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Solubilisation of waste-activated sludge by ultrasonic treatment

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Abstract

In order to enhance the efficiency of anaerobic digestion, the effects of ultrasonic pretreatment have been studied on waste-activated sludge. Solubilisation of chemical oxygen demand (COD), solid and nitrogen has been proved. Flocs were broken and compounds were made soluble. In the same time, particle size decreased with specific energy applied. In terms of biodegradability, ultrasound led to an increase in biogas production. Moreover, the relationship between biogas production and sludge fractions has been examined. For specific energy input lower than 3000 kJ/kg of total solids, biogas production linked to the particulate fraction of sludge was constant, even if the solids concentration decreased. On the other hand, biogas production linked to the soluble part of sludge increased with ultrasonic power.

Keywords: Ultrasound; Biodegradability; Particle size; Sewage sludge; Anaerobic digestion

1. Introduction

In response to the enforcement of the European legislation regarding the Urban Wastewater Treatments Directive (91/271/EEC), the research of more efficient treatments for wastewater is being increased. The Directive sets limits for some of the established sanitary determinants e.g. biological oxygen demand (BOD), suspended solids and the nutrients, nitrogen and phosphorus. This leads to an increase in sewage sludge production and causes a large problem to communities and wastewater treatment plants operators. They need to eliminate more sludge, whereas disposal routes (incineration, land disposal and land application) are subject to more social constraints. Anaerobic digestion, which allows a reduction of sludge quantity, has become one common method of sludge stabilisation, due to the production of biogas that makes the process profitable. In wastewater treatment plants, anaerobic digestion is generally applied to mixture of primary and secondary (waste-activated) sludge. But waste-activated sludge (WAS) is known to be more difficult to digest than primary sludge [1]. Anaerobic digestion process is achieved through several stages: hydrolysis, acidogenesis, methanogenesis. For WAS degradation, the rate-limiting stage is the hydrolysis [2]. In order to improve hydrolysis and anaerobic digestion performance, several pretreatments can be considered: mechanical, thermal, chemical or biological treatments. These pretreatments cause the lysis or disintegration of sludge cells [3,4]. Intracellular matter is released and becomes more accessible by anaerobic microorganisms. According to Wang et al. [5], ultrasonic lysis is the most interesting process for enhancement of methane generation. Retention time in the digester can also be reduced [6], which allows more compact plants.

Ultrasonic process leads to cavitation bubble formation in the liquid phase [7]. These bubbles grow and then violently collapse when they reach a critical size. Cavitational collapse produces intense local heating and high pressure on liquid–gas interface, turbulence and high shearing phenomena in the liquid phase. Because of the extreme local conditions, $OH^\bullet$, $HO_2^\bullet$, $H^\bullet$ radicals and hydrogen peroxide can be formed. Thus, sonication is a combination of different phenomena: chemical reactions using radicals, pyrolysis, combustion and shearing [8,9].

Mechanisms of the ultrasonic process are influenced by three factors: supplied energy, ultrasonic frequency and na-

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Nomenclature

BOD	biological oxygen demand (mg O_2/l)
COD	chemical oxygen demand (mg O_2/l)
$\mathrm{COD}_{\mathrm{NaOH}}$	soluble COD after an alkaline hydrolysis (mg O_2/l)
CODp	COD in particles (mg O_2/l)
CODp_0	initial COD in particles (mg O_2/l)
CODs	soluble COD (mg O_2/l)
CODs_0	initial soluble COD (mg O_2/l)
d_{50}	cut diameter (μm)
dS	Sauter mean diameter (μm)
$\mathrm{DD}_{\mathrm{COD}}$	degree of disintegration of COD (%)
E_s	specific supplied energy (kJ/kg TS)
MS	mineral solids (g/l)
MS,s	mineral solids in the soluble fraction (g/l)
$\mathrm{N\text{-}NH_4}^+$	concentration of ammonium (mg N/l)
OSMd	quantity of organic soluble matter to degrade (mg)
P	power (W)
S_{COD}	COD solubilisation (%)
S_{TS}	total solid solubilisation (%)
t	sonication time (s)
TKN	total Kjeldahl nitrogen (mg N/l)
TKNs	total nitrogen in the soluble fraction (mg N/l)
TS	total solids (g/l)
TS,s	total solids in the soluble fraction (g/l)
TS_0	initial total solids (g/l)
TSS	total suspended solids (g/l)
v	sample volume (l)
vbs	volume of biogas produced by the soluble fraction (ml)
VS	volatile or organic solids (g/l)
VS,s	volatile or organic solids in the soluble fraction (g/l)
VSS	volatile or organic suspended solids (g/l)
WAS	waste-activated sludge

