Effect of feed/inoculum ratio on anaerobic digestion of sonicated sludge

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C.M. Braguglia*, G. Mininni*, M.C. Tomei* and E. Rolle**

*CNR-Istituto di Ricerca sulle Acque, Via Reno 1-00198 Rome, Italy (E-mail: *braguglia@irsa.cnr.it*; *mininni@irsa.cnr.it*; *tomei@irsa.cnr.it*)

**Department of Hydraulics, Transportation and Roads, Faculty of Engineering, University of Rome "La Sapienza", via Eudossiana 18, 00184 Rome, Italy (E-mail: *enrico.rolle@uniroma1.it*)

Abstract In recent years, relevant interest has been devoted to activated sludge disintegration and solubilisation techniques in order to cope with the biological limitations related to particulate degradation. Mechanical disintegration with ultrasound can efficiently transform insoluble organics into a soluble form: the solubilised organic matter is released from the cells to the bulk phase, thus accelerating the hydrolysis step in the digestion process. Experiments were carried out on bench scale anaerobic reactors fed with either untreated or disintegrated excess sludge, added with a biomass inoculum taken from a full scale anaerobic digester. Digestion tests have been carried out at different feed/inoculum ratios (F/I) in the range of 0.1–2, kinetics of VS reduction has been investigated and a beneficial effect of sonication is observed for all the experimental conditions. Similar beneficial results have also been found for biogas production with a maximum gain of 25% at 0.5 F/I ratio.

Keywords Activated sludge; anaerobic digestion; biogas; COD; hydrolysis kinetics; ultrasound

Introduction

In the future, a considerable increase in the global sludge production is foreseen, caused by the expansion of wastewater treatment in developing countries and in small scale municipalities, and by more stringent effluent criteria. The produced sludge is highly septic and therefore requires stabilisation in order to enable environmentally safe utilisation or disposal. Anaerobic digestion is the standard stabilisation technique for large plants, resulting in the reduction of sludge volatile solids and the production of biogas. In the European Union, 50% of municipal sewage sludges are stabilised anaerobically (Davis and Hall, 1997). The anaerobic digestion process consists of four stages: hydrolysis, acidogenesis, acetogenesis and methanogenesis. In the case of sewage sludge digestion, the biological hydrolysis has been identified as the rate-limiting step, and that renders the degradation a slow process requiring large reactors. Therefore, the pre-treatment of excess sludge by mechanical disintegration can improve subsequent anaerobic digestion (Müller *et al.*, 1998; Neis *et al.*, 2000). The aim of the ultrasonic treatment is to increase the soluble available organic matter by disrupting the flocs and/or lysing the cells, exposing released material to further anaerobic digestion. There is, however, a lack of information on the effect of VS load on the process performances.

Untreated excess sludge digestion tests carried out in Germany on batch reactors at different F/I (from 0.15 to 8.37) showed that by decreasing the inoculum percentage (high F/I) the initial "lag-phase" in biogas production becomes longer but the total specific biogas production remains approximately constant, independently of F/I. The F/I optimum range, in terms of biogas production and volatile solids reduction, was assessed to be between 0.5 and 2 for untreated sludge (Engelhart, 2002).

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The objective of this study was to evaluate the effect of the initial ratio between raw activated sludge and anaerobic inoculum (F/I in terms of volatile solids) on the hydrolysis process kinetics, both for sonicated sludge and for the untreated one. For this purpose, VS biodegradation kinetics, soluble COD pattern and cumulative biogas production have been investigated in parallel batch tests.

Materials and methods

Sludge

Excess sludge was obtained from the municipal "Roma-Nord" wastewater treatment plant, one of the four wastewater treatment plants of the city of Rome managed by ACEA, the public company being in charge of the management of water and wastewater treatment plants in the Rome municipality.

The plant is conventional, including screening, primary clarification and secondary treatment by activated sludge.

The excess sludge was sampled from the recycle stream before thickener. This treatment plant serves approximately 700,000 P.E. and is operated with a quite high sludge age (20 d) due to the unusual dilution of incoming sewage (COD average value of 200 mg/L). The anaerobic inoculum was sampled from the full scale digester of the plant fed with primary and secondary sludge.

