

Ultrasonically enhanced anaerobic digestion of sludge

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Anaerobic digestion of wastewater treatment sludge is slow and incomplete as a result of the rate-limiting sludge hydrolysis step, due to the low biodegradability of the cell walls and the presence of extracellular biopolymers. Methods to enhance the biogas production should hence make the substrate more accessible to the anaerobic microorganisms. Low-frequency ultrasound treatment is emerging as the most appropriate technology. The present study couples laboratory sonification with digestion experiments (at 37°C). Results demonstrate that ultrasound treatment can – within certain ranges of specific energy-input (SE) – achieve (i) an increased disintegration of the sludge, observed from an increasing soluble COD and BOD; (ii) an increased release of volatile fatty acids (VFA) as a result of the oxidising radicals being formed through cavitation; and (iii) an increased biogas production. Ultrasound enhances the biogas production by over 40% at low SE, and approximately 15% at higher SE-values.

1. Introduction

Sludge disposal is a growing problem, representing up to 50% of the current operation costs of a waste water treatment plant (WWTP) (Neyens et al., 2004). Sludge stabilisation is a common practice with anaerobic digestion as dominant process since both organic matter is transformed into net energy in the form of biogas (60 to 70 vol% CH₄), thereby also reducing the final amount of sludge to be disposed, and pathogens present in the sludge are significantly destroyed, thus limiting possible hazards.

During anaerobic digestion, hydrolysis is generally accepted as being the rate limiting step in the process (Tiehm et al. 2001, Qasim 1999). Various pre-treatments have been studied and include mechanical, thermal, chemical and biological interventions (Dewil et al. 2007, Neyens and Baeyens 2003, Climent et al. 2007, Müller et al. 1998). All pre-treatments result in a certain degree of lysis or disintegration of sludge cells, hereby releasing and solubilising intracellular material into the water phase and transforming refractory organic compounds into biodegradable material. Previous research (Dewil et al. 2006) has demonstrated that ultrasound causes sludge flocs to be disrupted, altering its constituent structure and solubilising organics. This paper will address the use of

sonification to enhance the anaerobic digestion of WAS in order to define and quantify the optimum operating conditions, and the various associated phenomena.

2. Literature survey on ultrasonic WAS treatment

The cavitation phenomenon associated with the ultrasound treatment of liquids is well documented (Laborde et al. 1998, Lauterborn and Ohl 1997, Vera 2007). Cavitation only occurs at frequencies below 1 MHz. It was shown (Tiehm et al. 2001, Young 1989) that the radius of the bubbles and hence the energy released by implosion is inversely proportional to the frequency of the generating ultrasound. Cavitation will result in the promotion of chemical reactions due to locally high temperature and pressure; in creating extreme shear forces in the liquid (mechanically attacking components); and in the formation of highly reactive radicals ($H\bullet$ and $OH\bullet$) which facilitate chemical reactions to destroy organic contaminants.

The pre-cited mechanisms of ultrasound waves attack the bacterial cell walls whereupon exo-enzymes are released, resulting in the breakdown of organic material into readily biodegradable fractions. Bacterial kinetics are hereby enhanced, resulting in an increased biogas production (Bougrier et al. 2006, Vera 2007). The effect of intensity on enhanced biogas yield is not unanimously confirmed despite the enhanced biological hydrolysis, the preliminary step in the anaerobic treatment of WAS. Literature reports results of a increased biogas yields increased by as low as 10 to as high as 48%. Due to the ultrasonic destruction of organic material, hydrolysis was accelerated and volatile fatty acids (VFA) were more readily generated through acidogenesis and subsequently transformed into methane through methanogenesis (Quarmby et al. 1999). The dry solids content of the sludge is an important parameter for the disintegration with increased effectiveness at increased concentration of DS in the WAS (Tiehm et al. 2001, Onyeche et al. 2002, Dewil et al. 2006): more DS creates more nuclei for cavitation and that more particles were exposed to the resulting shear forces (Lehne et al. 2001).