ture of the influent. Cell disintegration is proportional to supplied energy [9,10]. High frequencies promote oxidation by radicals, whereas low frequencies promote mechanical and physical phenomena like pressure waves [11]. With complex influents, radical performance decreases. It has been shown that degradation of excess sludge is more efficient using low frequencies [12].

The objective of this study was to understand waste-activated sludge modification due to an ultrasonic process and its effects on sludge anaerobic biodegradability. This biodegradability was estimated without presence of oxygen. Solubilisation of chemical oxygen demand (COD), matter and nitrogen was measured and improvement of biogas production during batch anaerobic digestion was linked to this solubilisation.

2. Experimental

2.1. Sludge

Flotation-thickened WAS was collected from the municipal wastewater treatment plant of Carcassonne (South of France). This plant has a capacity of 90,000 people equivalent, that is to say that the pollution fed to the plant was equivalent to 4.5 tons of suspended solids per day. This plant-treated domestic and industrial wastewater was operated with a high-loaded aeration tank. A wastewater treatment plant produces purated water and sludge. Sludge is a suspension of complex compounds in water. For this paper, the term of “matter” or “solids” will be used in order to name these complex compounds. For the experiments, sludge was diluted to about 18.5 g/l of total solid (TS). The total volatile or organic solid (VS) content was 81% of TS.

2.2. Ultrasonic treatment

The ultrasonic apparatus used was an ultrasonic homogenizer Autotune 750 W (Bioblock Scientific). This apparatus was equipped with a probe and worked with an operating frequency of 20 kHz and a supplied power of about 225 W. Batch experiments were carried out in beakers without temperature regulation (no cooling). Treated samples had a volume of 0.5 l. Specific supplied energy ranged from 0 to 15,000 kJ/kg TS. Specific supplied energy has been chosen in order to compare results. Specific energy (E_s) is defined using ultrasonic power (P), ultrasonic time (t), sample volume (v) and initial total solid concentration (TS_0):

$$E_s = (Pt)/(v\mathrm{TS}_0) \tag{1}$$

2.3. Analysis

2.3.1. Chemical oxygen demand (COD)

COD measurement allows pollution concentration to be expressed as the amount of oxygen (O_2) consumed to oxidise matter. COD was measured in the total sludge, in supernatant and in the particulate fraction using the normalised method [13]. To separate supernatant and particulate mater, centrifugation was carried out using a Beckman J2 MC centrifuge [25,000 × g; 15 min; 5 °C]. For this paper, COD measured on the supernatant liquor will be called “soluble COD” and COD measured on the “solids of centrifuge” will be called “particulate COD”. “COD solubilisation” represents in fact the transfer of COD from the particulate fraction of the sludge (solids after centrifugation) to the soluble fraction of the sludge (supernatant after centrifugation). The same definitions will be used for matter and nitrogen solubilisation.

COD solubilisation (S_{COD}) and degree of disintegration ($\mathrm{DD}_{\mathrm{COD}}$) were determined. S_{COD} was calculated using the difference between soluble COD (CODs) and initial soluble COD (CODs_0), compared to the initial particulate COD (CODp_0):

$$S_{\mathrm{COD}} = (\mathrm{CODs} - \mathrm{CODs}_0)/\mathrm{CODp}_0 \times 100\% \qquad (2)$$

The degree of disintegration was defined by Müller and Pelletier [10] as the comparison between ultrasonic process and a maximum soluble chemical demand $\mathrm{COD}_{\mathrm{NaOH}}$ obtained by alkaline hydrolysis:

$$\mathrm{DD}_{\mathrm{COD}} = (\mathrm{CODs} - \mathrm{CODs}_0)/(\mathrm{COD}_{\mathrm{NaOH}} - \mathrm{CODs}_0) \times 100\% \qquad (3)$$

For alkaline hydrolysis, sludge was mixed with NaOH (1 mol/l), for 24 h, at room temperature.