Sludge disintegration by ultrasound

The disintegration by ultrasound was performed with an ultrasonic processor UP400S (Dr. Hielscher, Germany) operating at 255 W and 24 kHz. The power control had an amplitude of 20–100%. The sonotrode has a diameter of 22 mm making the device suitable for sample volumes from 100 to 2,000 mL.

To assess the degree of sludge disintegration, the chemical oxygen demand (COD) in the sludge supernatant was determined. The degree of disintegration (DD_{COD}) is calculated as the ratio between COD increase due to sonication and the total COD:

$$DD_{COD} = \left[\frac{(COD_{sol,son} - COD_{sol,tq})}{COD_{max}}\right] \cdot 100 \quad (1)$$

where $COD_{sol,son}$, $COD_{sol,tq,}$ and COD_{max} are the supernatant COD (mg/L), respectively, of the sonicated sample, of the untreated sample and of the reference sample after complete chemical mineralisation with H_2SO_4. The degree of disintegration is dependent on the specific energy supplied (E_{spec}), which can be calculated by equation (2) considering that P is the power of the ultrasonic homogeniser, t the sonication time, V the sludge treated volume, and TS the sludge total solid content:

$$E_{spec} = \frac{P \times t_t}{V \times TS} \quad (2)$$

Sonication was performed for 4 min on 500 mL of waste-activated sludge (2.5–3% TS) placed in a 1 L beaker with the probe located at 3 cm above the beaker bottom. The specific energy was in the range 4,500–5,000 kJ/kg dry solids, depending on the TS of the treated sample.

Anaerobic digestion tests

Experiments were carried out on bench scale anaerobic reactors of 0.4 L (working volume) that were operated in batch mode, immersed in a temperature controlled, agitated water bath at 37 °C. The reactors were fed with a mixture of inoculum and activated sludge, either untreated or disintegrated (degree of disintegration about 13%). *F/I* was calculated as follows:

$$\frac{F}{I} = \frac{V_{sludge} \cdot VS_{sludge}}{(V_{tot} - V_{sludge}) \cdot VS_{inoculum}} \tag{3}$$

The produced biogas was collected in a calibrated 1-L eudiometer tube placed on the digestion bottle via a ground-glass connection. The tube has a glass hose-coupling from which a sufficiently long hose connection leads to a levelling flask. The upper end of the eudiometer tube is fitted with a conical stopcock for adjusting the zero point. The liquid contained in the tube and in the levelling flask was NaCl at pH 3 to avoid CO_2 losses by carbonate formation. The biogas was read daily. At regular time intervals, one digestion reactor containing untreated and one containing sonicated sludge were stopped and sludge analysis was performed.

Analytical procedures

Volatile solids (VS), measured in triplicate, were determined after drying the samples at 105 °C for 24 h to obtain the concentration of dry solids. In the next step, the dry solids were incinerated at 550 °C for 2 h. The residues after incineration represent the inorganic part of the dry solids. The difference between the total dry solids and the inorganic ones gives the volatile solids.

To analyse the soluble phase, the particulate sludge matter was removed by centrifugation (10 min at 6,000 rpm) and the centrate was then filtered through 0.45 μm pore size membrane filters. Soluble COD (chemical oxygen demand), measured in duplicates, was determined by photometric determination of chromate consumption by the organic compounds, subsequent to digestion in concentrated sulphuric acid solution for 2 h at 148 °C by means of COD cell test by Spectroquant Merck (EPA method 410.4).

Results and discussion

VS degradation kinetics

Kinetics of volatile solids degradation has been investigated for untreated and sonicated sludge at F/I ratios in the range of 0.1–2. Anaerobic digestion tests have been carried for 12 d. Table 1 shows VS reduction efficiency.

A beneficial effect of ultrasound pretreatment on the efficiency of VS reduction for all F/I ratios was observed. The kinetics of the hydrolysis, that is the limiting step of the whole digestion process (Eastman and Ferguson, 1981), was assumed to follow a first order kinetic equation:

$$\frac{dXs}{dt} = -k_x Xs \tag{4}$$

where Xs (mg/L) is the anaerobically degradable fraction of the activated sludge, k_x is the hydrolysis constant (d^{-1}) and t is the time.