Since the concentration of DS affects the results of sonification, data are commonly presented in terms of SE, i.e. per kg DS. For anaerobic digestion, a definition on the basis of kJ/kg (ODS)₀ [(ODS)₀ being the initial ODS-content, before digestion] seems more appropriate, since only ODS participates in the biogas synthesis reaction.

Some additional effects on the sludge quality and properties are seen when treating it ultrasonically, i.e. a disruption of the extracellular polymeric substances (EPS) and the cell walls of the micro-organisms in the sludge (Onyeche et al. 2002); a reduction of the floc size (Dewil et al. 2006) and a reduced dewaterability unless extra polyelectrolyte is used; and inconclusive effects on the filamentous WAS components (Neis and Tiehm 1999, Dewil et al. 2006).

3. Experimental set-up and procedures

3.1. Ultrasonic equipment

WAS sludge was pumped through a tubular piezo-ceramic sonification reactor (Bandelin SONOREX TECHNNIK Sonobloc® SB 5.1-1002). The energy-input, expressed in kJ/kg DS or in kJ/kg (ODS)₀, was determined by the applied power (max. 1 kW) and the exposure time of the sludge together with the DS/ODS-content of the

sludge volume under scrutiny. A field of 25 kHz was used in accordance with literature recommendations, and the sonification time was varied from 0 to 250 min by using recirculation of the WAS. The amplitude and hence the intensity of the ultrasound waves can be varied. The maximum flow rate is 50 l/min.

3.2. Anaerobic digester set-up

Twelve anaerobic digesters were built as parallel operating mesophilic single stage batch reactors, each of 1 l content and mechanically stirred. Specific attention was paid towards avoiding the scum layer by adapting the mixing rate. The digestion experiments were run for 8 days. A mixture of 300 g WAS and 400 g seed sludge was used. For each experimental run, two control digesters were operated with non-sonificated sludge. Only the 300 g of WAS were sonificated at different energy input levels (as described above) prior to mixing with the seed sludge. The produced gas was collected in calibrated glass cylinders. The cylinders were filled with acidified de-ionised water to avoid losses of CO₂ due to the formation of carbonates. The digesters were moreover fitted with airtight septum sampling points for direct extraction of samples of the sludge mix at regular intervals for measurements of COD, VFA and biogas composition.

3.3. Analytical methods

The dry solids content (wt.% DS) of the wastewater, of the sludge cake and of the filtrate, as well as the organic content of these different samples (wt.% ODS) were measured by standard procedures (APHA 1992).

Analysis of total COD, soluble COD, BOD, volatile fatty acids (VFA) and biogas was performed by procedures described in detail elsewhere (Appels et al.2008).

3.4. Sludge characteristics

All sludge samples were obtained from the WWTP of Deurne-Antwerp (B). This 300,000 p.e. WWTP is operated without primary settling and with a low-load combined aerobic-anoxic plug flow reactor. The WAS was obtained from the sludge thickeners. The digested (seed) sludge was sampled from the underflow of the digesters. The sludge characteristics are given in Table 1.

Table 1: Characteristics of thickened WAS, seed sludge and seeded mixture

Parameters	WAS	Digested sludge (Seed)	Seeded Mixture
DS (g DS/kg sludge)	38.10 to 48.50	53.89 to 54.88	44.31 to 52.37
ODS (as % of DS)	64.06 to 67.26	54.22 to 58.42	55.34 to 63.11
COD _o (mg/l)	1875 to 2220	1160 to 1450	1475 to 1900

4. Results and discussion

4.1. Validation of the experiments

Prior to examining the effect of sonification, the digester design and operation procedures were validated by repeating the experiments 4 times on untreated WAS collected at 3 subsequent days. Essential digestion parameters (DS-content, ODS-

percentage, COD, biogas production) were measured. The DS-content of the seeded mixture was 48.34 g/kg of sludge with 60.94 wt% ODS. The initial COD of the seeded mixture was 1523 mg/l. The COD was reduced by digestion to 1356 mg/l.