2.3.2. Matter composition

Measures of total and organic solids (TS and VS) were realised on sludge and on solids of centrifugation. That permitted the whole concentration and particulate matter concentration to be obtained. The solids concentration of the supernatant, that is to say the soluble phase, was then deduced and thus the composition in the different parts in the sludge. The measures were obtained using the normalised method [13]. Samples were heated at 105 °C for 24 h: water was evaporated off and the total dry matter concentration determined. Then, the samples were heated at 550 °C for 2 h. That led to mineral matter concentration. Organic matter concentration was then deduced.

2.3.3. Nitrogen composition

Measures of ammonium ($\mathrm{N\text{-}NH_4^+}$) and total Kjeldahl nitrogen (TKN) were realised. $\mathrm{N\text{-}NH_4^+}$ concentration was determined using distillation and a colorimetric dosage. TKN concentration was determined in the whole sludge and in the supernatant using the normalised method [13]. This measure represents organic nitrogen and ammonia.

2.3.4. Particle size

Particle size measurements were realised in the Laboratoire de Génie Chimique (LGC) in Toulouse, using a laser diffraction sensor (Mastersizer 2000, from the Malvern firm). Particle size was determined using a sphere of same volume.

2.4. Examination of biodegradability

Anaerobic biodegradability tests were carried out to compare biogas production for different substrates, with the same inoculum. The inoculum needed to be quite active, with a good adaptability and a low endogenous respiration. For these experiments, the inoculum used was a sludge treating a mixture of wine effluents (80%) and sludge (20%). Tests were realised under mesophilic conditions (35–37 °C), in batch mode, in penicillin bottles ($v = 100$ ml). The inoculum was diluted to 4 g of suspended volatile solids (VSS) per litre. Penicillin bottles were agitated (200 rpm) in order to homogenize samples. COD (0.5 g) to degrade per gram VSS in the inoculum were introduced in each bottle. The volume of biogas produced was measured using a syringe. Several control samples were realised: a blank (no sludge added, only water), an ethanol sample (completely biodegradable compound) and a non-treated sample (raw sludge added). For each supplied energy level, three tests were realised. For the first test, bottles contained 5 ml of the whole sludge. This run was reproduced in triplicates. For the second and third tests, the bottles contained, respectively, the supernatant and the particulate fraction of 5 ml of sludge. These two runs were carried out in order to evaluate the contribution of the soluble and particulate fraction in the biogas production.

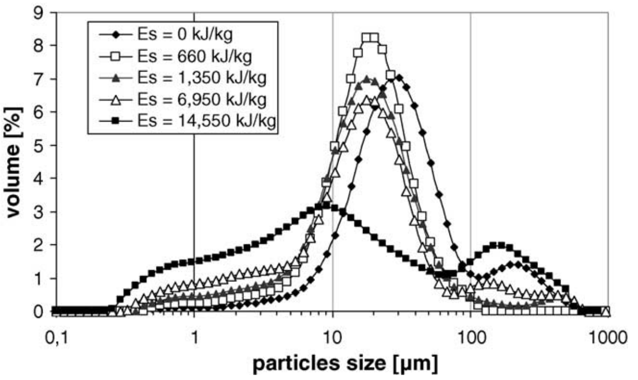

Fig. 1. Particles size distribution for different specific energy inputs.

3. Results and discussion

3.1. Particle size analysis

Particle size distribution is reported in Fig. 1. Particle size ranged from 0.4 to 1000 µm. For untreated sludge, particles size distribution was a peak centred on 30 µm and was quite narrow. For treated samples, peaks became larger: and the particle size distribution was more heterogeneous. The volume occupied by small particles increased with the specific supplied energy: for $E_s = 14{,}550$ kJ/kg TS particles of 1 µm occupied 1.5% of the whole volume, whereas they occupied 0.1% in the untreated sample.

