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Bio-methanization of organic fraction from municipal solid waste: temperature effects

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The main objective of this study was to analyse the development of dry anaerobic digestion process of OFMSW in batch reactors under two temperature ranges, thermophilic (55°C) and mesophilic (35°C). The experimental results lead to the conclusion that the thermophilic range has a greater rate of hydrolysis and is therefore more effective to degrade wastes, shortening the overall operating time. For example, the hydrolytic step in the thermophilic (T) process lasted an average of 8 days versus 14 days in the mesophilic (M) range. The methanogenic phase lasted for 18 and 29 days in the T and M processes, respectively. The mesophilic range showed higher removal of the organic effluent but with greater uptime requirements. Moreover, the thermophilic range showed greater productivities than the mesophilic range, and the productivities were approximately doubled in terms of the produced biogas from a given amount of consumed organic matter.

Keywords: anaerobic digestion, OFMSW, mesophilic, thermophilic, methane.

INTRODUCTION

Municipal waste production in Spain has increased from 18783442 tons (in the year 1998) to 24163199 tons (in 2003), which represents an increase of 28.64% during this period[1]. A 40–45% is the fraction of organic nature in the waste, known as OFMSW (Organic Fraction of Municipal Solid Waste). The OFMSW can be managed through biological treatment; in fact, Spain treated approximately 8.5 million tons of waste in biological treatment plants in 2006[2].

The increase in the generation of municipal waste has involved legal measures to minimise the negative effects on the environment. To this end, the current legislation sets a target of reducing the amount of Biodegradable Municipal Waste (BMW) sent to landfills by proposing and encouraging different methods of recovering such waste, among which is the process of biomethanisation or anaerobic digestion[3].

Anaerobic digestion has been considered to be the most economically viable option for the treatment and recycling of OFMSW and produces methane and generates a digested waste similar to compost or aerobically produced digestate[4]. The process is conditioned by certain operating variables such as the solids content of the waste feed, the solids retention time and the temperature.

Microorganisms are classified according to the temperature range in which they develop[5]: psychrophilic (T < 20°C), mesophilic (20 < T < 45°C) and thermophilic (T > 45°C). Operation at the mesophilic range (33–37°C) is more stable[6] and requires less energy expenditure. In addition, the advantages of the mesophilic range are a lower risk of inhibition by ammonium[7, 8] and by long chain fatty acids[9]. The thermophilic range is also associated with operational advantages. The temperature increase leads to higher metabolic rates and increases the production of biogas and methane[10].

In terms of the content of solids in the reactor, the process can be carried out in wet conditions (4–10% Total Solids-TS) or in dry conditions (15–30% TS). Focus on dry conditions, several studies[11, 12] compared systems operating at 30% TS and 20% TS in mesophilic anaerobic digestion of OFMSW. In accordance with the obtained results, the 30% ST showed a higher production of biogas and removal of organic matter and lower duration of the hydrolysis step, which is the limitation of the process. Additionally, a clear influence of solids on the methanogenic activity was observed[13].

This paper aims to compare the development of the dry anaerobic digestion process of OFMSW in batch reactors using two temperature conditions: thermophilic (55°C) and mesophilic (35°C).

MATERIALS AND METHODS

To study the effect of the temperature on the degradation of OFMSW and considering the results obtained in previous studies, an experimental plan was developed. The scheme consisted in an anaerobic biodegradability test for OFMSW under mesophilic and thermophilic conditions at 20% TS. Furthermore, studies at both temperatures have been carried out in duplicate to verify the reproducibility of the data.

Waste characterisation

The wastes used in this work were as follows:

– OFMSW was used as a source of organic matter. This waste was obtained from the recycling and composting plant "Las Calandrias" in Jerez de la Frontera, Spain.

– Mesophilic digested sludge was obtained from a Waste Water Treatment Plant (WWTP) and effluent was obtained from a thermophilic anaerobic reactor, which operated stably in the degradation of OFMSW as inocula. The digested sludge was obtained from the WWTP Guadalete, which is also located in Jerez de la Frontera, Spain. The T reactor effluent was obtained from the semi-stabilised Research Group Biological Waste, University of Cádiz. This group has extensive and proven experience in the Thermophilic Anaerobic Digestion of Organic Waste.