The biogas production was between 2460 and 2610 ml, i.e. within $\pm 5\%$ of the average production. The biogas production varied nearly proportionally with the initial amount of ODS $[(\text{ODS})_0]$ present. The average values of the digestion results are given in Table 2, for kJ/kg DS equal to zero (no sonification).

4.2. Effect of sonification on the COD release

The destruction of the sludge flocs can be measured in terms of the change of COD (ΔCOD) in the filtrate of the sonificated WAS. ΔCOD was determined by measuring both the total and the soluble fraction of COD (sCOD) before and after the ultrasonic treatment. Figure 1 illustrates both the change of COD and BOD in the filtrate in terms of the specific energy (SE) applied for a DS-concentration of 48.34 g DS/kg at 1 kW.

The destruction rate and hence the sCOD-fraction increases significantly with increasing specific energy (and consequently with increased duration of the experiments). Destruction starts at very low SE-values. Both the soluble COD and BOD increase with increasing SE, implying that a considerable fraction of the released COD is transformed into biodegradable organic components, thus increasing the BOD.

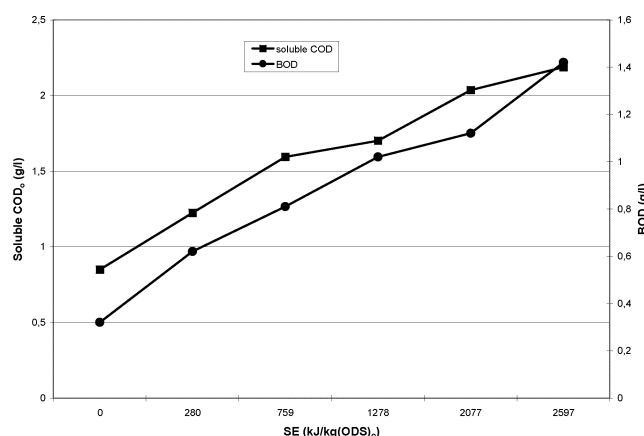


Figure 1: Effect of the dosage of ultrasound treatment on the sCOD and BOD of the filtrate at 20 kHz

4.3. Effect of sonification on the biogas production

Although high inputs of ultrasonic energy are economically not justified as illustrated in the final section of this paper, a full range of specific energy inputs were examined, in line with common ranges previously applied in literature, and in order to elucidate the very wide spread in biogas enhancements reported in literature.

Experiments were performed in triplicate, and average data of Table 2 are further used. These averages were within 5% of the maxima and minima measured.

Table 2: Experimental data obtained at different US energy-inputs, expressed in kJ/kg (ODS)₀, for $t = 8$ days (0 kJ/kg DS corresponds to the control digesters)

kJ/kg DS	0	168	456	768	1248	1560	5850	8180
kJ/kg ODS	0	280	759	1278	2077	2597	9737	13615
Δ DS (g)	5.51	5.7	5.6	5.35	5.51	5.39	5.43	6.41
Δ ODS (g)	5.46	5.65	5.2	5.15	5.2	5.24	5.2	5.61
COD _t (mg/l)	1356	1498	1075	1033	1028	1045	1213	1563
Biogas (ml)	2383	3570	2750	2591	2689	2660	2512	2501
Δ ODS/(ODS) ₀ (%)	15.70	16.24	14.95	14.81	14.95	15.07	14.95	16.13
Biogas (ml/kg Δ ODS) at 20°C	0.436	0.632	0.529	0.503	0.517	0.508	0.483	0.446
Biogas (Nm ³ /kg Δ ODS)	0.407	0.589	0.493	0.469	0.482	0.473	0.450	0.416

Sonification results in a marked enhancement of the biogas production at low SE-values with a gain of between approx. 20 and 45%. At moderately high values, the effect is less pronounced although still with a ± 15 % gain. Very high SE-inputs do not significantly increase the biogas production. Clearly, the digestion is less efficient despite a continued COD release (Figure 1 and Table 2): the released COD is not transformed in extra biogas. Parallel to the COD-effect, the rate of biogas formation needs also to be coupled to the VFA release, as will be discussed below: here too, high SE-values do not further promote VFA release.