On the other hand, the volume occupied by particles bigger than 100 µm increased largely with specific supplied energy. This could be explained by a re-flocculation phenomenon, already observed by Gonze et al. [14]. This re-flocculation may occur after ultrasonic treatment, due to the release of intracellular or extra-cellular material. Table 1 presents results obtained for cut diameter (d_{50}) and for Sauter mean diameter (dS). d_{50} is a cut diameter, that is to say that 50% of particles

Table 1
Cut diameter and Sauter mean diameter for different supplied energies

	E_s (kJ/kg TS)				
	0	660	1350	6950	14550
d_{50} (µm)	31.99	19.6	18.5	17.6	12.7
dS (µm)	18.5	11.2	8.3	5.8	3.7

(in volume) having a diameter lower or equal to d_{50}. Sauter mean diameter corresponds to the diameter of a sphere of the same surface area.

For an energy input lower than 1000 kJ/kg TS, the floc size reduction was important: d_{50} and dS strongly decreased, with a reduction of about 40%. Then, particles size decreased more progressively. First, energy applied was used to reduce flocs size. Then energy was used to break flocs and release extracellular material or to break cells and release intracellular material. This has not been really proved yet. In order to know if ultrasonic treatment at low frequency can break cells or not, it would be necessary to find an indicator of the cellular lyse. In this paper, expression "break cells" or "cellular lyse" will be used, even if they are misuses of language.

For this study, the minimum energy required to break cells was found to be about 1000 kJ/kg TS, that is to say about 20 kJ/l of sludge. This result is in agreement with Gonze et al. [14], who have found a minimal energy of 30 kJ/l, and with Lehne et al. [9], who have obtained an higher value but of the same order: 3000 kJ/kg TS.

3.2. Solubilisation

3.2.1. COD solubilisation

For each experiment, while the energy input increased, total COD was constant. During the experiments, the soluble/particulate COD repartition varied: soluble COD (CODs) increased whereas particulate COD (CODp) decreased. Cells underwent lysis and organic compounds were released into the liquid phase. CODs increased strongly for specific supplied energy between 0 and 10,000 kJ/kg TS: CODs ratio (that is to say CODs divided by total COD) increased from 4 to 32%. For higher specific energies applied, CODs and CODp were quite constant. Solubilisation and degree of disintegration are reported in Fig. 2.

For specific energy under 1000 kJ/kg, solubilisation and degree of disintegration are low: $S_{\mathrm{COD}} = 8\%$ and $\mathrm{DD}_{\mathrm{COD}} = 14\%$. These values are similar to those obtained by Lehne et al. [9] for $E_s = 3000$ kJ/kg TS (break point value). This energy was used to break flocs and not to lyse cells.

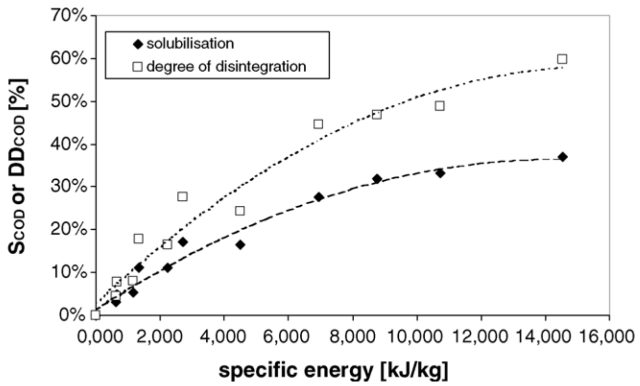

Fig. 2. Solubilisation and degree of disintegration of COD vs. supplied energy.

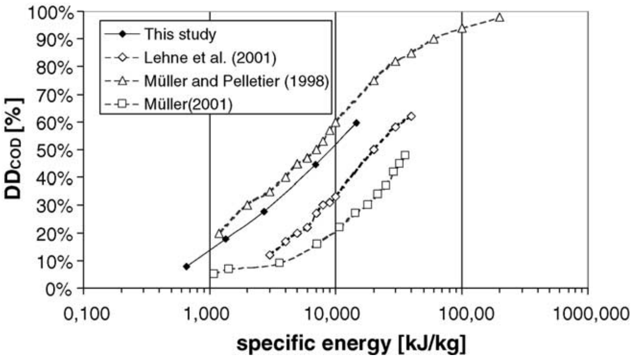

Fig. 3. Comparison of degree of disintegration obtained with different ultrasonic treatments.