Initially, both thermophilic and mesophilic reactors were loaded with 30% (w.w) of inoculum and 20% (w.w) of OFMSW, because of the greater production of biogas[11] and an improvement in the values of the kinetic parameters[12].

The physico-chemical characterisation of the waste and the initial mixtures used in the two processes, thermophilic and mesophilic, are shown in Table 1.

In order to characterise the waste and the inoculum, as well as to monitor the effluent of the process, the following parameters were analysed: pH, volatile fatty acids (VFAs), dissolved organic carbon (DOC), total solids (TS) and volatile solids (VS). These analyses were conducted in accordance with standardised methods[14] adapted for waste with high solids content and based on the previous leaching of the waste in an aqueous medium. The biogas volume and composition were determined using chromatographic methods.

Experimental equipment

The experimental equipment used in these tests in a batch system were designed and patented by the Biological Treatment of Waste research group from the University of Cádiz[15]. The experimental setup consists of a battery of 6 reactors, submerged in a thermostatic bath with a capacity of 45 L (Fig. 1). This bath has an electrical panel that allows for the independent operation of each reactor.

Each reactor consists of a 2 L stainless steel vessel with a total usable volume of 1.7 L. The used equipment is equipped with an agitator, to maintain homogeneity, a closure system that tightly secures the contents and various holes that allow for sampling at specific times.

The biogas is collected into the bags made of Tedlar material, a plastic polymer (polytetrafluoroethylene) with a low porosity and permeability to H_2. The bags are equipped with a connection valve and a septum that allows the sample gas to be collected from the interior. The samples are collected using a gas syringe of 1 mL (Dynatech Gastight).

RESULTS

The monitoring of the thermophilic and mesophilic anaerobic degradation processes involved the determination of the following control parameters: pH, soluble chemical oxygen demand (CODs), dissolved organic carbon (DOC), total acidity, alkalinity, and daily production of biogas, accumulated methane and biogas composition. In addition, the stages of the process have been studied for its characterisation and comparison of the two temperature ranges.

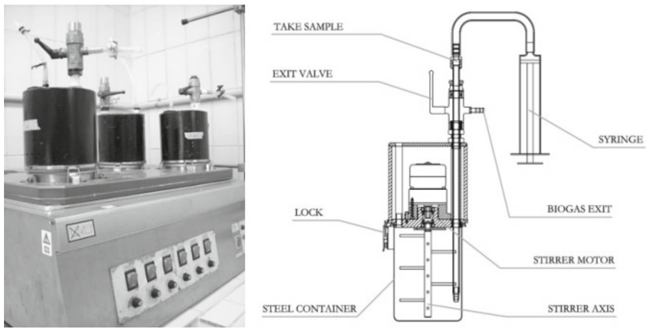

Figure 1. Batch type anaerobic reactors: Photograph (left) and schematic (right)

Monitoring of the thermophilic and mesophilic processes

The pH is a variable used in the monitoring of anaerobic systems that affects different chemical equilibriums in the reactor. In the present study, during the first days a decrease of pH was observed due to the hydrolysis of the waste in the hydrolytic step and the generation of acids in the acidogenic stage (Fig. 2). After this decrease, an increase in the pH was observed until day 25 in the M reactor, and this parameter stabilised at a pH of approximately 8. In the T reactor, the increase in pH occurred until day 18 and achieved stability at a pH of approximately 7.6. The difference observed in the pH value is therefore determined by the temperature regime studied.