The decrease of ODS is limited to some 15%. The differences in biogas yields can hence only be explained by the VFA release, and subsequent methanogenesis.

3.8. Release of volatile fatty acids

A clear distinction must be made between the effects of sonification, and the effects of digestion. During digestion, VFA are formed and consumed in the hydrolysis and/or acidogenetic digestion phase. It is therefore difficult to determine the exact VFA concentration during digestion, since the formation rate is expected to be equal (or proportional) to the further disappearance during methanogenesis (Appels et al., 2008). The results of the VFA concentration after sonification (VFA₀) and after 8 days of digestion (VFA_t) are shown in Table 3, where Δ ODS-values are taken from Table 2.

Table 3: VFA

kJ/kg DS	0	168	456	768	1248	1560	5850	8180
kJ/kg ODS	0	280	759	1278	2077	2597	9737	13615
VFA ₀ (mg/l)	94,4	242,3	372,2	524,0	565,0	351,6	268,2	256,7
VFA _t (mg/l)	13,0	21,7	63,6	6,7	31,3	30,7	28,1	80,4
Δ VFA (mg/l)	81,4	220,7	308,6	517,2	533,7	320,9	240,1	176,3
Δ ODS (g)	5,46	5,65	5,2	5,15	5,2	5,24	5,2	5,61
Biogas (ml)	2383	3570	2750	2591	2689	2660	2512	2501

From these results, it is clear that VFA are increasingly formed during sonification, in-line with the expected radical reactions ($H\bullet$ and $OH\bullet$) of cavitation, up to an energy input of about 1250 kJ/kg DS or 2100 kJ/kg (ODS)_o; organic compounds of the WAS are disrupted and oxidised to their carboxylic acid form. The reason why this transformation stops at higher SE-values has yet to be established, although it is expected that higher energy-inputs will lead to the disruption of the cells into larger building molecules, less prone to oxidation to VFA, although the soluble COD continues to increase. The initial significant release of VFA at low SE also explains the higher biogas synthesis (through acidogenesis) at lower SE-values. The available VFA (up to 5 times the value of non-treated WAS) increase the biogas production. Since the biological hydrolysis is the rate limiting step in a conventional anaerobic treatment, it is seen that the ultrasound release of VFA has a significant influence on the biogas yield.

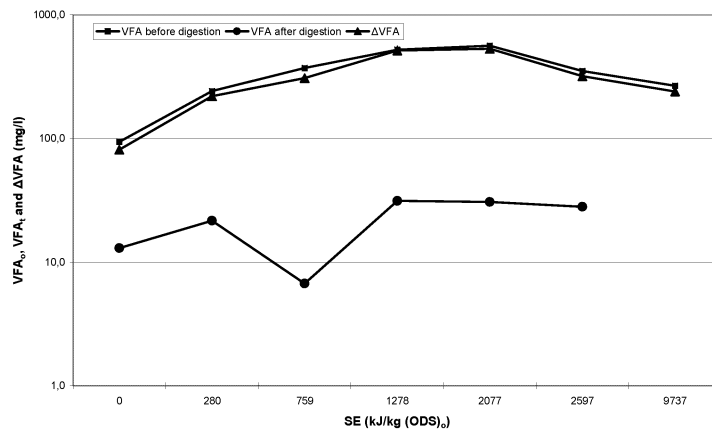


Figure 2: Influence of SE, expressed as kJ/kg (ODS)_o, on the release of VFA (mg/l)

4. Economic assessment of the ultrasound treatment

The economy of the ultrasound can be assessed by comparing the extra amounts of biogas (65 % CH₄, 22500 kJ/Nm³) with the energy-input needed. Anaerobic digestion itself, with a biogas production of approximately 0.4 Nm³/kg ODS-removed, has an economic benefit, despite the higher dosage of PE to be applied for dewatering (from 10 gPE/kg DS to 12 gPE/kg DS). The additional amount of biogas formed when applying 168 kJ/kg DS, or 280 kJ/kg ODS, accounts to ~ 0.18 Nm³/kg ODS or an extra 1050 kJ/kg ODS thus providing a net gain of 770 kJ/kg ODS. Beyond 280 kJ/kg ODS, the extra amount of biogas formed is ~20% only. The range of economic ultrasound-application is hence restricted to lower energy-inputs, certainly not exceeding ~4000 kJ/kg ODS or 2400 kJ/kg DS. Possible economic benefits are illustrated in Table 4.

