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Optimisation of sludge pretreatment by low frequency sonication under pressure

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A B S T R A C T

This work aims at optimizing sludge pretreatment by non-isothermal sonication, varying frequency, *US* power (P_{US}) and intensity (I_{US} varied through probe size), as well as hydrostatic pressure and operation mode (continuous *vs*. sequential – or pulsed – process).

Under non isothermal sonication sludge solubilization results from both ultrasound disintegration and thermal hydrolysis which are conversely depending on temperature. As found in isothermal operation:

- For a given specific energy input, higher sludge disintegration is still achieved at higher PUS and lower sonication time.
- US effects can be highly improved by applying a convenient pressure.
- 12 kHz always performs better than 20 kHz.

Nevertheless the optimum pressure depends not only on P_{US} and I_{US}, but also on temperature evolution during sonication.

Under adiabatic mode, a sequential sonication using 5 min *US*-on at 360 W, 12 kHz, and 3.25 bar and 30 min *US*-off gives the best sludge disintegration, while maintaining temperature in a convenient range to prevent *US* damping.

Keywords:
Audible frequency
Hydrostatic pressure
Sequential process
Pulsed ultrasound
Sludge disintegration

1. Introduction

Wastewater treatment plants (WWTP) commonly involve activated sludge and a large amount of excess bacterial biomass remains at the end of the process. After use, sewage sludge is usually landfilled, used for land fertilization or incinerated, but these disposal methods involve high energy consumption and may have adverse effects on health and environment. A sustainable solution for sludge management is anaerobic digestion (*AD*) resulting in biogas production. However, hydrolysis step is rate-limiting and sludge pretreatment is needed to break the cells wall and improve its biodegradability.

Apart from some popular techniques used in sludge processing, *e.g.* thermal, chemical or other mechanical methods, *ultrasound (US)* has gained interest for such purpose, as it provides efficient sludge disintegration (Pilli et al., 2011; Tyagi et al., 2014) and does not require any chemical additive. Ultrasonic pretreatment was reported to improve biodegradability and bio-solid quality (Khanal et al., 2007; Trzcinski et al., 2015), to enhance biogas/methane production (Barber, 2005; Braguglia et al., 2015; Khanal et al., 2007; Onyeche et al., 2002), to reduce excess sludge (Onyeche et al., 2002) and required sludge retention time (Tiehm et al., 1997).

Operating conditions of sonication can significantly affect the cavitation intensity and consequently the rate and/or yield of the *US*-assisted operation. Ultrasound efficiency is indeed influenced by many factors: *US* parameters (related to **frequency** F_S, **power** P_{US} and **intensity** I_{US}), presence of dissolved gas and particles, nature of the solvent (volatility), configuration of the acoustic field (standing or progressive wave), **temperature** (damping), **hydrostatic pressure** (P_h), *etc.* (Lorimer and Mason, 1987; Pilli et al., 2011; Thompson and Doraiswamy, 1999).

As regards *US*-assisted sludge pretreatment, specific energy input (*ES*) is recognized as the key parameter, but others have proved to have significant effects at given *ES* value, *e.g.* P_{US}, I_{US}, (Li et al., 2010; Liu et al., 2009; Show et al., 2007; Wang et al., 2005; Zhang et al., 2008b) and F_S (Tiehm et al. 2001; Zhang et al. 2008a). Previous investigations also indicated sonication without cooling (referred as “adiabatic” sonication although heat losses) to be much better than isothermal treatment thanks to the combined

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effects of cavitation and temperature rise due to ultrasound energy dissipated into the sludge (Chu et al. 2001; Kidak et al. 2009; Le et al., 2013a; Huan et al. 2009). In order **to better elucidate ultrasound effects** – *i.e.* without thermal interactions, our group first applied **isothermal conditions** thanks to an external cooling and highlighted the positive effect of audible frequency (12 *vs.* 20 kHz), the importance of hydrostatic pressure, and the separate roles of power density and power intensity (Delmas et al., 2015; Le et al. 2013a). 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. For a given equipment operating at the 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 of "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: with same conditions 12 kHz outperformed 20 kHz in any case. These results are of major interest for general sonochemistry, but they are probably not obtained at optimum temperature as sludge disintegration is known to be thermally activated. Thus in the **practical case – of non-isothermal ultrasonic sludge disintegration** – heat release would have a positive additional effect, but limited to some degree as conversely cavitation effects would decrease.

