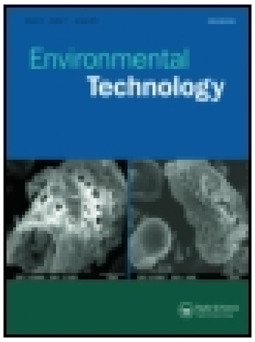

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Abstract

To increase the production of methane, when cattle manure is digested, pretreatments can be applied and/or the manure can be co-digested with other wastes. In this research work, a mixture of cattle manure (CM), food waste (FW) and raw glycerine (Gly) in a proportion in weight of 87% CM, 10% FW and 3% Gly was digested, a) without pretreatment and b) with pretreatment by ultrasound, applying a sonication energy of 1,040 kJ/kgTS. Specific methane production was 290 L CH_4/kg VS without pretreatment and 520 L CH_4/kg VS with pretreatment. With respect to the volumetric methane production, 1.07 L $CH_4/L_{reactor}$.day was produced in the first case, and in the second case, 1.98 L $CH_4/L_{reactor}$.day. We can conclude that the application of ultrasound pretreatment significantly improved the production of biogas.

Keywords: biogas; cattle manure; food waste; raw glycerine; ultrasound

1. Introduction

According to the Directive 2009/28/EC, the use of manure and other organic wastes to produce biogas can contribute significantly to sustainable development in rural areas. However, the biogas obtained by anaerobic digestion of cattle manure is not high enough for the economic viability of farm-scale plants.[1] Specific methane yields of 148-185 L CH_4/kg Volatile Solids (VS), 166 L CH_4/kg VS and 215 LCH_4/kgVS were reported by Castrillón et al. [2], Amon et al. [3] and Zarkadas et al. [4], respectively. To increase the production of biogas, pretreatments can be applied and/or the manure can be co-digested with other wastes to make the process profitable.[5-6]

A possible pretreatment is the application of ultrasound. This pretreatment has been used especially for sewage sludge.[7-10]

To a lesser extent, it had been used as pretreatments to other substrates, such as, meat processing effluent [11]; in this case, the specific energy applied was very high, 120,000 kJ/kg TS, to produce a 24% increase in methane production with respect to the non-pretreated effluent. The effect of ultrasound pretreatment (6,000 ± 500 kJ/kg TS) on the biological methane potential of dairy cattle slurry (containing 5.8% TS and 4.4% VS) was studied by Luste and Luostarinen [12]. They obtained an increase of 19% in methane production (250 ± 10 L CH_4/ kg VS) with respect to the untreated slurry. To enhance the anaerobic biodegradability of a mixture of sewage sludge and the organic fraction of municipal solid waste, Cesaro et al. [13] studied the effectiveness of ultrasound (90,000 kJ/kgTS), finding that, after 45 days, 24% more biogas was produced during anaerobic co-digestion compared to the untreated mixture. The energy costs of the pretreatment has to be offset by the increase in methane yield, otherwise the pretreatment is not economically feasible.[14]

Our research group has already studied the co-digestion of different mixtures of cattle manure (CM), raw glycerine (Gly) from biodiesel production (both substrates previously pretreated by ultrasound, using and energy of 520 kJ/kg TS) and food waste (FW) at 55ºC in an Induced Bed Reactor (IBR). The best results were obtained in the co-digestion of a mixture of 87% CM, 10% FW and 3% Gly, operating at an organic loading rate (OLR) of 7 kg COD/m^3.day, obtaining and specific and volumetric methane productions of 640 L CH_4/kg VS and 2.6 m^3 CH_4/m^3.day (3.3 m^3 biogas/m^3.day).[15]

It is not known whether these outstanding results are due only to the co-digestion process of these three wastes or if an application of ultrasound pretreatment improves enough to be energetically feasible. Moreover, this paper, aims to check if a higher energy (1,040 kJ/kg TS) could improve it.