For an operation of 8000 hrs/year, the benefit is 28,800 €/year. Considering the investment cost of an ultrasound treatment of max. 20 kW to be around € 40000, capable of treating sludge from a WWTP of 10000 p.e., the benefit yields a payback period of less than 2 years since the biogas generated will benefit from green-power certificates. These findings are confirmed by industrial applications. Capital costs today are roughly € 20000 /kW. Operation and maintenance costs are minimal although the

ultrasound probes need replacement every 1.5 to 2 years. WWTP reports, ranging from 50000 to 750000 p.e. (for a total of several Mp.e.). reveal an improved ODS destruction at around 30 %, an increased biogas production by about 30%. Since the degradation rate is accelerated, the dimensions of the digesters can moreover be reduced for a given load, thus reducing the impact of high capital requirements. This has been applied in Bamberg (D, 250000 p.e.) (Ultrawaves, 2008), where the application of ultrasound avoided the construction of a third 3000 m³ digester.

Table 4: Economic evaluation for a sludge treatment(20 m³/hr, 5 wt% DS and 60% ODS i.e. ~ 10,000 p.e.). 15% ODS removal through digestion [PE = polyelectrolyte]

cost of PE after ultrasound, with 10 % reduction of DS; 12 g PE/kg DS: 900 kg DS/h x 5 €/kg PE x 0.012 kg PE/kg DS	+ 54 €/h
cost of PE before treatment at 10 g PE/kg DS: 1000 kg DS/h x 5 €/kg PE x 0.010 kg PE/kg DS	- 50 €/h
Replacement of 40000 € sonotrode (every 2 years, for 8000 hrs/year)	+ 2.5 €/h
cost of ultrasound treatment: 20 kWh x 0.08 €/kWh	+ 1.6 €/h
Extra production of biogas (20 % on a 15 % reduction of ODS): (600 kg ODS/h x 0.15 x 0.2 Nm ³ / kg ODS x 0.65 € /Nm ³)	- 11.7 €/h
Total extra gain	3.6 €/h

5. Conclusions

The present paper demonstrated that ultrasound treatment can at low specific energy-input achieve (i) an increased disintegration of the sludge, as witnessed by an increasing soluble COD-fraction accompanied by an increasing presence of BOD; (ii) an increased release of volatile fatty acids (VFA) as a result of the oxidising radicals being formed through cavitation; and (iii) an increased biogas production of up to 40 % at low SE-values, and about 15 % at moderate SE. High SE-values do not promote digestion.

The economics of the treatment are excellent.

6. References

- APHA, 1992. *Standard methods for the examination of water and wastewater*. 18th ed. Washington DC: American Public Health Association.
- Appels, L., Dewil, R. and Baeyens, J. 2008. Ultrasonically enhanced anaerobic digestion of municipal sewage sludge, accepted for publication in *Int.Jnl.Sust.Engrg.*, April 2008 issue
- Bougrier, C., Albasi, C., Delgenes, J.P., Carrere H., 2006. Effect of ultrasonic, thermal and ozone pre-treatments on waste activated sludge solubilisation and anaerobic biodegradability. *Chemical Engineering and Processing*, 45(8), 711-718.
- Chiu, Y.-C., Chang, C.-N., Lin, J.-G. and Huang, S.-J., 1997. Alkaline and ultrasonic pretreatment of sludge before anaerobic digestion. *Water Science and Technology*, 36(11), 155-162.
- Clark, P.P. and Nuijoo, I., 1998. *Ultrasonic sludge pre-treatment for enhanced sludge digestion*. Proceedings of Innovation 2000 Conference, UK, Cambridge.

