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Ultrasonic sludge pretreatment under pressure

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A B S T R A C T

The objective of this work was to optimize the *ultrasound (US) pretreatment of sludge*. Three types of sewage sludge were examined: mixed, secondary and secondary after partial methanisation ("digested" sludge). Thereby, several main process parameters were varied separately or simultaneously: stirrer speed, total solid content of sludge *(TS)*, thermal operating conditions (adiabatic vs. isothermal), ultrasonic power input *(P_{US})*, specific energy input *(ES)*, and for the first time external pressure. This parametric study was mainly performed for the mixed sludge. Five different *TS* concentrations of sludge (12–36 g/L) were tested for different values of *ES* (7000–75,000 kJ/kg_{TS}) and 28 g/L was found as the optimum value according to the solubilized chemical oxygen demand in the liquid phase (*SCOD*). P_{US} of 75–150 W was investigated under controlled temperature and the "*high power input – short duration*" procedure was the most effective at a given *ES*. The temperature increase in adiabatic *US* application significantly improved *SCOD* compared to isothermal conditions. With P_{US} of 150 W, the effect of external pressure was investigated in the range of 1–16 bar under isothermal and adiabatic conditions for two types of sludge: an optimum pressure of about 2 bar was found regardless of temperature conditions and *ES* values. Under isothermal conditions, the resulting improvement of sludge disintegration efficacy as compared to atmospheric pressure was by 22–67% and 26–37% for mixed and secondary sludge, respectively. Besides, mean particle diameter (*D[4,3]*) of the three sludge types decreased respectively from 408, 117, and 110 μm to about 94–97, 37–42, and 36–40 μm regardless of sonication conditions, and the size reduction process was much faster than *COD* extraction.

1. Introduction

Due to economic reasons and/or negative impacts on environment, incineration, composting, ocean discharge, and land spreading, known as the most common sludge disposal options used over the years, are no longer sustainable. Meanwhile, anaerobic digestion (*AD*) of sludge is an efficient and sustainable technology for sludge treatment. However, a pretreatment of sludge, which ruptures the cell wall and facilitates the release of intracellular matters into the aqueous phase, is required to enhance the *AD* as hydrolysis is the rate-limiting step of microbial conversion.

Ultrasound (*US*) irradiation has been reported as a promising mechanical disruption technique, resulting in improved biodegradability and bio-solid quality [1], increased methane production [1–3], sludge reduction [3,4], and less sludge retention time [5].

Despite ultrasonic sludge treatment reached commercial developments and gave rise to many works, none of them was carried out to investigate the effect of pressure. Changing the hydrostatic pressure will change the resonance condition of cavitation bubbles via their equilibrium radius and then may drive the system toward resonance conditions [6]. At resonance conditions, the rate and yield of reactions will increase [7–9]. More probably, both the cavitation threshold and the intensity of cavity collapse increase following an increase in external pressure [10], suggesting a possible optimum pressure. Brett and Jellinek [11] stated that bubbles could be visible for gas-applied pressure as high as 16 atm. Nevertheless, nearly all the *US* experiments have been carried out at atmospheric pressure. Only a few studies have been focusing on how increasing static pressure affects cavitation.

Most works on pressure effects concern sonoluminescence, and no consensus emerges about an optimum value as reported by Chendke and Fogler [11,12]. The early works of Finch (1955) cited by the authors indicated that the greatest sonoluminescence intensity was observed in water at a static pressure of about 1.5 atm (over an investigated range of 1–8 atm), but Chendke and Fogler recommended a value of 6 atm to promote sonoluminescence in nitrogen-saturated water [11]. In aqueous carbon tetrachloride solutions, the intensity of the sonoluminescence did not show any monotonous behavior: it first went up to 6 atm, then reached a minimum at 8 atm, got a new maximum at 12 atm, and was finally almost inhibited above 18 atm [12]. Whillock and Harvey [13] investigated the effects of hydrostatic pressure on the corrosion of 304L stainless steel in an ultrasonic field. An increase in pressure

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Table 1
Characteristics of the sludge samples.

Parameter	Value				
	Mixed sludge		Secondary sludge		Digested sludge
Raw sludge samples					
Total solids (*TS*)	285 mg/g		37.5 g/L		14.0 g/L
Volatile solids (*VS*)	238 mg/g		32.2 g/L		11.9 g/L
VS/*TS*	83.5%		85.8%		84.7%
Synthetic sludge samples					
Total solids (*TS*)	28.0 g/L	28.0 g/L	28.0 g/L	14.0 g/L	14.0 g/L
$SCOD_{NaOH\ 0.5\ M}$	18.5 g/L	11.3 g/L	22.9 g/L	14.0 g/L	11.0 g/L
TCOD	36.5 g/L	18.3 g/L	38.2 g/L	19.1 g/L	15.0 g/L

up to 4 bar at a constant temperature caused a strong increase in corrosion rate. Closer to the present subject, Neppiras and Hughes [14] investigated the influence of pressure (up to 5.8 atm) on the disintegration of yeast cells and found an optimum value of 4 atm.

Following these researches, static pressure seems to be an important parameter, but it has been marginally investigated due to the complex equipments required. In case of sludge pretreatment, external pressure should be varied simultaneously with other related parameters, including total solid content of sludge (*TS*), *US* power input (P_{US}), specific energy input (*ES*), thermal operating conditions, etc., in order to select optimal conditions for actual application. The effect of ultrasound will be presented in terms of disintegration degree (organic matters solubilized in liquid phase) and particle size reduction. The objective of this work is to optimize high-power low-frequency ultrasonic pretreatment of sludge, and especially to emphasize on static pressure for the first time, which is expected to enhance sludge disintegration, to increase methane production, and to facilitate the *AD*.