For supplied energy over 1000 kJ/kg TS, solubilisation and degree of disintegration rose strongly; for $E_s = 15{,}000$ kJ/kg TS, $S_{\mathrm{COD}} = 35\%$ and $\mathrm{DD}_{\mathrm{COD}} = 55\%$. This degree of disintegration is in the same range of those obtained by other authors, as shown in Fig. 3.

COD measurements are normally applied for soluble pollution at low concentration. For these experiments, sludge was concentrated (total COD ≈ 22.5 g O_2/l) and had a non-negligible particulate fraction (CODp ≈ 14–21 g O_2/l). So, COD measurement was thus not well adapted for total COD measures, but was realised in order to compare results with those in the literature. The given sample results vary by up to 15%. It was concluded that it would be more effective to focus on parameters, which were more easily obtained such as matter distribution.

3.2.2. Matter distribution

Using ultrasound did not change total matter quantity. Total solids concentration (TS) was constant. Thus ultrasound, in terms of energy input, did not induce an evaporation phenomenon. Fig. 4 shows matter distribution.

The total mineral solid content and the total organic solids content were constant. Thus ultrasound did not induce a mineralisation phenomenon. However, the solid content of soluble (supernatant of centrifugation) and particulate (solids

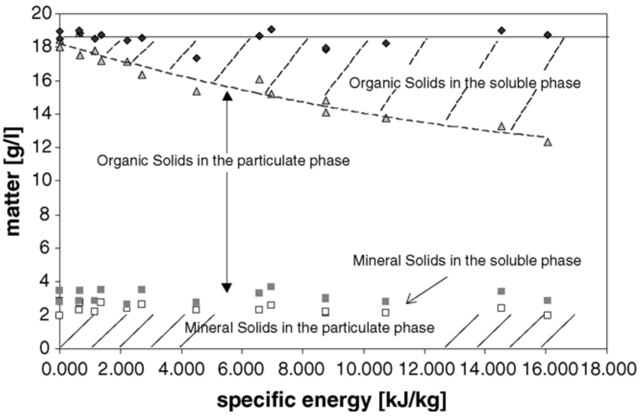

Fig. 4. Matter distribution as function of specific supplied energy.

Table 2
Synthesis of solubilisation results

	E_s (kJ/kg TS)					
	0	660	1355	2700	6951	14547
CODs/COD (%)	5.8	10.5	16.1	22.3	33.1	41.6
TS,s/TS (%)	5.7	10.5	12.2	16.8	25.9	36.5
VS,s/VS (%)	3.1	8.3	10.0	14.7	24.9	36.9
MS,s/MS (%)	17.6	20.3	21.9	25.4	29.8	29.6
TKN,s/TKN (%)	3.1	8.6	11.3	15.6	29.4	44.7
N-NH_4^+/TKN (%)	2.3	4.1	4.6	5.7	11.3	18.2

of centrifugation) parts varied with specific supplied energy. Soluble matter concentration increased, whereas particulate matter concentration decreased. Thus, ultrasound led to a solubilisation phenomenon of organic solids but also of mineral solids. Solubilisation of mineral matter was very low, less than 3%, whereas organic solubilisation was quite high 29% for a specific energy of 15,000 kJ/kg TS. In fact, total solid solubilisation (S_{TS}) increased with energy input. This increase was fast for specific energy between 0 and 8000 kJ/kg TS (S_{TS} = 25% for E_s = 7000 kJ/kg TS). For higher specific supplied energies, total solids solubilisation seemed to be slower (S_{TS} = 32% for E_s = 15,000 kJ/kg TS).

In terms of matter solubilisation, optimum results were obtained for a supplied energy of 10,000 kJ/kg TS that is to say about 200 kJ/l, for this study.

3.2.3. Nitrogen solubilisation

Cells were broken due to ultrasound. Intracellular compounds were released into the liquid phase and were made soluble. So ultrasounds led to a nitrogen release. Fig. 5 shows nitrogen distribution.