The objective of this study was to know what was the influence on biogas production by: a) the addition of raw glycerine and food waste as co-substrates when cattle manure is anaerobically treated, and b) the implementation of ultrasound irradiation (520 and 1,040 kJ/kg TS) to cattle manure and raw glycerine prior to co-digestion with food waste (87% CM, 10% FW and 3% Gly).

2. Materials and methods

2.1. Materials

Cattle manure was collected from the cesspit of a dairy farm with 120 livestock units after stirring the contents of the cesspit. After that, it was stored in the laboratory at 4°C (for no more than three weeks). The food waste was collected from a local retirement home.

The crude glycerine was obtained from a local industrial plant which produces biodiesel from used vegetable oil. The major components of crude glycerine were: pure glycerine, soap (sodium oleate), glycerol ester and methanol, with percentages of around 46.4%, 30%, 9.3% and 7.8% (w/w), respectively. The COD of the crude glycerine was very high, around 1250 g/kg.

2.2. Equipment employed

The manure was ground using a domestic triturator and the food waste using an industrial STR-2000 triturator. The ultrasonic device used was a Hielscher UPS 400S (400 W, 24 kHz). Anaerobic co-digestion was carried out in an IBR, with a useful volume of 18.75 L.[15] The operating temperature was kept constant at 55±1ºC.

2.3. Analytical methods

The parameters analysed to characterise the three types of waste and monitor the performance of the reactors were: pH, total solids (TS), volatile solids (VS), chemical oxygen demand (COD), total nitrogen (TN), ammonium nitrogen (NH_4^+-N), total phosphorus (TP) and volatile fatty acids (VFA). The protocol described in Castrillón et al. [15] was followed in this work.

Samples from the reactor (digestate and biogas) were taken twice a week to monitor the biodegradation process. The volume of gas produced was measured daily using a HI-TEC F101D thermal effect mass gas flow apparatus equipped with an electronic totalizer. All gas volumes reported have been corrected to standard temperature and pressure (0ºC, 101.3 kPa). The methane and carbon dioxide contents of the biogas were determined on an Agilent gas chromatograph using a TCD detector and a Porapack N packed column plus a molecular sieve, employing the following temperature ramp: starting temperature 35ºC (1.5 min), increasing up to 55ºC at a rate of 1.5 ºC/minute.

2.4. *Experimental methods*

Bearing in mind the results obtained in previous studies [15], the co-digestion of mixtures of 87% CM, 10% FW and 3% Gly was carried out at 55ºC. The percentages refer to mass fractions.

Prior to co-digestion, ultrasound was applied to the mixture of CM and Gly at a specific energy of 1,040 kJ/kg TS. As previously stated, this energy was chosen to compare the biodegradability and methane yield with previous results obtained when applying a lower energy, 520 kJ/kg TS.[15] Once the sonication energy (Es) was selected, the sonication time was calculated using the expression: Es = (P . t)/(V TSo) (kJ/kg TS), where P is the microwave irradiation power (kW), in our case 0.4 kW; t is the time (s) of exposure of the sample to microwave irradiation; V is the volume (mL) of the sample treated; and TSo is the initial Total Solids.[16]

After the ultrasound treatment, FW was added and the mixture was stirred for 3-5 minutes to obtain a homogeneous mixture to be fed into the reactor. Co-digestion without previously applying ultrasound was also studied under similar operating conditions for the sake of comparison.

It was not necessary to inoculate the digester as it had previously been used in the thermophilic anaerobic co-digestion of a similar mixture of CM, FW and Gly. The digester was fed once a day. The hydraulic residence time (HRT) was 20 days in all experiments, which supposes an OLR of around 7 kg COD/m^3.day (3.6-3.8 kg VS/m^3.day).

Total biogas produced per day, biogas composition and COD, solids and volatile fatty acids were determined.