Table 1 Efficiency of volatile solids degradation (%)

F/I	VS degradation efficiency (%)	
	Untreated	Sonicated
0.1*	25	28
0.5	21	23
1	21	24
2	23	28

*A different sludge stock has been utilised for this test

It is hypothesised that the total VS in the sludge (X_T) to be digested are constituted by a degradable fraction (Xs) and by a non-degradable fraction (X_I) originated by the endogenous biomass decay process. The active biomass in the hydrolysis process is, in the first approximation, assumed equal to the VS content of the inoculum.

The ratio $f_x = Xs/X_T$ for sewage activated sludge is reported in the range 0.3–0.4 (Engelhart, 2002), depending on the operating conditions of the wastewater treatment plant: in our case, the full scale plant is operated at high sludge retention time (SRT = 20 d) so a quite high non-biodegradable fraction is expected in the produced sludge and, consequently, a low f_x (equal to 0.3) was assumed in the kinetic analysis.

Integrating equation (4) it follows that:

$$\frac{Xs_f}{Xs_0} = e^{-k_x t_f} \tag{5}$$

where the subscripts 0 and f indicate the initial and final time, respectively. Considering that $Xs_0 = f_x X_{T0}$, equation (5) can be written as:

$$Xs_f = f_x X_{T0} e^{-k_x t_f} \tag{6}$$

Defining the non-biodegradable fraction of the biomass as $X_I = (1 - f_x)X_{T0}$, and considering that total volatile solids X_T concentration is the sum of the inert and the biodegradable fraction X_S, the VS mass balance referring to the whole digestion time gives for the final VS concentration X_{Tf}:

$$X_{Tf} = X_{T0}\left|1 - f_x\left(1 - e^{-k_x t_f}\right)\right| \tag{7}$$

The VS degradation efficiency $\eta = 1 - \dfrac{X_{Tf}}{X_{T0}}$ can be therefore evaluated by:

$$\eta = f_x\left(1 - e^{-k_x t_f}\right) \tag{8}$$

Kinetic data are well correlated by equation (8) with correlation coefficient R^2 always > 0.96. An example of comparison between experimental data and correlation functions (in the linearised form) is shown in Figure 1 for the test performed at F/I = 0.1.

Figure 2 highlights the beneficial effect of the sonication pretreatment on the hydrolysis kinetics with increases of k_x values from 30 to 250% for all investigated F/I.

The hydrolysis rate values show a decreasing trend with increasing F/I for untreated sludge. This behaviour could be explained considering that by increasing F/I ratio there is an excess of substrate with respect to the available hydrolytic biomass, with resultant hydrolysis rate reduction. The same pattern is observed for the sonicated sludge, except for F/I = 2 that gives the highest k_x: this unexpected value may be due to a synergic

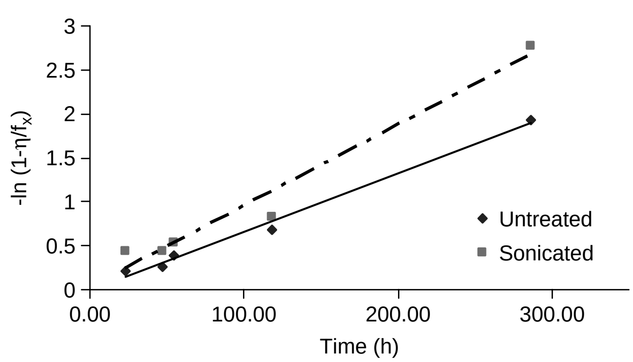

Figure 1 Experimental data and correlation lines for digestion test at F/I = 0.1

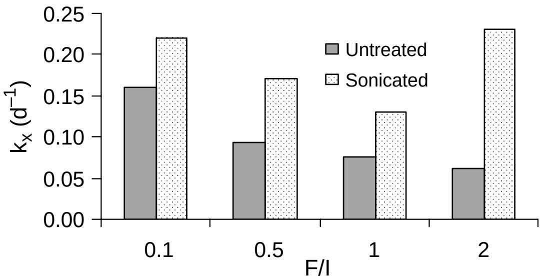

Figure 2 Hydrolysis constant k_x vs. F/I ratio

positive effect of high F/I and sonication, but the result has to be considered preliminary and needs to be verified with additional tests.