On the whole sludge, total nitrogen (TKN) was constant whatever the specific energy. This means that ultrasound did not lead to a nitrogen mineralisation or volatilisation. For a specific energy increase, the quantity of organic nitrogen in particles decrease and organic nitrogen concentration in soluble phase and ammonia concentration increased. Thus organic nitrogen was made soluble. For specific supplied energy of 15,000 kJ/kg TS, organic nitrogen solubilisation was about 40%. Very little organic nitrogen was transformed into ammonium. Nitrogen is mainly in proteins or amino acids. Proteins were made soluble but were not completely degraded. In terms of nitrogen solubilisation, maximum results were obtained for an supplied energy of 10,000 kJ/kg TS that is to say about 200 kJ/l, for this study.

3.2.4. Solubilisation synthesis

Table 2 summarises results obtained in terms of solubilisation during this study. Solubilisation increased with specific supplied energy.

In terms of COD, solubilisation was fast for specific energies lower than 10,000 kJ/kg TS, and then slower. In terms of solids, solubilisation was centred on organic matter. Total solid ratio (TS,s/TS) and organic solid ratio (VS,s/VS) were close, especially for high-specific supplied energy. Nitrogen was made soluble, but was not degraded: TKN ratio (TKN,s/TKN) increased strongly, whereas N-NH_4^+ ratio increased very slowly.

3.3. Sludge biodegradability

Fig. 6 shows results obtained for whole sludge biodegradability tests. Biogas production for the blank was very low, i.e. during all the experiments endogenous biogas production was low.

For each treatment, biogas production was higher than that for untreated sludge: minimum increase of 25% in biogas vol-

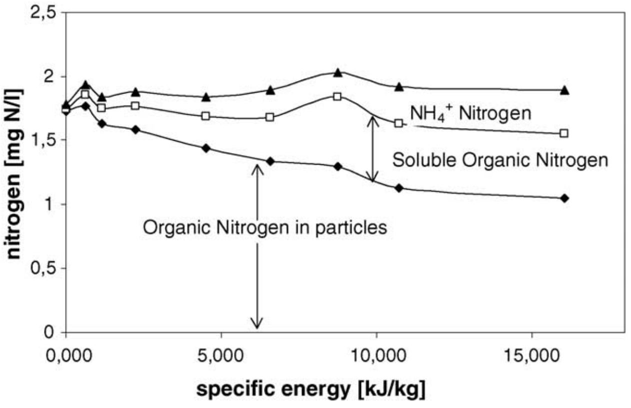

Fig. 5. Nitrogen distribution as function of specific supplied energy.

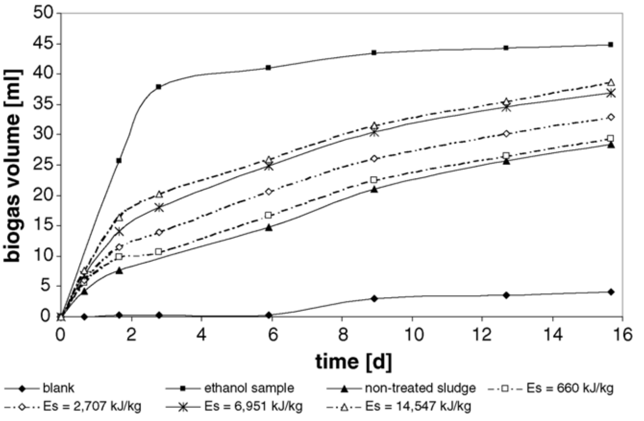

Fig. 6. Biogas volume produced by treated sludge.

Table 3
Biogas produced linked to soluble and particulate organic matter

	Specific energy (kJ/kg TS)					
	0	660	1350	2700	6950	14547
Total organic matter to degrade (bottle 1) (mg)	81.5	80.9	79.9	79.3	76.8	82.0
Particulate organic matter to degrade (bottle 2) (mg)	79.0	74.2	71.9	67.6	57.7	51.7
Soluble organic matter to degrade (bottle 3) (mg)	2.5	6.8	8.0	11.7	19.1	30.3
Biogas produced with the whole sludge (bottle 1) (ml)	20.5	23	25.6	25.7	31.2	32.8
Biogas produced by particulate matter (bottle 2) (ml)	19.5	19.6	20.9	19.3	17.3	16.6
Biogas produced by soluble matter (bottle 3) (ml)	0.7	1.6	5.2	5.4	11.4	13.8