3. Results and Discussions

3.1. Influence of the ultrasound pretreatment

3.1.1. Removal efficiencies

Table 1 shows the physicochemical characteristics of the influents of the reactor when not ultrasound was applied to the co-substrates and when applying 1,040 kJ/kg TS. This table also includes the characteristics of the effluents once steady-state conditions were achieved (constant biogas production and constant effluent COD and VS).

The pH remained stable throughout the digestion process, around 7.3-7.4, and there was no need to add alkalinity to the digester. The simultaneous presence of ammonia and bicarbonate in the digester results in the formation of a buffer system.[17]

Ultrasound pretreatment increased biodegradation. COD removal increased from 84.8% to 90.9% and VS removal varied from 83.4% to 83.6% when 1,040 kJ/kg TS were applied.

Although VFA concentrations in the effluents were low during all experiments, acetic acid, propionic acid and isobutiric acid were detected when no ultrasound pretreatment was applied. In the case of applying a specific energy of 1,040 kJ/kg TS, only acetic acid and propionic acid were detected.

3.1.2. Methane production

Figures 1 and 2 show the influence of the pretreatment on specific and volumetric methane productions throughout the study. Table 2 shows the methane yields obtained in the co-digestion of the mixture without pretreatment or when ultrasound pretreatment was applied. The maximum methane yield was obtained once steady-state conditions were achieved in the reactor. When ultrasound irradiation at 1,040 kJ/kg TS was compared to no pretreatment, the pretreatment upgraded the specific methane production from 290 to 520 L CH_4/kg VS (79%) and the volumetric methane production from 1.07 versus 1.91 L CH_4/L reactor·day (78%).
In order to enhance the economic viability of an anaerobic digestion system, more than 30 m^3 biogas per ton waste must be achieved.[4] In this study, 34.1 m^3 biogas per ton waste was achieved without pretreatment. When 1,040 kJ/kg TS were applied, the biogas produced was 51.6 m^3 biogas per ton of waste. Moreover, the methane content in the biogas increased from 62.7% in the untreated waste to 70.2% when applying 1,040 kJ/kg TS. Braeutigam et al. [18], had obtained similar behaviour, the methane content in biogas increased from 66.9% when the waste (chicken manure) was not pretreated by ultrasound, to 70.4% when it was pretreated. These results clearly indicate that methanogenic activity is enhanced by the application of ultrasound.

3.1.3. Comparison of results and energy balance

If the results presented in this paper are compared with those obtained in a previous work [15], it can be noted that when a lower energy was applied (520 kJ/kg TS), the process performance was improved (92.7% COD removal and 86.8% VS removal) and concerning VFA, acetic acid was only detected with values of around 87 mg/L. With respect to the methane production, an increase of 120% in the specific methane production (640L CH_4/kg VS *versus* 290 L CH_4/kg VS) and 136% in volumetric methane production (2.6 *versus* 1.1m^3 CH_4/m^3.day) with respect to the untreated substrate was achieved. When 520 kJ/kg TS were applied, 58.2 m^3 biogas per ton of waste were produced (51.6 m^3 biogas per ton of waste when 1,040 kJ/kg TS were applied). It can be assumed that the application of a higher specific energy did not achieve greater efficiencies. This may be related to the fact that, at short sonication times (i.e., lower specific energy), floc deagglomeration occurs without the destruction of bacteria cells.[7] Cattle manure is used as inoculum in the anaerobic treatment. Thus, the reduction in the number of cells introduced in the process could lead to lower biogas productions (i.e. a less efficient process).

Table 3 shows the net energy produced (energy produced as biogas minus energy consumed in sonication). As can be seen, the net energy obtained doubled when applying low energy ultrasound to the substrates (1,622 kJ/kg fed substrate *versus* 796 kJ/kg fed substrate).

Furthermore, the application of low energy ultrasound leads to a slight increase in temperature, reaching values around 30ºC, therefore, less energy is needed to heat the substrate up to the digestion temperature.