2. Materials and methods

2.1. Sludge samples

Three types of sludge were collected from Ginestous wastewater treatment plant (Toulouse, France) with a sufficient amount for all experiments in this study: mixed sludge (solid form, after centrifugation), secondary sludge (liquid form), and digested sludge

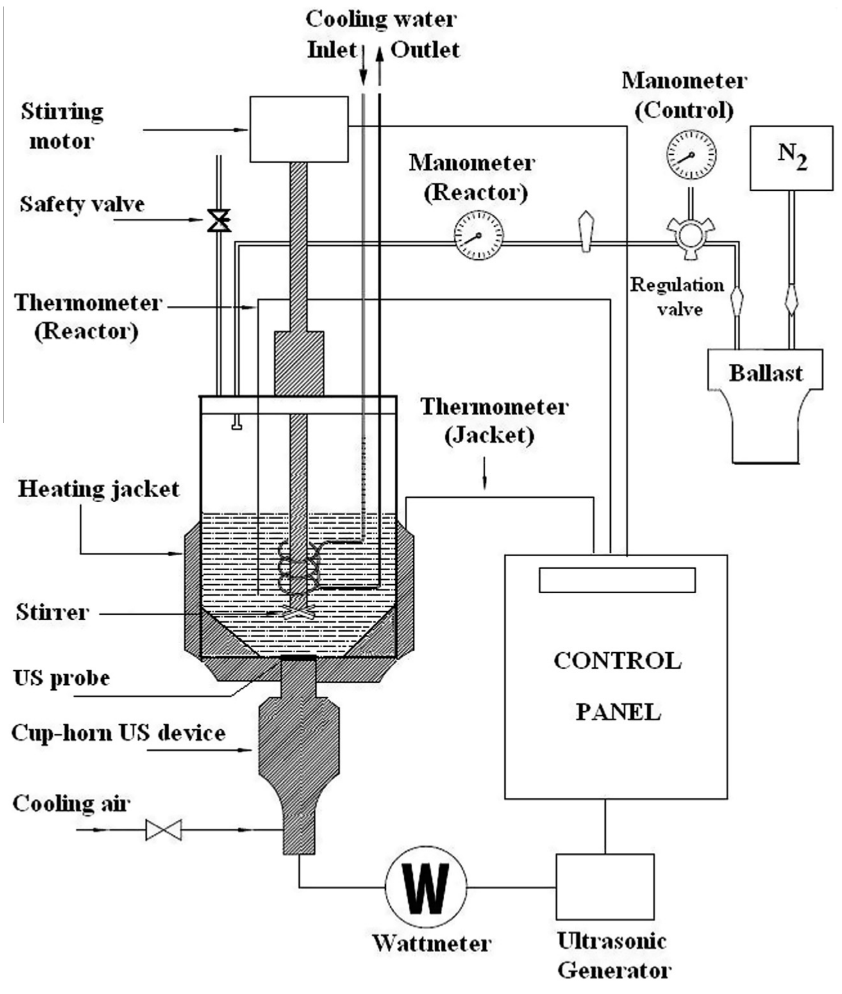

Fig. 1. Ultrasonic autoclave set-up.

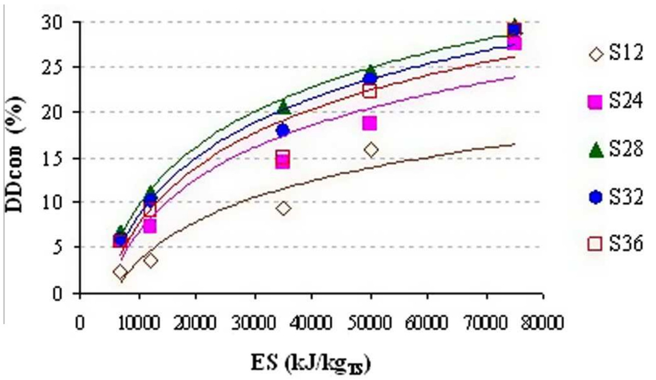

Fig. 2. Effect of *TS* content on mixed sludge disintegration (DD_{COD}): P_{US} = 150 W, adiabatic condition and atmospheric pressure.

(liquid form, after anaerobic digestion process of the secondary sludge). Their properties, given in Table 1, were evaluated according to standard analytical methods (see Section 2.3).

For mixed sludge, it was sampled in 100 g plastic boxes and preserved in a freezer. Kidak et al. [15] reported that this preliminary maintaining step might change some physical characteristics of the sludge, but it should not significantly affect *COD* solubilisation results. It was confirmed in the present study, the difference in sludge disintegration between fresh sludge (without freezing) and frozen sludge was less than 5% on the whole *ES* range (7000–75,000 kJ/kg$_{TS}$).

Secondary and digested sludge lots were sampled in 1L plastic boxes and stored at a constant temperature of 3–4 °C.

When performing experiments, the required amount of sludge was defrosted (for frozen sludge) and diluted with distilled water to prepare synthetic sludge samples with a given *TS* content.

2.2. Ultrasound application

Ultrasonic irradiation was emitted by a cup-horn ultrasound unit (see Fig. 1) included in an autoclave reactor which was connected to a pressurized N_2 bottle.