This work thus aims at optimizing sonication process for non-isothermal sludge disintegration by simultaneous investigation of the significant parameters, *i.e.* P_{US}, I_{US} (varied both through P_{US} and emitter surface), F_S (20 and 12 kHz) and P_h. Without any cooling but heat losses, temperature rise might be controlled – and possibly optimized through the operation mode (continuous *vs.* sequential – or pulsed – sonication).

2. Materials and methods

2.1. Sludge samples

Waste activated sludge (*WAS*) was collected from a French wastewater treatment plant. Standard analytical methods (see § 2.2) were used to evaluate its properties gathered in Table 1. Note that sludge sampling was performed at different periods in relation with the changes in *US* equipment along this work. Synthetic *WAS* samples labeled "a" and "b" in Table 1 were used for investigating the efficiency of "adiabatic" sonication under pressure (varying P_{US} and probe size) and for optimizing the *US*-assisted process (continuous *vs.* sequential treatment), respectively.

Table 1
Properties of the sludge samples (a and b).

Parameter		Sample	
		a	b
Raw sludge sample			
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
Synthetic sludge sample			
Total solids (*TS*)	g/L	28.0	28.0
Mean $SCOD_0$	g/L	2.8	4.1
$SCOD_{NaOH0.5M}$	g/L	22.7	22.1
TCOD	g/L	36.3	39.1
$SCOD_{NaOH}/TCOD$	%	62.5	56.5

Sludge was sampled in 1 L and 100 mL boxes and frozen. As mentioned in previous studies (Kidak et al., 2009; Le et al., 2013b), it was verified that this conditioning method did not significantly affect *COD* solubilization results (variation less than 8%).

Synthetic samples were prepared by diluting defrosted raw sludge with distilled water up to a total solid concentration of 28 g/L – an optimum value for *US* sludge disintegration according to our previous work (Le et al., 2013a).

2.2. Analytical methods

Standard Methods (APHA, 2005) were applied to measure ***total*** and ***volatile solid*** (*TS* and *VS*) contents. *TS* content was obtained by drying the sludge sample to a constant mass at 105 °C. Then the residue was ignited at 550 °C and *VS* content was calculated from the resulting weight loss.

In order to get normalized data the ***degree of sludge disintegration*** (DD_{COD}) was calculated by measuring the chemical oxygen demand in the supernatant (*SCOD*) before and after treatment. *SCOD* was measured by Hach spectrophotometric method after preliminary vacuum filtration using a cellulose nitrate membrane with 0.2 μm pore size. Following Schmitz et al. (2000), DD_{COD} was given as the ratio between the soluble COD increase during sonication and that resulting from a strong alkaline disintegration of sludge (0.5 M NaOH for 24 h at room temperature (Huan et al., 2009)):

$$DD_{COD} = (SCOD - SCOD_0)/(SCOD_{NaOH} - SCOD_0)*100(\%) \quad (1)$$

Besides, potassium dichromate oxidation method (standard AFNOR NFT 90–101) was used to measure the total chemical oxygen demand (*TCOD*).

The ***particle size distribution (PSD)*** of sludge before and after treatment was measured by laser diffraction on a Mastersizer 2000 (Malvern Inc.). After dilution in osmosed water (300 fold), the suspension was pumped into the measurement cell (suction mode). As found in previous studies (Bieganowski et al., 2012; Minervini, 2008), the refractive index and absorption coefficient were set to 1.52 and 0.1, respectively (default optical properties). Moreover it was checked that these mean optical properties led to a weighted residual parameter of less than 2% as recommended by the manufacturer. An average of five consecutive measurements (showing less than 3% deviation) was made and the volume mean diameter *D[4,3]* (or de Brouckere mean diameter) was calculated.

2.3. US equipment and experimental procedure

The experimental set-up (see Fig. S1 in Supplementary Materials) used a cup-horn sonicator included in an autoclave reactor (internal diameter of 9 cm and depth of 18 cm, for a usable capacity of 1 L). The stainless steel reactor was connected to a pressurized N_2 bottle and a safety valve (HOKE 6500) limited overpressure to 19 bar.

To achieve experiments at a selected temperature, the reactor was cooled by circulating fresh water stream (15 °C) in an internal coil. It could be also heated by two 500 W annular heaters whose power can be adjusted thanks to a PID controller. The suspension was stirred by a Rushton type turbine of 32 mm diameter. According to our previous work (Le et al., 2013a), its speed was set to 500 rpm to prevent centrifugation of the particles. The same synthetic sludge volume (V = 0.5 L) was used for each experiment.