Estimated k_x values for non-treated sludge are generally lower than those reported in literature (Table 2), while values of sonicated sludge are well in the range of the above data. It must be pointed out that sampled secondary sludge of our study derives from a very prolonged aeration and therefore it is reasonable that this matrix is hardly digestible.

Soluble COD pattern

During the kinetic tests at F/I = 0.5–2, both soluble and particulate COD were controlled to assess the sonication effects on COD release and on its digestibility. Table 3 shows the soluble COD (CODs) variations during the tests for a digestion time of 12 d.

For untreated sludge at all investigated F/I, soluble COD shows a slight increase after 12 d due to the released CODs, not degradable or not completely removed by the subsequent methanogenic phase, which therefore accumulates in the reactor.

The trend of soluble COD was different for sonicated sludge: the high initial CODs (demonstrating the effectiveness of the ultrasound treatment) was rapidly removed.

Figure 3 shows the CODs profiles for sonicated and untreated sludge at F/I = 2. It must be pointed out that for treated sludge there is a 20% increase of CODs in the first 55 h followed by rapid removal, whereas in the case of the untreated sample, COD remains approximately constant after an initial increase.

Table 2 Hydrolysis constant: comparison with literature data

k_x (d^{-1})	Substrate	Reference
0.22	Activated sludge	Gosset and Belser, 1982
0.15	Sonicated activated sludge	Pavlostathis and Gosset, 1986
0.25	Mixed sludge	Siegrist *et al.*, 1993
0.06–0.17	Activated sludge	This study
0.13–0.23	Sonicated activated sludge	This study

Table 3 Initial and final soluble COD concentration (mg/L) for untreated and sonicated sludge

F/I	Untreated		Sonicated	
	CODs (t = 0)	CODs (t = 12d)	CODs (t = 0)	CODs (t = 12d)
0.5	104	204	981	254
1	177	408	1,605	686
2	117	322	1,890	800

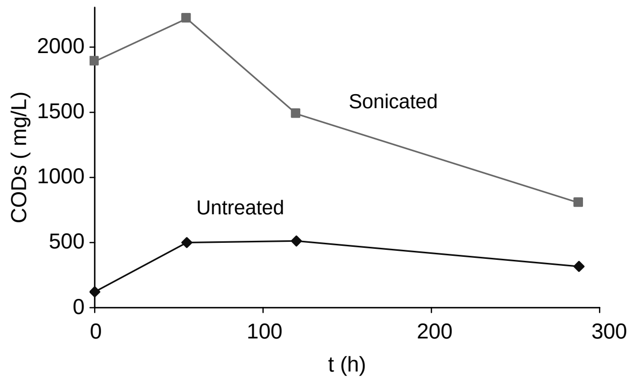

Figure 3 Soluble COD profiles at F/I = 2

The initial accumulation of organic compounds in the case of the sonicated sample at F/I = 2 can be explained by different kinetics of hydrolysis and conversion, with a prevalent effect of hydrolysis in the first phase of the process.

Biogas production

Biogas production of untreated sludge was, as expected, higher for the test with F/I = 2, being higher than the amount of available volatile solids. The cumulative production decreases by decreasing F/I (Figure 4), but the produced biogas volume per unit of degraded substrate remains almost constant. The specific biogas production, for the digestion time of 12 d, was 449 Nm^3/kgVS degraded. This value is consistent with the total COD balance and with the values reported in literature for anaerobic digestion processes on activated sludge that are in the range of 430–650 Nm^3/kg VS degraded (Tiehm *et al.*, 2001; Engelhart, 2002).

As regards sonicated sludge, the biogas production increased by increasing F/I; the highest cumulative production was observed in the test at F/I = 2 (Figure 5), characterised by the greatest amount of available substrate with respect to the inoculum. As for the untreated sludge, the cumulative biogas production is consistent with the total COD mass balance.