The reactor and its internals were made of 316L stainless steel. The reactor internal diameter was 9 cm and its depth 18 cm, for a usable capacity of 1 L. A safety valve (HOKE 6500) limited overpressure to 19 bar. The solution was stirred by a Rushton type turbine of 32 mm diameter, with an adjustable speed up to 3000 rpm. A cooling water stream (15 °C) was continuously circulated in an internal coil to maintain a constant temperature (*T*) of the solution at 28 ± 2 °C during sonication.

The ultrasound system had a fixed frequency of 20 kHz and a maximum total power of 200 W corresponding to P_{US} of 158 W. The *US* device, supplied by Sinaptec, was composed of four elements: a piezoelectric transducer (M202045), a titanium booster (B20B), an aluminum flange (AU4G) ensuring a good mechanical connection, and an ultrasonic cup horn (PLANUS P2035041, 35 mm diameter probe) placed at the bottom of the reactor. The transducer was cooled by compressed air during operation.

Prior to the application of external pressure to *US* pretreatment, some process parameters were examined separately to identify adequate values. For each experiment, a constant volume of synthetic sludge sample (0.5 L) was poured into the stainless steel reactor. Five different sonication times corresponding to five values of *ES* (7000, 12,000, 35,000, 50,000, and 75,000 kJ/kg$_{TS}$) were tested.

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

with *ES*: specific energy input, energy per total solid weight (kJ/kg$_{TS}$), P_{US}: *US* power input (W), *t*: sonication duration (s), *V*: volume of sludge (L), and *TS*: total solid concentration (g/L).

First, the influence of *TS* content (12–36 g/L), stirrer speed (250–1500 rpm), and P_{US} (75–150 W) along with ultrasonic duration was investigated for mixed sludge disintegration. Afterwards, separate and combined effects of ultrasound and temperature (which increased due to *US*) were examined for mixed and secondary sludge. The effect of external pressure (in the range 1–16 bar) was then evaluated for these two types of sludge. Finally, the best combination of process parameters was subsequently tested for all the sludge samples.

2.3. Analytical methods

Total and volatile solids contents (*TS* and *VS*, respectively) were measured according to the following procedure (APHA, 2005). *TS* was determined by drying a well-mixed sample to constant weight at 105 °C. *VS* was obtained from the weight loss on ignition of the residue at 550 °C.

The *degree of sludge disintegration* (DD_{COD}) was calculated by determining the soluble chemical oxygen demand after strong alkaline disintegration of sludge ($SCOD_{NaOH}$) and the chemical oxygen demand in the supernatant before and after treatment ($SCOD_0$ and *SCOD*, respectively) [16]:

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

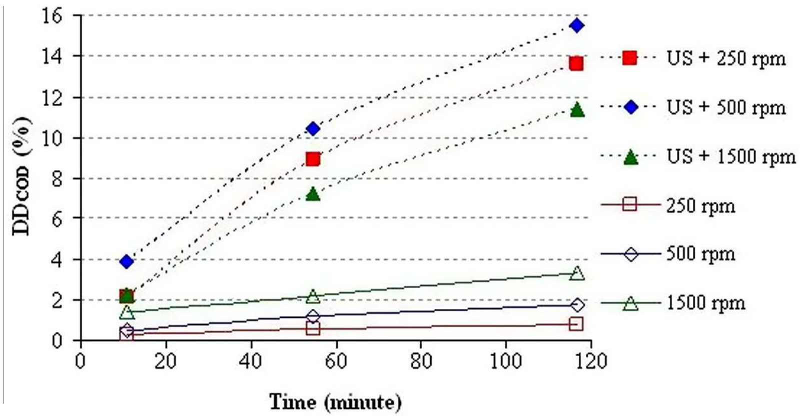

Fig. 3. Effect of the stirrer speed on mixed sludge disintegration (DD_{COD}): P_{US} = 150 W, (when under *US*), controlled *T* (28 ± 2 °C), and atmospheric pressure.

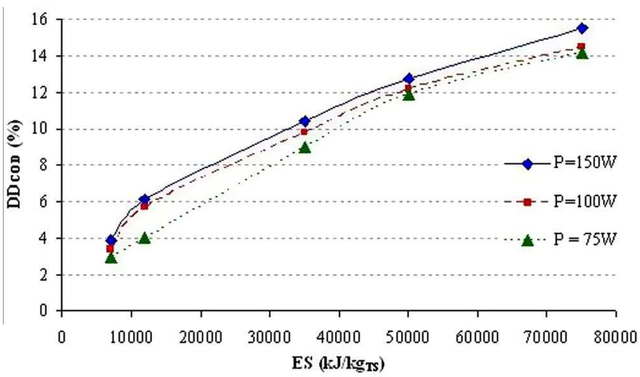

Fig. 4. Effect of specific energy input *ES* with three P_{US} on mixed sludge disintegration (DD_{COD}): t = 0–233 min, controlled T (28 ± 2 °C), and atmospheric pressure.

To measure the $SCOD_{NaOH}$ value, used as reference to evaluate the efficiency of organic matter solubilization under *US*, the sludge sample was mixed with 0.5 M NaOH at room temperature for 24 h [17]. Besides, total chemical oxygen demand (*TCOD*) was also measured by potassium dichromate oxidation method (standard AFNOR NFT 90-101).