As stated before, the increases in biogas production after sonication found by other researchers [11-12,19] were lower than those obtained in this study; nevertheless, higher sonication energies were usually applied.

4. Conclusions

Application of ultrasound pretreatment to a mixture of cattle manure and crude glycerine prior to the co-digestion of that mixture with FW (87% CM, 10% FW and 3% Gly) enhances the biodegradability and the viability of biogas plants (COD removal higher than 90% were obtained).

A specific energy, around 1040 kJ/kg TS, increases specific methane production of said mixture around 70% with respect to untreated one. A higher increase in methane yield (120%) was achieved when applying a lower energy value (520 kJ/kg TS).

The overall results indicated that applying ultrasound pretreatment with the lowest specific energy (520 kJ/kg TS) is sufficient to increase COD and VS removal and, subsequently, biogas production.

The net energy obtained doubled when applying low energy ultrasound (520 kJ/kg TS) to the substrates (1622 kJ/kg fed substrate with sonication *versus* 796 kJ/kg fed substrate without sonication).

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Table 1. Physicochemical characteristics of the influents and effluents of the reactor for the non-sonicated and sonicated waste

$E_{sonication}$ (kJ/kg TS)	0		1,040	
HRT (days)	20		20	
OLR (kg COD/m^3.day)	6.4		7.0	
OLR (kgVS/m^3.day)	3.6		3.8	
	Influent	Effluent	Influent	Effluent
pH	7.3 ± 0.1	7.3 ± 0.1	7.3 ± 0.1	7.4 ± 0.1
COD (g/kg)	128.5 ± 6.2	19.5 ± 5.1	139.8 ± 5.3	12.7 ± 3.8
$COD_{removal}$ (%)	84.8		90.9	
TS (g/kg)	105.3 ± 1.0	19.1 ± 3.8	103.9 ± 1.4	18.7 ± 1.3
VS (g/kg)	72.5 ± 1.1	12.5 ± 1.1	75.4 ± 1.1	12.3 ± 0.9
$VS_{removal}$ (%)	83.4		83.6	
Total N (g/kg)	1.8 ± 0.6	1.80 ± 0.3	1.9 ± 0.3	1.8 ± 0.2
Acetic acid (mg/L)	830 ± 56	101 ± 21	727 ± 35	115 ± 4
Propionic acid (mg/L)	1088 ± 99	87 ± 8	1135 ± 106	103 ± 8
Isobutyric acid (mg/L)	75 ± 30	12 ± 3	62 ± 11	n.d.
Butyric acid (mg/L)	90 ± 1	n.d	97 ± 9	n.d.
Isovaleric acid (mg/L)	92 ± 6	n.d	11 ± 3	n.d.
Valeric acid (mg/L)	97 ± 5	n.d	85 ± 4	n.d.

n.d.: not detected

Table 2. Comparison of the methane yield: without sonication and with sonication (1,040 kJ/kg TS)

Es (kJ/kg TS)	0	1,040
HRT (days)	20	20
OLR (kg COD/m^3.day)	6.4	7.0
OLR (kg VS/m^3.day)	3.6	3.8
L CH_4/kg VS	290	520
L CH_4/kg wet waste	21.4	36.2
L CH_4/$L_{reactor}$.day	1.07	1.91
% CH_4	62.7	70.2