The equipment included two generators working at 12 and 20 kHz, and for each two different probes of 13 and 35 mm diameter, labeled as *SP* and *BP*, respectively. Maximum P_{US} (transferred from the generator to the transducer) was 100 W and 400 W for *SP* and *BP*, respectively. During operation, the transducer was cooled by compressed air.

For a given set of operating conditions, different sonication times (t), corresponding to four values of *ES* (7000, 12,000, 35,000, and 50,000 kJ/kg_{TS}), were usually applied, where:

$$ES = (P_{US}*t)/(V*TS) \quad (2)$$

First, the effect of temperature on sludge disintegration (DD_{COD}) was investigated for both isothermal and "adiabatic" sonication under standard conditions — 20 kHz, atmospheric pressure. Then the influence of *US* parameters and hydrostatic pressure was evaluated under non-isothermal conditions. Finally, a pulsed-mode procedure was applied to further optimize the *US*-assisted process. In some cases, experiments were duplicated and the coefficients of variation of DD_{COD} were about 5%.

3. Results and discussion

3.1. Temperature effect

Two different effects result from the ultrasonic pretreatment: extreme macro and micro mixing due to cavitation and increase in the bulk temperature. To evaluate the contribution of each on sludge disintegration, different tests were applied: (1) sonication (150 W, BP) under isothermal conditions (cooling at 28 ± 2 °C), (2) "adiabatic" sonication (*i.e.* same conditions, but without any cooling), (3) thermal hydrolysis: without *US* and with a progressive increase as recorded in (2), and (4) 5 min of *US* and progressive temperature increase afterwards.

Results are presented in Fig. 1. Based on DD_{COD} values, treatment efficiency could be ranked as follows: (2) ("adiabatic" sonication) > (4) (short sonication time and thermal hydrolysis) > (1) (low temperature sonication) ~ (3) (thermal hydrolysis only). DD_{COD} values of sonicated samples under adiabatic conditions were about twice those obtained under cooling (28 °C). Note that in any case after 5 min of *US* at 150W-*BP*, sludge particles were almost disrupted: *D[4,3]* was about 110 μm as compared to 380 μm of raw sludge, proving particle size not to be the convenient quantity for sludge treatment.

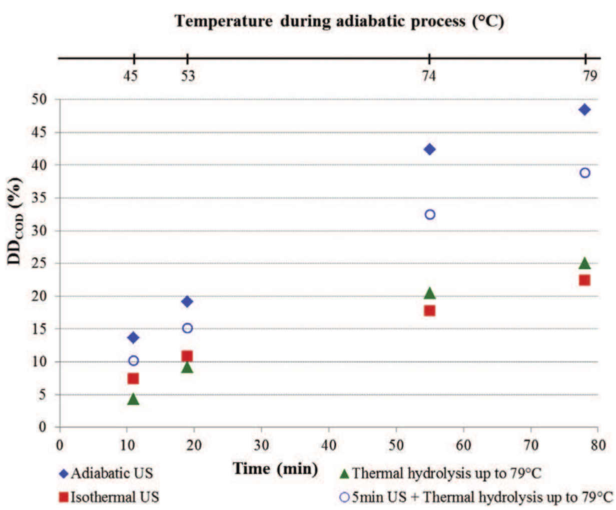


Fig. 1. Effect of temperature profile* on time-evolution of DD_{COD} under sonication (F_S = 20 kHz, P_{US} = 150 W, *BP*, *WAS* "a" from Table 1, and atmospheric pressure) and/or thermal hydrolysis. **The upper x-axis indicates the evolution of temperature during adiabatic US and thermal hydrolysis.*

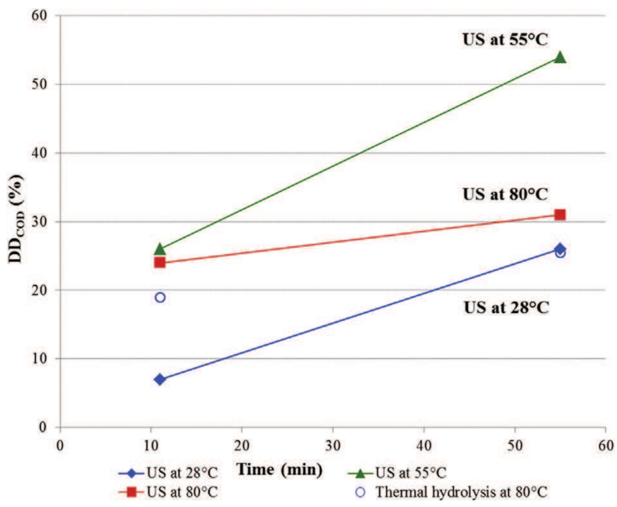


Fig. 2. Effect of temperature on sludge disintegration by isothermal sonication (F_S = 20 kHz, P_{US} = 150 W, *BP*, *WAS* "b" from Table 1, and atmospheric pressure); comparison to thermal hydrolysis.