Prior to *SCOD* determination, the supernatant liquid was filtered under vacuum using a cellulose nitrate membrane with 0.2 μm pore size. *COD* of the filtered liquid was measured as per Hach spectrophotometric method. The change in the *SCOD* indirectly represents the quantity of organic carbon that has been transferred from the cell content (disruption) and solid materials (solubilisation) into the external liquid phase of sludge. The experiments were triplicated and the coefficients of variation (*CV*) were about 5%.

The *particle size distribution* (*PSD*) of sludge before and after treatment was determined by using Malvern particle size analyzer (Mastersizer 2000, Malvern Inc.), a laser diffraction-based system (measuring range from 0.02 to 2000 μm). Each sample was diluted approximately 300-fold in osmosed water, before being pumped into the measurement cell (suction mode). The *PSD* was based on the average of five measurements showing deviations of less than 3%. Optical properties of the material were set as default (refractive index 1.52, absorption 0.1) appropriate for the majority of naturally occurring substances. Since the primary result from laser diffraction is a volume distribution, the volume mean diameter *D[4,3]* (or de Brouckere mean diameter) was used to illustrate the mean particle size of sludge.

3. Results and discussion

3.1. Effect of TS concentration on DD_{COD}

Five synthetic mixed sludge samples (S12, S24, S28, S32, and S36 corresponding to 12, 24, 28, 32, and 36 g/L of *TS*, respectively) were treated at atmospheric pressure, under adiabatic condition, and at a constant P_{US} of 150 W – close to the maximum, because "high power and short time" of *US* should be preferred for a given *ES* [15]. The respective *ES* was varied (7000, 12,000, 35,000, 50,000, and 75,000 kJ/kg_{TS}) via the sonication time. The stirrer speed was adjusted to 500 rpm. The results are presented in Fig. 2.

SCOD gradually increased with sonication time (0–150 min) but less and less and the relation between *SCOD* and *TS* content was not simple because the best DD_{COD} was not found at the maximum *TS*. For example, at *ES* of 7000 kJ/kg_{TS}, *SCOD* was improved by 2.4-fold when increasing *TS* from 12 to 24 g/L, but did not significantly change for higher values. Fig. 2 actually exhibited a *TS* optimal value of 28 g/L in terms of DD_{COD} over the whole ES range. This behavior is in agreement with other studies [4,15,18–21] and can be explained by explained by opposite effects. The increase in *TS* provides more cells and aggregates to be in contact with cavitation bubbles; thereby, the *US* power input, which is required to generate cavitation, is more efficiently consumed. Nevertheless, at higher sludge loading, the acoustic pressure field decreases faster from the emitter due to the degraded propagation of the ultrasonic wave in a denser suspension. Consequently, acoustic cavitation intensity will be reduced. These two opposite effects lead to an optimum *TS* concentration that could slightly depend on sludge characteristics, operating conditions, reactor design, *US* power, and *US* frequency, etc.

For all the following experiments of this work (excepting those with digested sludge), synthetic samples were prepared to match this 28 g/L *TS* concentration corresponding to values of *TCOD* and $SCOD_{NaOH}$ given in Table 1.

3.2. Effect of stirrer speed on DD_{COD}

To decorrelate the effect of stirrer speed on DD_{COD} when *US* is applied, preliminary experiments at 250, 500, and 1500 rpm were carried out under ambient conditions (controlled *T* of 28 ± 2 °C,

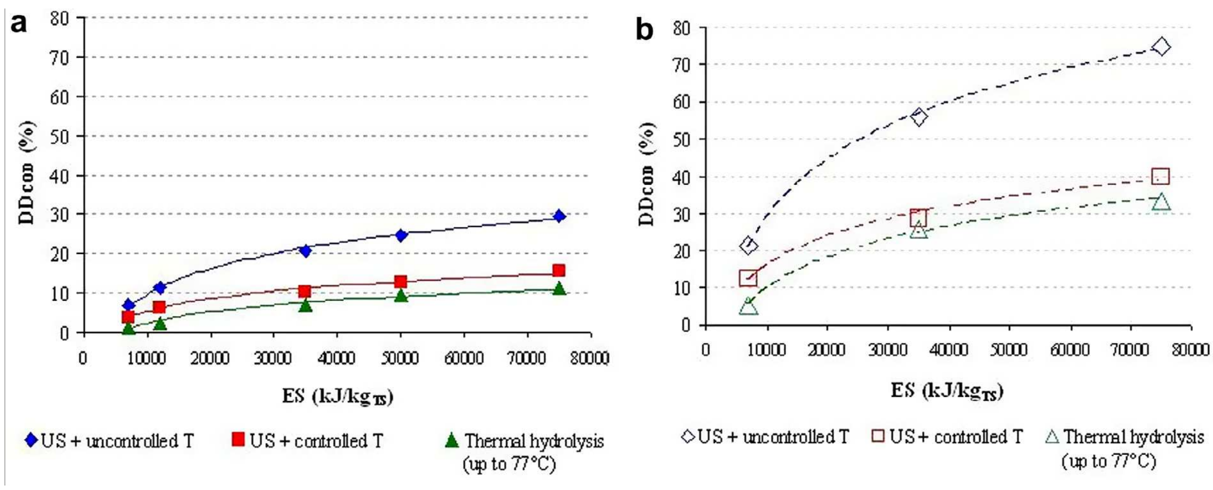

Fig. 5. Effect of temperature on (a) mixed sludge and (b) secondary sludge disintegration (DD_{COD}): P_{US} = 150 W (0 W for thermal hydrolysis), *TS* = 28 g/L, and atmospheric pressure.