ume was obtained with ultrasonic pretreatments. These volumes were always lower than for a completely biodegradable substrate (ethanol). Ultrasound led to an increase of sludge biodegradability, but they were not fully biodegradable. For specific supplied energies of 660 and 1350 kJ/kg TS, biogas production was the same. This can be explained by variation in particle size. Indeed, these energies were very close to the minimal energy necessary (1000 kJ/kg). Energy was used to reduce flocs size, not to break cells. Organic compounds were not released in the liquid phase, but matter was much more available. This was also confirmed by results on matter and nitrogen solubilisation. For $E_s = 1000–7000$ kJ/kg TS, gas production increased with supplied energies. Solubilisation made compounds more available for bacteria of anaerobic digestion. For supplied energies of 7000 and 15,000 kJ/kg TS, biogas production was almost the same. Biogas volume was 1.4 times higher than that for untreated sludge.

Some experiments were conducted in order to determine the effect of each part (soluble and particles) in the biogas production. Table 3 shows the importance of soluble and particulate parts in the biogas production. The volume of biogas produced with the soluble part of the sludge increased with specific supplied energy, whereas the volume of biogas produced with the particulate fraction was quite constant. Moreover, the sum of the gas volumes produced with soluble and particulate parts (bottle 3 + bottle 2) was equal to the volume produced with the whole sludge (bottle 1) in 8% near. That allows assumptions to be made with regard to biogas origin. Thus, for non-treated sludge, biogas was produced at 97% from the particulate matter, whereas for sludge treated at about 7000 kJ/kg TS, biogas was produced at 60% only from the particulate matter.

Biogas volume linked to soluble fraction increased quickly for $E_s < 7000$ kJ/kg TS. Then volume seemed to be constant. For $E_s = 7000$ kJ/kg TS, the volume of biogas produced by soluble fraction was three times higher than that for untreated sludge. It was possible to link organic soluble matter to degrade with the volume of biogas produced. For the conditions of these biodegradability tests, the relation between biogas produced by the soluble fraction (vbs) and organic soluble matter to degrade (OSMd) was linear: vbs (ml) = 0.49 OSMd (mg) with $r^2 = 0.93$. At the same time, the volume of biogas produced with the particulate fraction was almost constant for low specific energies lower than 3000 kJ/kg TS, even if solids content in particles decreased with supplied energy (TSS reduction of 15% for $E_s = 3000$ kJ/kg TS). Organic solids in the particulate fraction were not easily available for bacteria and were only slowly biodegraded. For $E_s > 3000$ kJ/kg TS, biogas produced with the particulate fraction was a bit lower, but there was no enough data to determine a relation between biogas produced and organic matter in particle. However, these results show that for low energy input ($E_s < 3000$ kJ/kg TS), biogas was mainly produced by the particulate fraction. For higher specific energies applied, the volume of biogas produced by particulate and soluble fractions was of the same order of magnitude. Total biogas volume increased because more matter was soluble. Solids contained in the particulate part of the sludge were made soluble due to ultrasound. They were more available in the soluble fraction for bacteria: thus biogas production increased.

4. Conclusions

Ultrasounds were shown to have an effect on sludge solubilisation and on biogas production during batch anaerobic digestion of sonicated sludge. COD, matter and nitrogen solubilisation increased with supplied energy. In the same time, biogas production increased too. The ultrasonic process led to floc size reduction and cells lysis. It was necessary to supply enough energy in order to lyse cells. For specific supplied energy lower than 1000 kJ/kg TS, energy was used in order to reduce flocs size. Then, supplementary energy was used to break flocs or cells. That permitted the release of organic substances into the liquid phase. Organic substances were more available, so biodegradability was improved. In term of biogas production, it did not seem interesting to have a supplied energy higher than 7000 kJ/kg TS. Indeed, when the supplied energy was higher than 7000 kJ/kg TS, biogas generation was constant and solubilisation was less marked. Moreover, biogas production linked to the particulate fraction did not depend on matter concentration for low energy input. Biogas production linked to particles was limited. But if matter was solubilised, this matter became available for bacteria.

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