The main information brought by these experiments is: first, cavitation and thermal hydrolysis seem to show almost additional effects during adiabatic sonication; second, thermal hydrolysis of early disrupted sludge is faster than that of raw sludge. Therefore the combined effect is actually more complex: cavitation acts mainly during the early stage of the adiabatic sonication, then *US* being progressively damped by the increasing temperature, thermal hydrolysis takes over, being "boosted" by the initial work of *US*. The resulting positive effect of combining *US* and temperature rise for sludge disintegration is in agreement with the conclusion of earlier works (Chu et al., 2001; Kidak et al., 2009; Huan et al., 2009), but opposite to most power *US* applications in which temperature only damps cavitation.

To further understand the effect of temperature on cavitation efficiency, additional experiments were conducted on *WAS* "b" presented in Table 1, under a constant temperature of 28, 55 or 80 °C. Results, given in Fig. 2, show an increase in DD_{COD} when increasing *T* from 28 to 55 °C, but a decrease at 80 °C. It is well known that at high temperature cavitation bubbles accumulate water vapor during the growth phase at low acoustic pressure, which will cushion bubble collapse and make it much less violent. Moreover, there was only small differences in DD_{COD} between isothermal *US* and sole thermal hydrolysis at the same *T* of 80 °C. It is then clear that cavitation intensity is severely dampened at high temperature.

3.2. Effect of US parameters on non-isothermal sonication at atmospheric pressure

The effect of P_{US} on DD_{COD} under non-isothermal sonication was investigated using the following ranges: 50–100 W for *SP* and 50–360 W for *BP*. Experiments were conducted at 20 kHz under atmospheric pressure and using *WAS* "a" from Table 1. Results are reported in Fig. 3.

As expected, the evolution of sludge temperature was found to depend on P_{US}: higher P_{US} resulted in a faster temperature increase and yielded a higher final value at given *ES* as the reactor was not fully insulated. In addition, and more surprisingly, different temperature profiles were also observed with same P_{US} but different probe sizes: at 50 W, final *T* increased from 40 °C to 46 °C when switching from *SP* to *BP*. This unexpected result means that the efficiency of *US* transmission to the sludge is significantly better

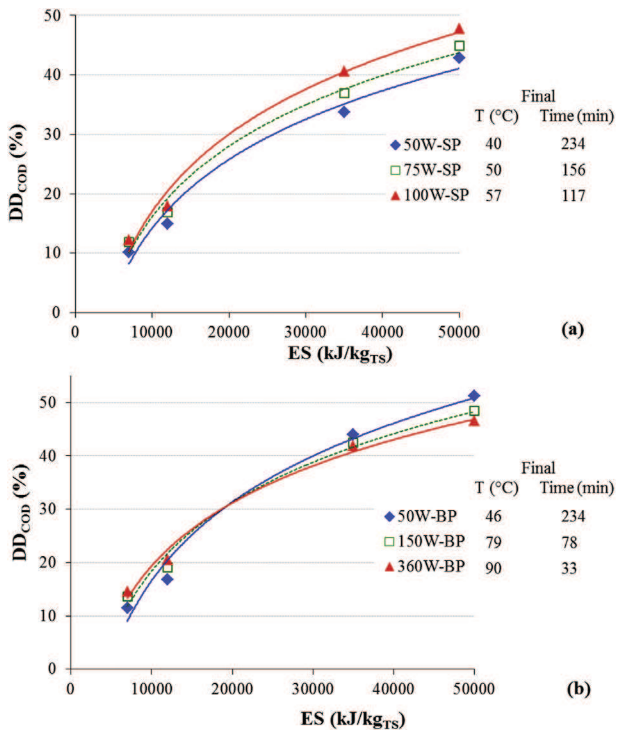


Fig. 3. Effect of *ES* and P_{US} on DD_{COD} under "adiabatic" sonication ($F_S = 20$ kHz, *WAS* "a" from Table 1, and atmospheric pressure): (a) *SP* and (b) *BP. Final temperatures of adiabatic US are also given.*

with the big probe than with the small one, maybe due to limited wave propagation under intense cavitation.