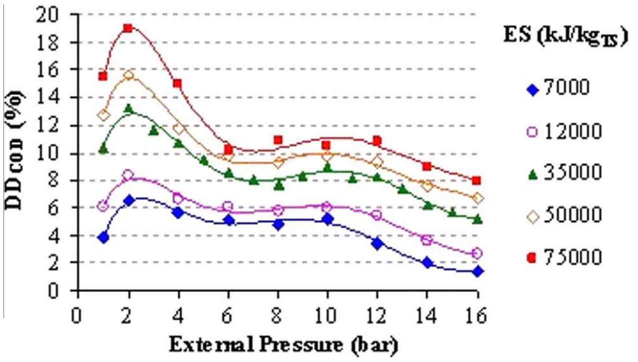

Fig. 6. Effect of external pressure on mixed sludge disintegration (DD_{COD}): P_{US} = 150 W, controlled T (28 °C), and TS = 28 g/L.

atmospheric pressure) with mixed sludge. Fig. 3 exhibits the resulting time-evolution of DD_{COD}.

As expected, for blank experiments (without *US*), the faster the stirring was, the higher the sludge disintegration was: after 2 h of stirring, DD_{COD} was 0.8%, 1.8%, and 3.3% for a stirrer speed of 250, 500, and 1500 rpm, respectively. However, these DD_{COD} values as well as the differences observed among the three corresponding series under *US* were rather low, which indicated that the main role of the stirrer was to make a homogeneous solution, rather than to significantly enhance the transfer of organic matters from solid to aqueous phase.

Under *US*, DD_{COD} increased when raising the stirrer speed from 250 rpm to 500 rpm, but decreased at 1500 rpm. The reactor was not equipped with baffles. Consequently high rotation speed of the whole liquid resulted in the centrifugation of particles, leading to less particles present in the central zone where *US* is concentrated, then to a decrease of the sludge *US* pretreatment efficiency. In addition, aeration could occur and its main effect would be to severely damp the acoustic waves. Therefore, a stirrer speed of 500 rpm was applied in subsequent experiments of this work.

3.3. Effect of US power input along with sonication duration

Three different P_{US} (75, 100, and 150 W) were tested under a controlled T of 28 °C and at atmospheric pressure. In each case, *ES* values of 7000, 12,000, 35,000, 50,000, and 75,000 kJ/kg$_{TS}$ were applied by varying the sonication duration. The corresponding performance reflected by DD_{COD} is illustrated in Fig. 4.

For all P_{US}, the disintegration of sludge increased gradually with sonication time t. A quasi-linear increase of DD_{COD} was observed in the *ES* range of 0–50000 kJ/kg$_{TS}$ (up to about 12–13%), followed by a slower increase until the end of the process (about 14–16% at *ES* of 75,000 kJ/kg$_{TS}$). This complies with recent researches [5,19,21].

For a given *ES* value, DD_{COD} was the highest in 150 W experiments, followed by 100 W and 75 W experiments. This effect was best observed in the first stage of the process (ES < 50000 kJ/kg$_{TS}$). Afterwards ($ES \geqslant$ 50000 kJ/kg$_{TS}$), DD_{COD} values did not exhibit notable discrepancies for most combinations of P_{US} and t. For instance, the highest differences were observed at *ES* of 12,000 kJ/kg$_{TS}$: DD_{COD} of [75 W–37 min] and [100 W–28 min] experiments represented respectively, 66% and 93% of that measured after applying 150 W during 19 min. At *ES* of 75,000 kJ/kg$_{TS}$, DD_{COD} values obtained for all P_{US} differed by less than 10%.

Although it did not result in a significant enhancement of DD_{COD}, the *"high power input – short duration"* sonication procedure proved, again, to be the most effective combination for sludge pretreatment in isothermal conditions, as already reported by other researchers [4,15,18,22,23]. The reason could be attributed to the relative resistance of municipal sludge particles to ultrasonic disruption (especially fibrous particles), requiring high values of P_{US} [15]. A *US* power input of 150 W was applied in all following experiments.

3.4. Effect of temperature and of sludge type on DD_{COD}

The ultrasonic pretreatment has two simultaneous effects: (i) extreme macro and micro mixing caused by the cavitation, and (ii) increase in the bulk temperature. To evaluate their individual contribution, three operating procedures were carried out for mixed and secondary sludge: (1) *US* under isothermal conditions (cooling at 28 °C), (2) *US* under adiabatic conditions, (3) thermal hydrolysis: without *US*, progressive increase of T up to 77 °C as found in (2).

Results, illustrated in Fig. 5a and b, show the disintegration (ultrasonic or thermal pretreatments) of secondary sludge to be about 3-fold higher than that of mixed sludge. As confirmed by Show et al. [21], secondary sludge, mainly composed of biological substances (derived from activated processes), is readily disrupted, while mixed sludge (mixture of primary and secondary sludge) contains many non-degradable materials from primary sludge (plastic, textile, fibrous, born, sand...) that cannot be easily disintegrated.

At all observed times and with both types of sludge, DD_{COD} values under adiabatic sonication were the highest, followed by those at low temperature sonication and thermal hydrolysis. DD_{COD} values of sonicated samples under cooling (28 °C) were about half those obtained under adiabatic conditions (uncontrolled T).