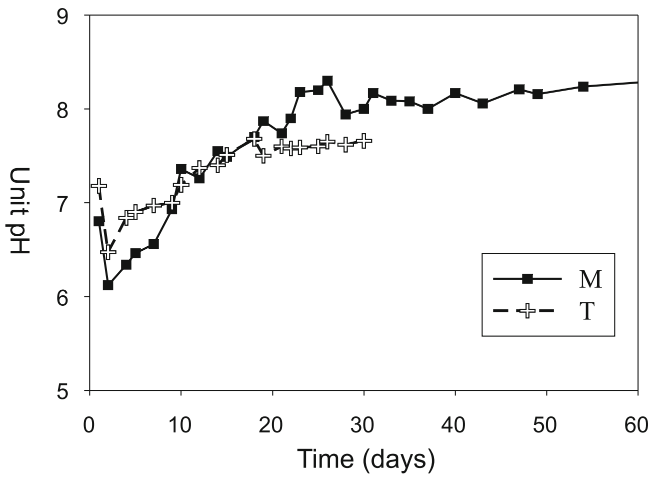

Figure 2. The pH evolution in the mesophilic (M) and thermophilic (T) reactors

Table 1. Physical-chemical characterisation of the waste, the inoculums and the initial content of the different processes: mesophilic (M) and thermophilic (T)

	OFMSW M	OFMSW T	Inoculum M	Inoculum T	Digester M	Digester T
pH	6.51	6.59	7.49	7.84	6.68	6.87
Density [kg/L]	0.666	0.638	0.971	0.965	1.035	1.029
TS [%]	82.34	81.09	3.64	4.92	16.61	18.32
VS [%]*	29.89	26.76	1.85	3.87	10.71	11.66
COD_s [mg O_2/L]	1862.54	1898.23	1134.12	794.15	1259.19	1552.77
DOC [ppm]	1283.5	1105.56	662.15	593.45	931.1	847.4
Total N [g N/L]	27.43	21.79	0.44	0.35	–	–
Ammonia N [mg NH_3-N/L]	10.93	10.07	12.15	13.98	–	–
Total Acidity [mg AcH/L]	301.2	306.3	19.24	256.87	–	–
Alkalinity [mg Ca CO_3/L]	14.0	17.3	0.64	1.13	9.2	9.9
Total P [g P/kg $_{dry\ weight}$]	6.07	5.39	–	–	–	–

* VS expressed as % of total sample weight.

The CODs of the batch processes show significant differences depending on the operating temperature. Figure 3 illustrates the evolution of this parameter for the two tested conditions (T and M). Although this parameter has a large scatter, mainly due to the complexity and heterogeneity of the waste, we can observe an increase in the early days due to the solubilisation of organic matter in the solid waste. The duration of this increase depends on the temperature range studied. The increase occurs during the first 10 days in the thermophilic process and during the first 22 days in the mesophilic process. The final concentrations of the organic matter (measured as CODs) are lower in the M reactor and reaches values close to 300 mg/L, representing a decrease of 90.62% in the methanogenic stage. In the T reactor this decrease was apparently lower, although it is noted that the operating time was different for both systems. Thus, for the same time of the operation, the thermophilic and mesophilic systems reach final values and percentages of organic content of purification at the same order of magnitude.

The DOC evolution (Fig. 4) exhibits a more definite trend that the CODs evolution. The hydrolysis and acidogenesis periods are similar for both processes in respect to this parameter. Thus, we observe an increase in the soluble organic matter during the first 15 days of the process in the mesophilic range and the first 9 days for the thermophilic range. From that time, all of the biodegradable organic matter starts to be consumed. The hydrolysis and acidogenesis phases result in 70.11% reduction of the DOC in the M reactor (operation day 60) and 49.34% reduction of the DOC in the T reactor (operation day 30). For the same time of operation, for approximately 22 days, both systems achieve similar removal rates, but the rates are slightly higher in the mesophilic system.

The hydrolysis and acidogenesis stages can be observed in Figure 5, which shows the evolution of the total acidity. The value of this parameter increases until day 15 in mesophilic conditions and until day 10 in thermophilic conditions. Thereafter, there is acid consumption by acetoclastic methanogenic archaea. The removal in the M reactor was 98.33% and 78.43% in the T reactor until the end of the process. If we consider the same operation time, the percentage of removal is in the same order in both processes.