Fig. 3a, corresponding to the **small probe**, proves that *high* P_{US} *− short time* is the most effective for *US* sludge pretreatment at atmospheric pressure as found in isothermal condition at 28 °C (Delmas et al., 2015). Nevertheless, the positive effect of P_{US} in adiabatic mode was not better than in isothermal mode: for instance, at *ES* of 50,000 kJ/kg$_{TS}$, DD_{COD} increased by 12% from 50 to 100 W as compared to 13% for sonication at 28 °C (Delmas et al., 2015). That means there was no positive effect of the slight temperature gain at 100 W as compared to 50 W (up to 17 °C) despite the temperature level reached was still moderate.

Conversely, the 50 W-sonication could have benefit from the temperature increase when switching from small to **big probe**, as in the latter case higher DD_{COD} was reached despite lower I_{US} (Fig. 3b). With *BP*, high power was only efficient in adiabatic conditions for *ES* lower than 20,000 kJ/kg$_{TS}$ (when the increase in sludge temperature and *US* duration were still small). The apparently surprising reverse trend at higher *ES*, then higher *t*, might be explained by a lower *US* efficiency at higher temperature. So in this high range of *ES*, the beneficial effect of temperature through thermal hydrolysis should be overpassed by its detrimental effect on cavitation efficiency (as yet suggested on Fig. 2).

However, it should be mentioned that the results in Fig. 3 were achieved on samples rapidly cooled at the end of sonication. In this case, the beneficial effect of thermal hydrolysis (a slow process) could not be fully recovered during the shortest treatments, *e.g.* 33 min for 360 W and 78 min for 150 W, as compared to 4 h for 50 W (Fig. 3b). Another comparison could then be made based on the same treatment period, including sonication plus maturation under stirring only ("thermal hydrolysis" after *US*). Thereby, additional experiments were conducted using *BP* at both same *ES* and treatment time. At 50 W, sonication was applied in the *ES* range of 7000–50,000 kJ/kg$_{TS}$ and the suspensions were then cooled down immediately to 28 °C. At 150 W and 360 W, *US* was turned off after same *ES* values were reached, but the stirrer was still working (without cooling) until the whole durations equaled those of 50 W experiments. Results of DD_{COD}, given in Fig. 4, show again the *high* P_{US} *− short time* sonication to be the best mode for sludge disintegration at atmospheric pressure, thanks to thermal hydrolysis after *US* disintegration. Nevertheless only very slight difference was observed between 150 and 360 W due to reduced cavitation effects at high temperature. Temperature evolutions (due to heat losses) corresponding to experiments at 50,000 kJ/kg$_{TS}$ are depicted in Supplementary Materials (Fig. S2). Of course, one may suggest that thermal insulation of our equipment would provide even better results by keeping higher temperature after sonication. Note that such energy saving by insulating the reactor could also save *US* energy for the same result in terms of DD_{COD}.

To sum up, the effect of heat released by sonication is rather complex and cannot be neglected. Besides, at atmospheric pressure, sludge disintegration still benefits from high P_{US} if enough time is let for thermal hydrolysis induced by *US* heating to operate.

3.3. Effect of US parameters on the optimum pressure and subsequent DD_{COD}

Optimum pressures under adiabatic *US* were searched in the 1–5 bar range at a given *ES* value, but for different P_{US} (100–360 W) and probe sizes using *WAS* "a" from Table 1. Results are shown in Fig. 5 where same *ES* (50,000 kg/kg$_{TS}$) but different total treatment durations were applied (contrary to recommendations from previous section). This should however not much change the location of the optimum pressure, but only the final corresponding DD_{COD} value.

Under isothermal sonication at 28 °C (Delmas et al., 2015), the optimum pressure was found to shift toward higher pressures when increasing P_{US} (and thus I_{US} proportionally):

- 1 bar (or even lower) at 50 W, 2 bar at 150 W and 3.5 bar at 360 W for *BP*,
- 1.5 bar at 50 W and 2.5 bar at 100 W for *SP*.

Surprisingly, under temperature rise as in the present work, the same optimum pressure of 2 bar was obtained with the same probe (*BP*) at different P_{US} (150 and 360 W) while an increase would be expected at higher power according to isothermal data. The respective evolution of optimal pressure *vs.* P_{US} is more complex in non-isothermal conditions, due once again to the result of opposite effects of temperature on cavitation intensity and thermal hydrolysis: the optimal pressure values found at 28 °C slightly increase at