Figure 5 shows that in the case of F/I = 2 during the first 55 h, a slowdown in the biogas production occurs and this is attributable to the observed accumulation of soluble organic substances suggesting a kinetic discoupling between hydrolysis (i.e. solubilisation of organics) and methanogenesis (i.e. biogas production). Nevertheless, in the second phase of the test, the biogas production rate increases and overtakes the other ones. The specific biogas production remains comparable (280 $Nm^3/kg_{VSdegrad}$) by varying the F/I. The production results, however, are lower compared to the untreated control, and this effect might be due to different biodegradability characteristics of the COD fractions

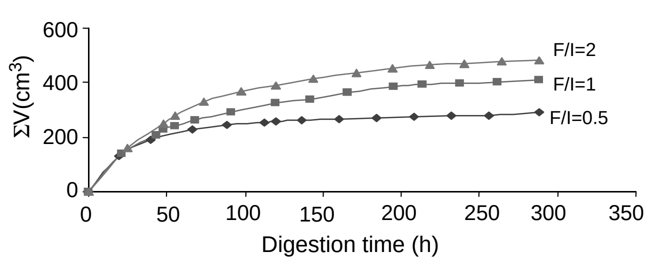

Figure 4 Cumulative biogas production at different F/I with untreated sludge

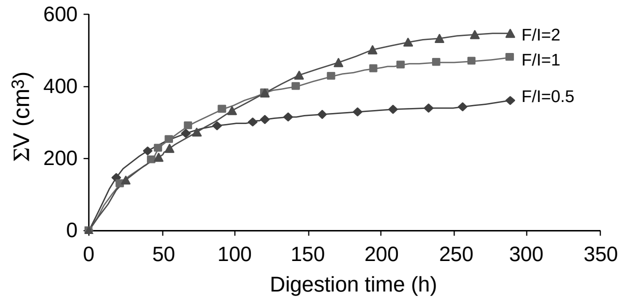

Figure 5 Biogas produced in the tests at different F/I with sonicated sludge

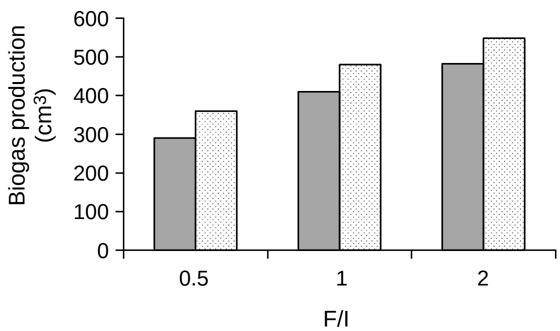

Figure 6 Cumulative biogas production vs. F/I ratio

present in the sonicated sample that involve different biochemical fermentation pathways (Tiehm *et al.*, 2001).

From the energy point of view, sonication pretreatment had a positive effect (see Figure 6) for all investigated F/I. The more convenient gain (25%) was obtained for the test at high inoculum (F/I = 0.5) while in the test with low inoculum (F/I = 2) the untreated sludge produced even more biogas in the first 5 d with respect to the sonicated one, then the "overtaking" occurred but only a difference of 14% was detected at the end of the test.

Conclusions

The comparison between batch anaerobic digestion tests carried out with untreated and sonicated sludge showed a beneficial effect of the pre-treatment by ultrasound, both on hydrolysis kinetics and biogas production for all the investigated F/I.

For untreated sludge, VS degradation data are well correlated by a first order kinetic equation: the hydrolysis rate values, in the range of $0.06-0.17\,d^{-1}$, show an increasing trend with decreasing F/I. Soluble COD measured after different digestion times did not change significantly for tests with F/I = 0.5 and 1, whereas a small increase in the first 55 h was monitored in the test with F/I = 2.

In the tests with sonicated sludge, the first order kinetics of VS degradation was confirmed. It is worth noting that ultrasound pre-treatment considerably accelerates the reaction rate (k_x values are in the range $0.13-0.23\,d^{-1}$) and this effect is more evident with F/I = 2. The trend of soluble COD during the digestion process was basically different from that observed for the untreated sludge. The soluble COD, initially very high (so demonstrating the effectiveness of the ultrasound treatment), was rapidly removed during the first days.

For all F/I tested, biogas production was higher for sonicated sludge, and the more convenient gain (25%) was obtained for the test at high inoculum (F/I = 0.5) where no biogas slowdown due to kinetic decoupling was observed, thus allowing a major gain in biogas production with respect to untreated sludge.

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