Regarding the ratio total acidity/alkalinity (Fig. 6), there are large differences between the mesophilic and thermophilic systems. In the mesophilic system, this ratio reaches the values between 0.15 and 0.4, increasing during the first 15 days due to the generation of acids in the acidogenic stage. It declines until day 60, coinciding with the end of the methanogenic stage, until the values less than 0.01 are reached. This decrease was associated with the use of acids at this stage.

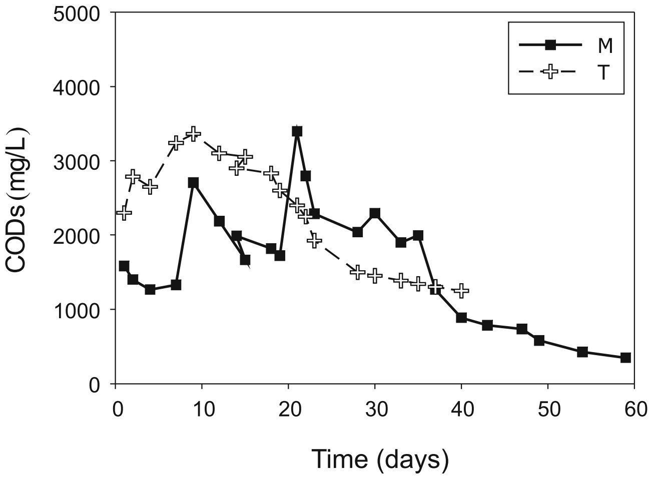

Figure 3. The CODs evolution in mesophilic (M) and thermophilic (T) reactors

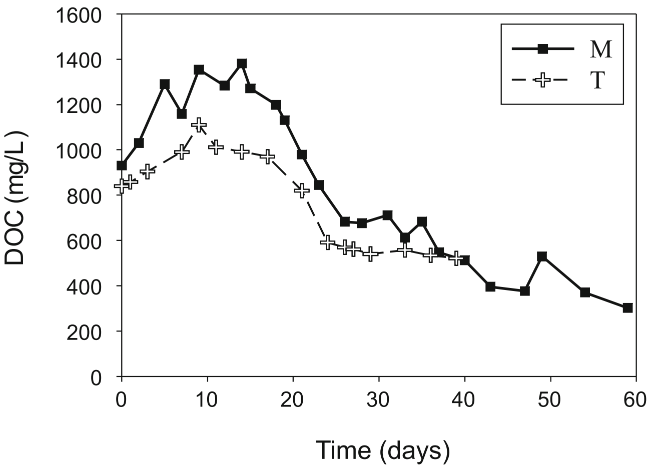

Figure 4. The DOC evolution in the mesophilic (M) and thermophilic (T) reactors

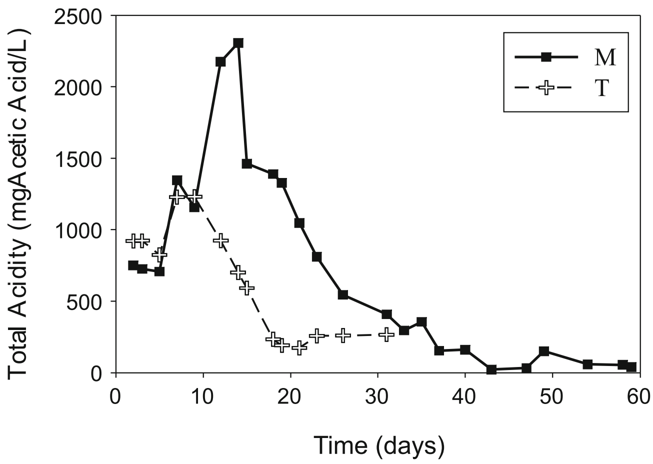

Figure 5. The total acidity evolution in mesophilic (M) and thermophilic (T) reactors