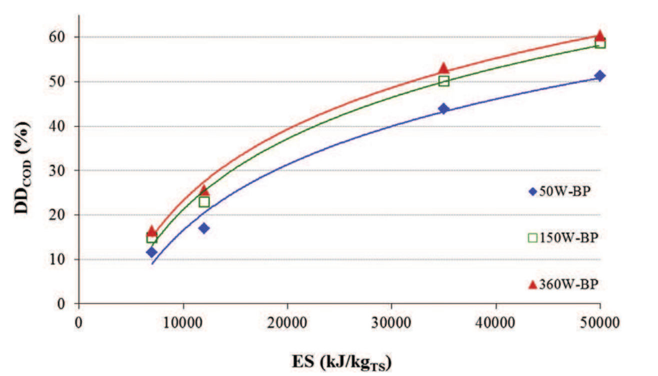


Fig. 4. Effect of *ES* and P_{US} on DD_{COD} under "adiabatic" sonication followed by stirring up to 240 min ($F_S = 20$ kHz, *WAS* "a" from Table 1, atmospheric pressure).

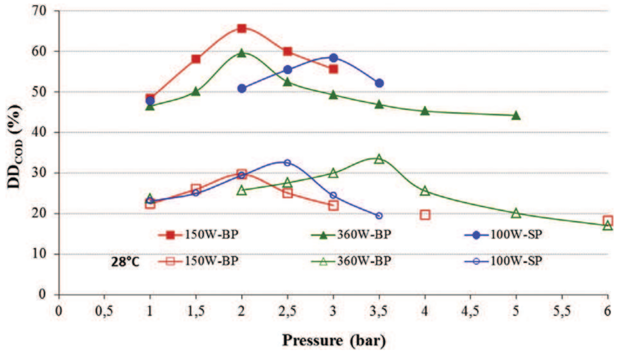


Fig. 5. Comparison of pressure effects on DD_{COD} under adiabatic and isothermal (28 °C) sonication for different combinations of P_{US}-probe sizes (F_S = 20 kHz, ES = 50,000 kJ/kg$_{TS}$, *WAS* "a" from Table 1).

the moderate temperatures resulting from sonication at 100 W with *SP* when no cooling is applied (from 2.5 bar to 3 bar -Fig. 5), but they decrease at the extreme temperatures found at 360 W with *BP* (from 3.5 bar to 2 bar -Fig. 5). This unexpected result (due to the negative effect of very high *T*) would deserve more analysis based on single cavitation bubble dynamics at high temperature and high pressure. It should be additionally noticed that the optimum is less marked in "adiabatic" conditions where only a part of DD_{COD} is due to acoustic cavitation, the other part being due to temperature rise and not dependent on the hydrostatic pressure.

In short, sonication effect can be improved by applying a convenient pressure and this optimum is due to opposite effects of hydrostatic pressure. At high external pressure, the increase of the cavitation threshold reduces the number of cavitation bubbles but their collapse is more violent (Lorimer and Mason, 1987). Associated with our previous work under isothermal sonication, it can be concluded that location of the optimum pressure is dependent on P_{US}, I_{US}, as well as on temperature.

3.4. Optimization of sludge sonication pretreatment

High P_{US}-short time, low F_S (12 kHz according to our previous work, Delmas et al., 2015), and adiabatic conditions should be preferred to improve *US* disintegration of sludge. Moreover, the optimum pressure was found to depend on *US* parameters and thermal effects induced by high power ultrasound. Then this section is devoted to finalizing optimization of *US* sludge disintegration by searching for the optimum pressure, while setting the other parameters at the most favorable conditions expected (*i.e.* 12 kHz, *BP* working at 360 W, and adiabatic conditions) using *WAS* "b" from Table 1.

It can be also noted that sonication at high P_{US} resulted in too high sludge temperature, more than 80 °C, while the safety range recommended by the manufacturer is less than 65 °C for the 12 kHz device. Extreme temperatures might harm the transducer, lead to unstable P_{US}, and are not convenient to provide intense cavitation. In fact, several runs were interrupted due to the high temperature. Sequential (or pulsed) sonication was therefore investigated to limit the temperature increase and possibly improve the process. The comparison of continuous and sequential modes contributes to the optimization of sludge *US* pretreatment.

Fig. 6a compares continuous *vs*. sequential *US* sludge disintegration using same *ES* value of 35,000 kJ/kg$_{TS}$ and varying pressure within 1–3.25 bar, as the optimum was expected in this range (cf. § 3.3, 3.25 bar being the value found for isothermal sonication (28 °C) at 12 kHz and 360 W with *BP*). Besides, 35,000 kJ/kg$_{TS}$ was chosen to have a relatively short treatment time in the most severe conditions (continuous sonication at 360 W), not to harm the transducer (by limiting temperature rise).

