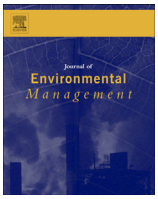
Journal of Environmental Management 165 (2016) 206–212

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman



Research article

Optimisation of sludge pretreatment by low frequency sonication under pressure

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ARTICLE INFO

Article history:
Received 28 May 2015
Received in revised form
9 September 2015
Accepted 11 September 2015
Available online 2 October 2015

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

ABSTRACT

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.

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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http://dx.doi.org/10.1016/j.jenvman.2015.09.015

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