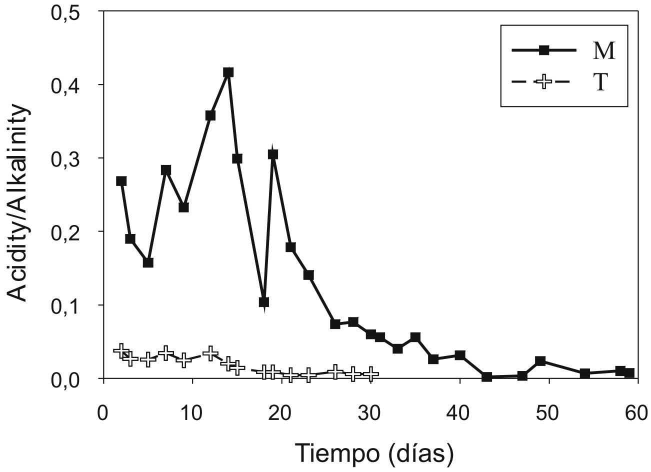

Figure 6. The acidity/alkalinity evolution in mesophilic (M) and thermophilic (T) reactors

For the thermophilic system, this ratio remains constant during the hydrolysis and acidogenesis stages at approximately 0.03. With the onset of methanogenesis (day 10), the ratio begins to decrease to values below 0.01 at day 30 of the experiment.

Monitoring of biogas

The daily production of biogas shows a different evolution depending on the operating temperature (Fig. 7). In the mesophilic process, the methanogenic phase starts on day 15, coinciding with the degradation of organic matter in the system (as seen by the DOC and CODs levels).

In terms of the composition of the biogas produced, H_2 is detected during the first 3 days in the M reactor and during the first 2 days in the thermophilic one (Fig. 8 b), as a result of the hydrolytic activity. The methane production in the M reactor is delayed until day 10 (Fig. 8 a), whereas, as it has been discussed above, in the T reactor, the methane production takes place from the 2nd or 3rd day. The final volume of the accumulated methane per litre is different in each case and is 7.31 L in M and 9.26 L in T (Fig. 9). During the methanogenic stage, the biogas detected presents a high enrichment in methane, between 85 and 90% in both cases.

Overview of the processes

Table 2 shows a summary of the main results obtained in this study regarding the anaerobic degradation of OFMSW at the mesophilic and thermophilic ranges. For a better understanding of the results, the two stages of the process: hydrolysis-acidogenesis and methanogenesis have been distinguished.

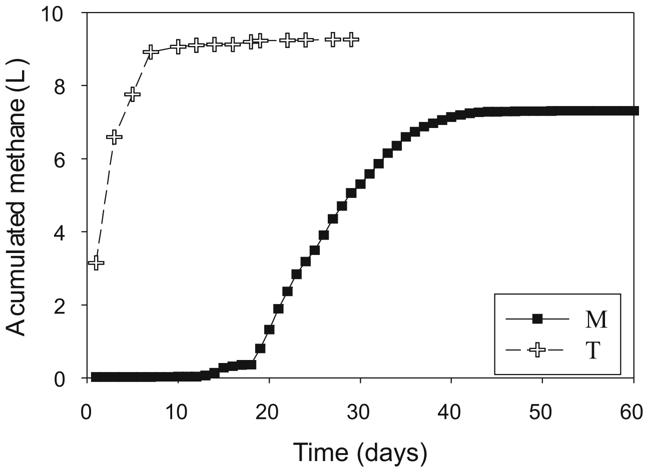

Figure 9. Accumulated methane in the systems M and T

The temporal extension of the hydrolytic step is different for the M and T processes and is 14 and 8 days, respectively. The methanogenic phase lasts for 29 days in M and 18 days in T. The values for the methanogenic stage were calculated based on the overall duration of the process until the end of the consumption of the biodegradable organic matter. This implies that the operating time was different for the two systems, T and M.

The generation of hydrogen was 1.57 L and 0.25 L in the T and M processes, respectively, and methane production was 7.95 L and 0.16 L in T and M processes, respectively.