- Climent, M., Ferrer, I., del Mar Baeza, M., Artola, A., Vázquez F. and Font, X., 2007. Effects of thermal and mechanical pretreatments of secondary sludge on biogas production under thermophilic conditions. *Chemical Engineering Journal*, 133(1-3), 335-342.
- Dewil, R., Baeyens, J., Goutvrind, R., 2006. Ultrasonic treatment of waste activated sludge, *Environmental Progress*, 25(2), 121-128.
- Dewil, R., Appels, L., Baeyens, J. and Degreè, J., 2007. Peroxidation enhances the biogas production in the anaerobic digestion of biosolids. *Journal of Hazardous Materials*, 146(3), 577-81.
- Laborde, J.L., Bouyer, C., Caltagirone, J.P. and Gérard, A., 1998. Acoustic bubble cavitation at low frequencies. *Ultrasonics*, 36(1-5), 589-594.
- Lehne, G., Mümler, A. and Schwedes, J., 2001. Mechanical disintegration of sewage sludge. *Water Science and Technology*, 43(1), 19-26.
- Müller, J., Lehne, G., Schwedes, J., Battenberg, S., Nèveke, R., Kopp, J., Dichtl N., Scheminski, A., Krull R. and Hempel, D.C., 1998. Disintegration of sewage sludges and influence on anaerobic digestion. *Water Science and Technology*, 38(8-9), 425-433.
- Neis, U. and Tiehm, A., 1999. *Ultrasound in wastewater and sludge treatment*. Reports on Sanitary Engineering N°25, Technical University Hamburg-Harburg.
- Neis, U., 2000. Ultrasound in water, wastewater and sludge treatment. *Water*. 21, 36-39.
- Neyens, E. and Baeyens, J., 2003. A review of thermal sludge pre-treatment processes to improve dewaterability. *Journal of Hazardous Materials*, 98(1-3), 51-67.
- Neyens, E., Baeyens, J., Dewil, R. and De heyder, B., 2004. Advanced sludge treatment affects extracellular polymeric substances to improve activated sludge dewatering. *Journal of Hazardous Materials*, 106(2-3), 83-92.
- Onyeché, T.I., Schläfer, O., Bormann, H., Schröder, C. and Sievers, M., 2002. Ultrasonic cell disruption of stabilized sludge with subsequent anaerobic digestion. *Ultrasonics*, 40(1-8), 31-35.
- Qasim, S.R. 1999. *Wastewater Treatment Plants: Planning, Design and Operation*. 2nd ed. Boca Raton: CRC Press.
- Quarmby, J., Scott, J.R., Mason, A.K., Davies, G. and Parsons, S.A., 1999. The application of ultrasound as pre-treatment for anaerobic digestion. *Environmental Technology*, 20(11), 1155-1161.
- Tiehm, A., Nickel, K. and Neis, U., 1997. The use of ultrasound to accelerate the anaerobic digestion of sewage sludge. *Water Science and Technology*, 36(11), 121-128.
- Tiehm, A., Nickel, K., Zellhorn, M. and Neis, U., 2001. Ultrasonic waste activated sludge disintegration for improving anaerobic stabilisation. *Water Research*, 35(8), 2003–2009.
- Ultrawaves, 2008. Ultrasound sludge disintegration for anaerobic digestion acceleration and enhancement. Bamberg WWTP, Germany. www.ultrawaves.de/downloads/case-studie-bamberg.pdf (accessed on 15/02/2008).
- Vera, M.A., 2007. *Improved anaerobic digestion with ultrasound disintegration*. In: Proceedings of Hawaii Water Environment Association, Oahu.
- Vesilind, P.A. and Davis, H.A., 1988. Using the CST device for characterising sludge dewaterability. *Water Science and Technology*, 20, 203-205.
- Young, F.R., 1989. *Cavitation*. London: McGraw-Hill.