In accordance with recent works [15,17,22], the higher the temperature, the higher the ultrasonic disintegration efficiency. This is

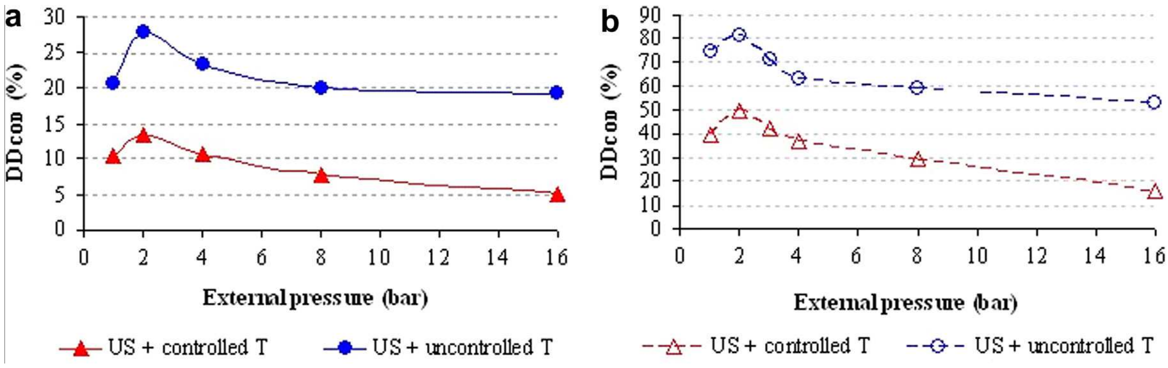

Fig. 7. Effect of external pressure on (a) mixed sludge and (b) secondary sludge disintegration (DD_{COD}): under different temperature conditions: P_{US} = 150 W, TS = 28 g/L. (a) ES = 35,000 kJ/kg$_{TS}$. The final temperature in adiabatic mode was about 75 °C. (b) ES = 75,000 kJ/kg$_{TS}$. The final temperature in adiabatic mode was about 85 °C.

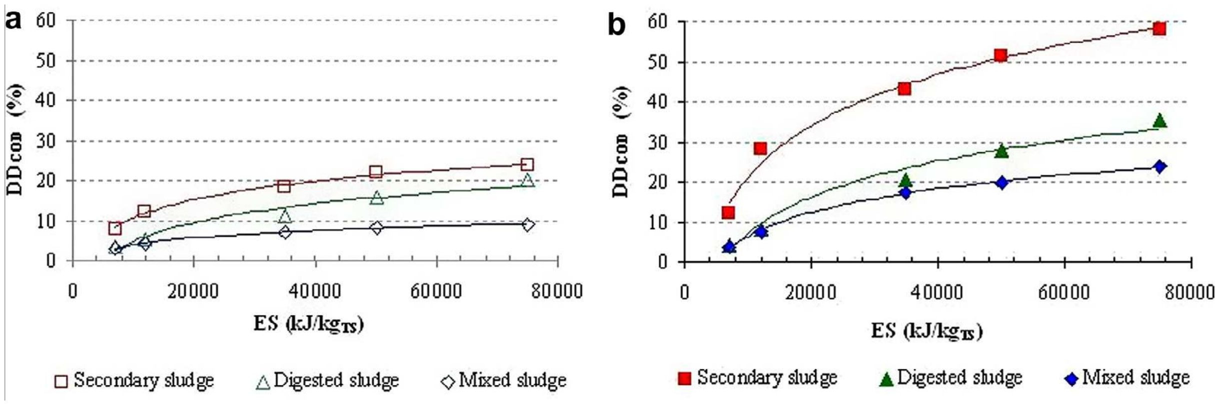

Fig. 8. Effect of specific energy input *ES* on ultarsonic pretreatment efficacy of different sludge types at optimum pressure and different temperature conditions: P_{US} = 150 W and *TS* = 14 g/L (a) isothermal condition (28 °C) and (b) adiabatic condition.

opposite to most power *US* applications as cavitation intensity is higher at low temperature. In short, it is clear that ultrasonic disintegration of sludge is the result of two different effects: the specific cavitation effect and the thermal effect. Despite lower performances, next experiments were conducted under isothermal condition to have a clear understanding of *US* effect under different values of static pressure.

3.5. Effect of external pressure on DD_{COD}

Experiments to investigate the effect of the external pressure (1–16 bar) on the efficacy of ultrasonic pretreatment of sludge were carried out for mixed sludge in the following conditions: optimum *TS* of 28 g/L, isothermal mode, P_{US} of 150 W, and *ES* in the range of 0–75,000 kJ/kg_{TS}. Results are presented in Fig. 6.

All curves corresponding to different *ES* values show the same trends of DD_{COD}: an initial increase up to 2 bar and a decrease thereafter, 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 and this effect was relatively high at low *ES*, with a maximum improvement of 67% at 7000 kJ/kg_{TS} (Fig. 6). It is interesting to note that beyond the optimum pressure (about 2 bar), the decrease of DD_{COD} was faster at higher *ES*. With the exception of the lowest *ES* (7000 and 12,000 kJ/kg_{TS}), all DD_{COD} values were lower at 6 bar than those at atmospheric pressure. Nevertheless, the positive pressure effect up to 2 bar might lead to energy savings in sludge pretreatment applications with ultrasound. For instance, at the optimum pressure, DD_{COD} obtained with ES of 7000, 35,000, and 50,000 kJ/kg_{TS} were higher than those at atmospheric pressure with *ES* of 12,000, 50,000, and 75,000 kJ/kg_{TS}, respectively.