The following conditions were investigated:

(i) 50 W continuous sonication at 1 bar (164 min)
(ii) 360 W continuous sonication at 1, 2, and 3.25 bar (23 min)
(iii) 23 min of 360 W continuous sonication, as in (*ii*), but followed by stirring (no *US*) up to 164 min, to get the same treatment time as in (*i*) (marked as *360W-'xx' bar + stirring*) and let thermal hydrolysis operate after the temperature rise due to sonication
(iv) Sequence made of 1 min *US* at 360 W followed by 6 min stirring (no *US*) and pursued for a total duration of 164 min (marked as *360W-1/6-'xx' bar*)
(v) Sequence made of 5 min *US* at 360W followed by 30 min stirring (no *US*) and pursued up to 164 min of treatment (marked as *360W-5/30-'xx' bar*).

Two *US* pulses of 1 min and 5 min were selected in order to vary the temperature fluctuations around the smooth continuous temperature profile (at 50 W). Temperature profiles during sequential sonication are given in Fig. 6b.

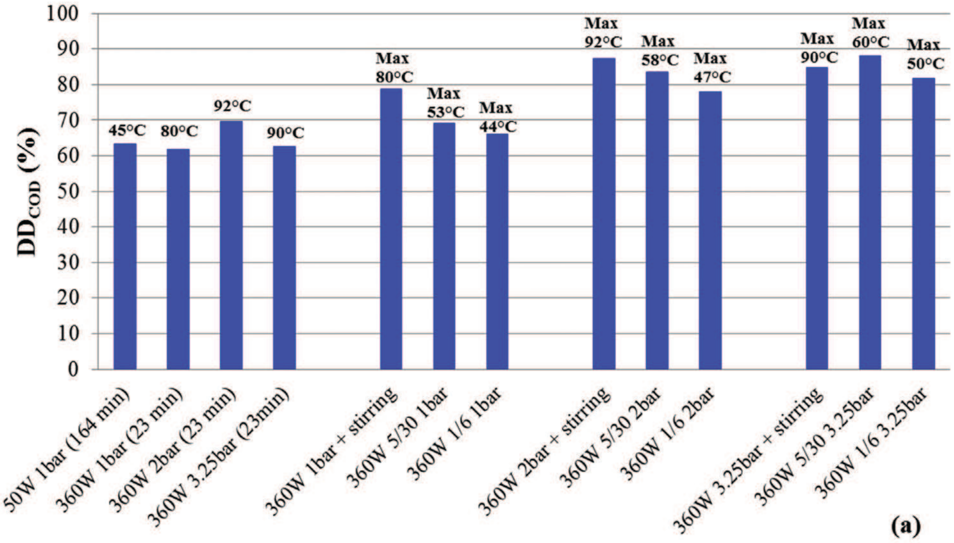
For the continuous "adiabatic" process, sonication at 360 W under 2 bar was found as the best condition regardless of the total treatment time. It is interesting to note that the final temperature under 360 W *US* increased from 80 °C to 99 °C with increasing pressure from 1 to 3.25 bar, proving a better energy transmission at high pressure. Nevertheless this better transmission does not mean better efficiency for sludge disintegration: as yet mentioned, too high temperature is very detrimental for cavitation intensity, due to the less violent collapse of cavitation bubbles containing too much vapor. The 360 W runs including a consecutive maturation period up to 164 min (mentioned as "+ stirring" in Fig. 6a) showed much better disintegration than those cooled just after sonication, thanks to thermal hydrolysis, and resulted in closer DD_{COD} values at 2 and 3.25 bar, clearly higher than that at 1 bar. The benefit as compared to the 50 W operation was only significant if the whole treatment period was indeed kept unchanged. However, temperature at the end of the 360 W continuous sonication was too high (both for equipment safety and cavitation efficiency). Then its disadvantages as abovementioned could be avoided by a sequential *US* application mode.

For the sequential mode, 360 W sonication at 3.25 bar was the most efficient, followed by that at 2 bar, then 1 bar. The pressure of 2 bar was no longer an optimum in the sequential process which provided a very similar temperature profile at 2 and 3.25 bar. Besides, the advantage of the 35 min period cycle (5/30) as compared to 7 min period cycle (1/6) at all applied pressures might be again due to temperature effect: the maximum sludge temperatures during 5/30 mode were indeed higher than those during 1/6 mode (see Fig. 6b). At the same *ES* value of 35,000 kJ/kg$_{TS}$ and same treatment time of 164 min, DD_{COD} resulting from the "optimal" sequential process was about 40% higher than that from 50 W continuous sonication. However, this sequential mode did not perform much better than the continuous operation at 360 W, while yielding more reasonable temperatures.