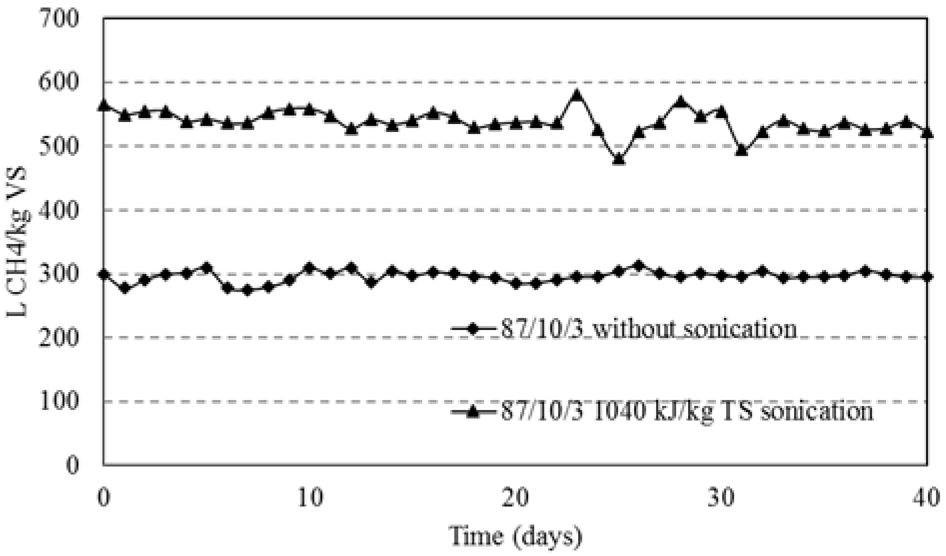
Table 3. Energy balance in the anaerobic co-digestion of CM+FW+Gly with and without ultrasound pretreatment

Energy consumed for sonication	kJ/kg TS	0	520	1,040
	kJ/kg fed substrate	0	54.2	108.4
Energy produced	kJ/kg fed substrate	796	1676	1511
Energy balance	kJ/kg fed substrate	796	1622	1457

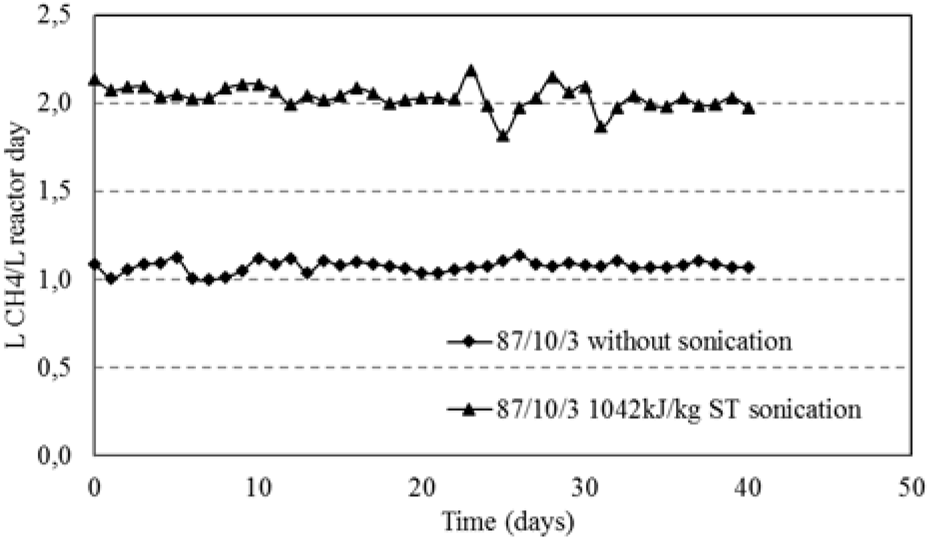
Figure Captions

Figure 1. Specific methane production in the thermophilic co-digestion of CM + FW+ Gly with and without pretreatment by ultrasound

Figure 2. Volumetric methane production in the thermophilic co-digestion of CM + FW+ Gly with and without pretreatment by ultrasound



700
600
500
400
300
200
100
0
L CH4/kg VS
0
10
20
30
40
Time (days)
87/10/3 without sonication
87/10/3 1040 kJ/kg TS sonication



2,5
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1,5
1,0
0,5
0,0
L CH4/L reactor day
0
10
20
30
40
50
Time (days)
87/10/3 without sonication
87/10/3 1042kJ/kg ST sonication