mar. Geol.* 141 (1997) 183–192.
- [33] H.K. Ha, Acoustic measurements of cohesive sediment transport: suspension to consolidation (PhD thesis), The Faculty of the School of Marine Science, The College of William and Mary in Virginia, 2008, <<http://web.vims.edu/library/theses/Ha08.pdf?svr=www>>, (accessed 20.07.14).
- [34] B.L. Johnson, M.R. Holland, J.G. Miller, J.I. Katz, Ultrasonic attenuation and speed of sound of cornstarch suspensions, *J. Acoust. Soc. Am.* 133 (2013) 1399–1403.
- [35] R. Weser, S. Wöckel, U. Hempel, B. Wessely, J. Auge, Particle characterization in highly concentrated suspensions by ultrasound scattering method, *Sens. Actuators, A: Phys.* 202 (2013) 30–36.
- [36] APHA, AWWA, WEF, Standard Methods for the Examination of Water and Wastewater, 21st ed., American Public Health Association, Washington D.C., 2005.
- [37] U. Schmitz, C.R. Berger, H. Orth, Protein analysis as a simple method for the quantitative assessment of sewage sludge disintegration, *Water Res.* 34 (2000) 3682–3685.
- [38] F. Contamine, F. Faid, A.M. Wilhelm, J. Berlan, H. Delmas, Chemical reactions under ultrasound: discrimination of chemical and physical effects, *Chem. Eng. Sci.* 49 (1994) 5865–5873.
- [39] N. Ratoarinoro, F. Contamine, A.M. Wilhelm, J. Berlan, H. Delmas, Power measurement in sonochemistry, *Ultrason. Sonochem.* 2 (1995) S43–S47.
- [40] G.O.H. Whillock, B.F. Harvey, Ultrasonically enhanced corrosion of 304 L stainless steel II: the effect of frequency, acoustic power and horn to specimen distance, *Ultrason. Sonochem.* 4 (1997) 33–38.
- [41] C.P. Chu, B.V. Chang, G.S. Liao, D.S. Jean, D.J. Lee, Observations on changes in ultrasonically treated waste-activated sludge, *Water Res.* 35 (2001) 1038–1046.
- [42] T. Mao, S.Y. Hong, K.Y. Show, J.H. Tay, D.J. Lee, A comparison of ultrasound treatment on primary and secondary sludges, *Water Sci. Technol.* 50 (2004) 91–97.
- [43] A. Grönroos, H. Kyllönen, K. Korpjärvi, P. Pirkonen, T. Paavola, J. Jokela, J. Rintala, Ultrasound assisted method to increase soluble chemical oxygen demand (SCOD) of sewage sludge for digestion, *Ultrason. Sonochem.* 12 (2005) 115–120.
- [44] J. Quarmby, J.R. Scott, A.K. Mason, G. Davies, S.A. Parsons, The application of ultrasound as a pre-treatment for anaerobic digestion, *Environ. Technol.* 20 (1999) 1155–1161.
- [45] J.L. Laborde, C. Bouyer, J.P. Caltagirone, A. Gérard, Acoustic bubble cavitation at low frequencies, *Ultrasonics* 36 (1998) 589–594.
- [46] E.A. Neppiras, Acoustic cavitation, *Phys. Rep.* 61 (1980) 159–251.