In short, sequential sonication at 12 kHz and under 3.25 bar – with 5 min of adiabatic sonication at 360 W and 30 min of stirring – appears as the best combination to achieve a high sludge disintegration degree with the advantage of maintaining temperature in the recommended range.

4. Conclusions

This work shows how non-isothermal ultrasonic sludge disintegration may be improved by lowering frequency (under audible



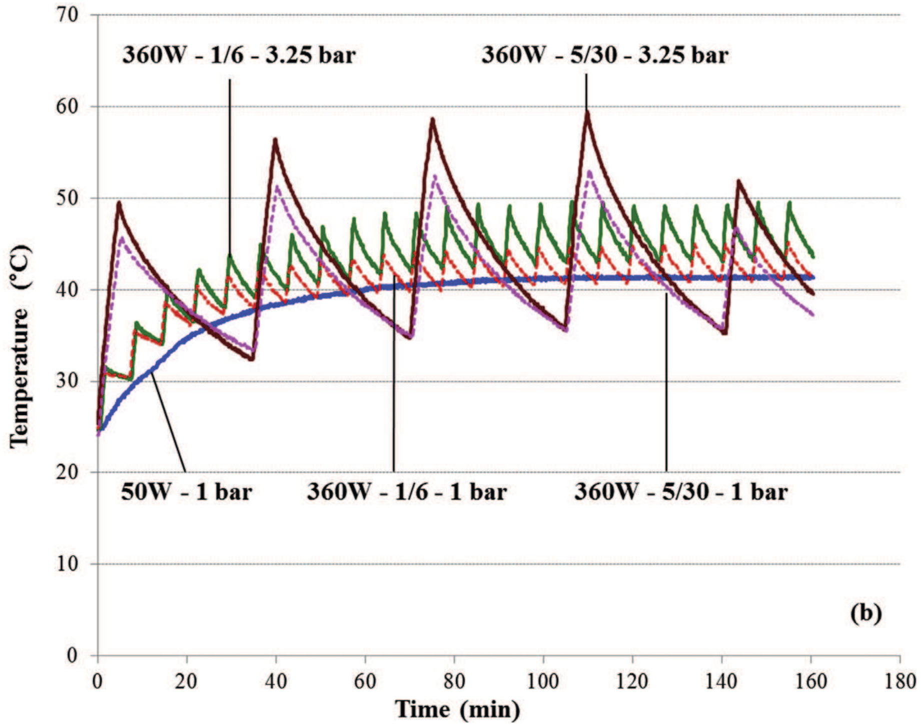


Fig. 6. Continuous and sequential *US* sludge disintegration at different pressures under adiabatic conditions (a) DD_{COD} and (b) temperature profiles (*BP*, ES = 35,000 kJ/kg$_{TS}$, F_S = 12 kHz, *WAS* "b" from Table 1).

threshold), increasing power while decreasing sonication time, finding the optimal pressure, and using sequential mode.

First, the effect of temperature increase due to sonication without cooling could not be neglected both during and after the process, accounting for resulting thermal hydrolysis of sludge is rather slow at moderate temperature. As a result, at a given specific energy, more efficient sludge disintegration was still achieved when applying higher power if same total time was kept. This temperature evolution also affected the optimum value of pressure to be applied for sonication enhancement, which differed from that observed during isothermal operation. Concerning disintegration, a slight improvement was obtained at moderate temperature, mainly due to conjugate effects of higher number of cavitation bubbles and thermal hydrolysis, but a decrease at extreme temperatures (>80 °C) due to the less violent collapse of cavitation bubbles containing too much vapor. Due to combined cavitation and thermal effects, the optimum temperature should be higher than in most other *US* applications.

Then, a sequential operation using 5 min *US*-on at 360 W, 12 kHz, and 3.25 bar and 30 min *US*-off showed the best efficiency of sludge disintegration and the advantage of maintaining temperature in the recommended safety range. In a large continuous equipment with a convenient thermal insulation, same optimum temperature would be achieved with much less *US* energy consumption increasing the economic viability of this process.

It is clear that 12 kHz – much more efficient than 20 kHz – is probably not the optimal frequency and additional work would be deserved. This improvement at low frequency would probably be observed on many other applications of physical effects of power ultrasound. Nevertheless equipment is not directly available and should be designed specifically.

Finally these optimal conditions should be used in future experiments on methane production to quantify the positive effect of sonication on both yield and kinetics.

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Appendix A. Supplementary data

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