To examine the effect of pressure (1–16 bar) along with temperature during sonication, further experiments were performed under adiabatic condition. The results, shown in Fig. 7a and b, once again confirmed the optimum pressure found in this work to be about 2 bar regardless of temperature and sludge type.

According to Thompson and Doraiswamy [6], increasing the external pressure increases the cavitation intensity and consequently results in an overall improvement of the *US* efficiency. Conversely, increasing the external pressure also leads to an increase in the cavitation threshold [10]. Thereby, 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, a too high static pressure prevents bubble formations, cavitation, and then sludge ultrasonic disintegration. To sum up, as suggested by a simple analysis, an optimum pressure was expected due to opposite effects of external pressure: a reduction of the number of cavitation bubbles due to a higher acoustic cavitation threshold, but a more violent bubble collapse. The major result is that the optimum pressure seems to depend neither on the energy input, nor on the sludge type, nor on temperature that might be surprising.

Although mixed and secondary sludge led to very different DD_{COD}, the same order of sludge disintegration effectiveness was observed regardless of sludge type: (i) *US* + uncontrolled *T* + optimum pressure of 2 bar > (ii) *US* + uncontrolled *T* + atmospheric pressure > (iii) *US* + controlled *T* (28 °C) + optimum pressure of 2 bar > (iv) *US* + controlled *T* + atmospheric pressure. These conditions (ii) and (iii) showed the effect of pressure to be less than that of the temperature increase due to *US*.

The disintegration of different sludge types (mixed, secondary, and digested sludge) was investigated for a reduced *TS* of 14 g/L (as digested sludge was not available at 28 g/L), the optimum pressure of 2 bar, and both isothermal and adiabatic modes. Results are given in Fig. 8a and b. As previously found, adiabatic *US* was more efficient than isothermal *US* in terms of sludge disintegration, with an improvement of 22–82%, 39–88%, and 33–86% for mixed, secondary, and digested sludge, respectively. The results indicated the highest disintegration of secondary sludge, followed by digested sludge and mixed sludge regardless of temperature control.

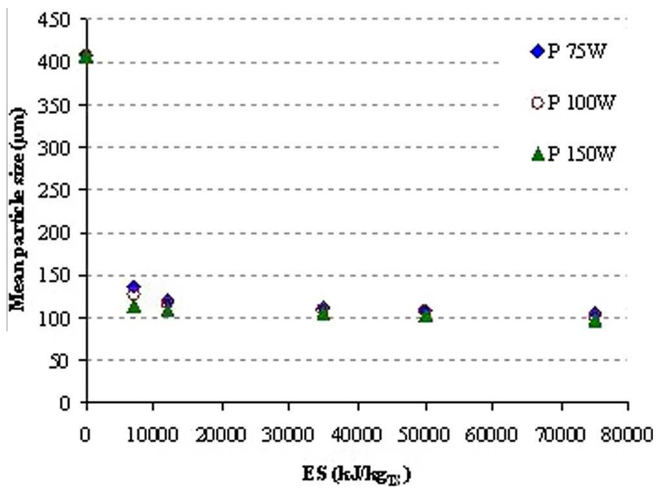

Fig. 9. Mean particle size evolution of mixed sludge (based on *D[4,3]*) during *US* pretreatment with different P_{US} values: controlled *T* (28 ± 2 °C) and atmospheric pressure.

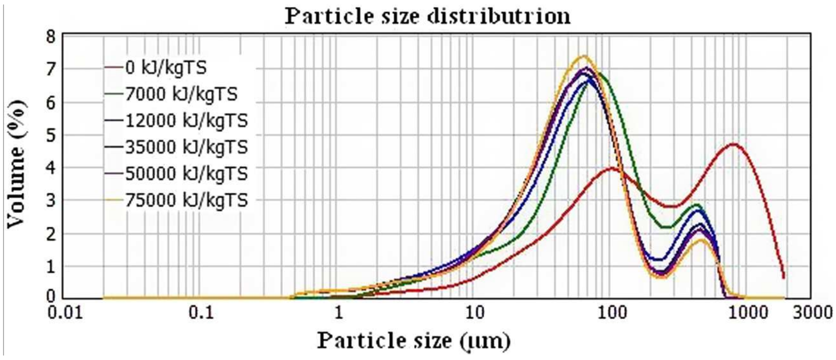

Fig. 10. Evolution of particle size distribution of mixed sludge during *US* pretreatment: P_{US} = 150 W, controlled *T* (28 °C), *TS* = 28 g/L, and atmospheric pressure.

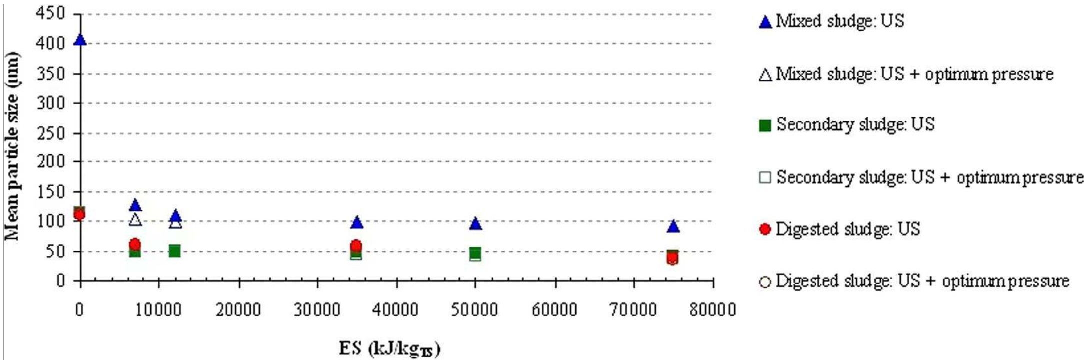

Fig. 11. Mean particle size evolution of different types of sludge during *US* pretreatment (based on *D[4,3]*): P_{US} = 150 W and controlled *T* (28 °C).

3.6. Particle size reduction

Ultrasonic pretreatment is also very effective in reducing the particle size, which is sometimes used to assess the degree of sludge disintegration and commonly analyzed by laser diffraction. The reduction in particle size should accelerate the hydrolysis stage of sludge *AD* and enhance degradation of organic matters. However, this parameter was not advised for process optimization [24].

Fig. 9 describes *D[4,3]* evolution of mixed sludge samples as a function of *ES* for the three investigated P_{US} at atmospheric pressure. Gonze et al. [25] found that particle size was decreased gradually with the increase in sonication time and a reverse trend occurred after 10 min of sonication due to the re-flocculation of the particles. However, this phenomenon was not found in this work, probably due to higher ultrasound power. In order to better understand the effect of sonication on particle charges, zeta potential measurements were performed. First, zeta potential could not be measured with the actual suspension -due to too high particle size- but only with filtered suspension (<1 µm). Sonication was shown to have only marginal effect on zeta potential: −11.3 and −13.2 mV corresponding to pretreated sludge at 7000 kJ/kg_{TS} and 50,000 kJ/kg_{TS}, respectively, as compared to that of −6.94 mV for unpretreated one. These small variations indicate a very low modification of surface charges leading to even more stability. This result is then in agreement with the absence of re-flocculation.

Compared with the untreated sludge, particle size was reduced by 68–77% following the subsequent increase in *ES* of 7000–75,000 kJ/kg_{TS}. Main reduction of *D[4,3]* was observed within a much shorter duration compared to the time required for a significant *COD* release in the aqueous phase: after 10–20 min of sonication, a quasi-plateau was reached to about 100 µm regardless of P_{US}. Other works [5,22] came to the same conclusion of a fast particle size reduction within a very short sonication time.

In the *ES* range of 7000–75,000 kJ/kg_{TS}, d_{90}, d_{50}, and d_{10} values of mixed sludge decreased by 74%, 70% and 58%, respectively. This indicated that different particle sizes had slightly different reduction extents, in which large particles were disrupted more effectively by *US* than smaller ones due to their larger surface exposed to sonication or to different consistency. This point, also illustrated in Fig. 10 showing a very fast reduction of the class of large particles (about 1000 µm), is similar to conclusions in previous works [19,21]. As shown on Fig. 10, the distribution of initial sludge was cut at 1950 µm corresponding to 99.86% of cumulative volume. Then the residual larger particles may be ignored.

When *US* experiments were conducted at optimum pressure, although the kinetics of disruption was slightly faster, the difference in final particle diameter compared to that at atmospheric pressure was negligible (Fig. 11). For instance, the enhancement of particle size reduction of mixed sludge dropped from 9.3% at 7000 kJ/kg_{TS} to less than 1% at 35,000–75,000 kJ/kg_{TS}. In accordance with Show et al. [21], the mean particle size of secondary and digested sludge was lower after *US* treatment than that of mixed sludge due to the aforementioned differences of properties.

4. Conclusions

Mixed sludge samples with different *TS* contents were pretreated with various sonication durations. An initial value of 28 g/L always yielded the highest *COD* release in the aqueous phase (DD_{COD}).

Different *US*/temperature combinations were then investigated to evaluate the effect of *US*. At any sonication time (or *US* specific energy *ES*), DD_{COD} values were the highest under adiabatic sonication, followed by those obtained by sonication under cooling, and then thermal hydrolysis ones regardless of sludge type. The effect of *US* was clearly more important than that of sole thermal hydrolysis obtained with the same temperature–time profile. The effect of external pressure on *US* sludge pretreatment was studied for the first time on mixed and secondary sludge using pressurized nitrogen in the range of 1–16 bar. At 150 W of P_{US}, DD_{COD} exhibited an optimum with respect to applied pressure at about 2 bar for all applied *ES* values. At the optimum pressure and low *ES* (7000 kJ/kg_{TS}), disintegration efficacies of secondary and mixed sludge were improved up to 37% and 67%, respectively, compared to those at atmospheric pressure.

Compared with the untreated sludge samples, mean particle size of mixed, secondary, and digested sludge was decreased by 68–77%, 55–68%, and 44–67%, respectively and particles were almost entirely disrupted in the initial period of the ultrasonic process. At 2 bar, the final size was nearly obtained at the first sampling time. The great difference in the kinetics of the two phenomena- fast size reduction and slower *COD* removal in liquid phase- should be emphasized and demonstrate that particle size is not the key parameter to follow *COD* solubilization.

All these data suggest that the best energy efficiency would correspond to short *US* exposure at the optimal pressure and under adiabatic condition. The under pressure ultrasonic pretreatment of sludge might offer a significant potential of energy savings in sludge pretreatment applications with ultrasound.

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