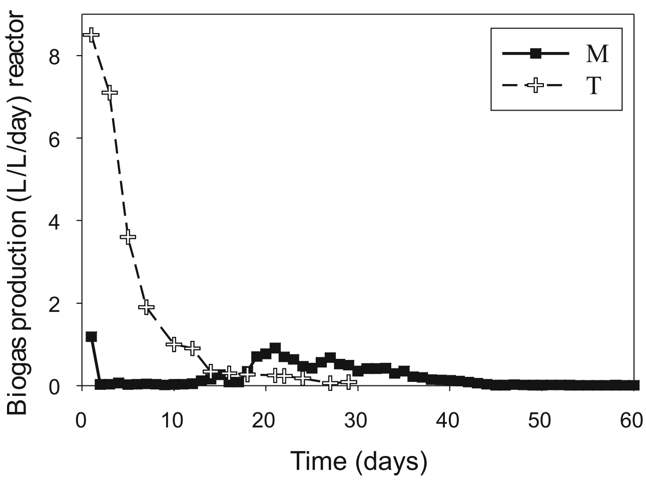

Figure 7. The evolution of the daily production of biogas in mesophilic (M) and thermophilic (T) reactors

DISCUSSION

Analysing the thermophilic and mesophilic processes

The optimum **pH** for the development of methanogenesis in the thermophilic process is usually higher than in the mesophilic process[16]. In this study, we observed that the final pH is slightly lower for the thermophilic process, due to the OFMSW used in both studies was not the same. The differences in the OFMSW is shown in the waste characterisation in Table 1, and the alkalinity was higher in the OFMSW used in the thermophilic

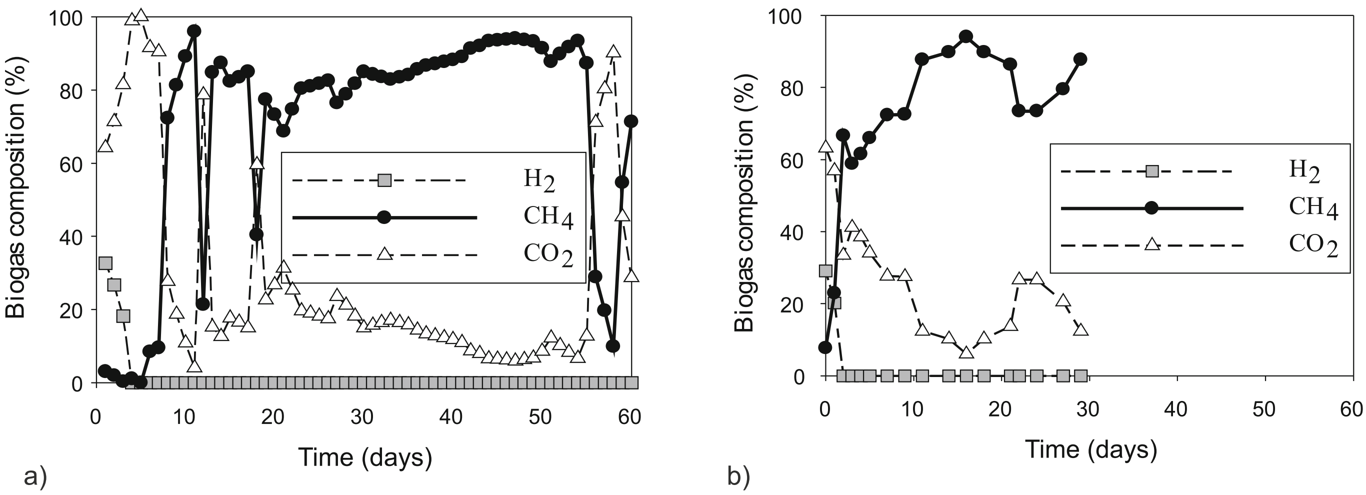

Figure 8. Evolution of the biogas composition in M reactors (a) and T ones (b)

Table 2. Summary of the main results of the batch anaerobic reactors for the degradation of the OFMSW under the two temperature ranges, mesophilic and thermophilic

Hydrolytic-acidogenic stage		
	Average M	Average T
Time [days]	14	8
Production DOC [%]	28.98	32.92
Production CODs [%]	54.88	45.60
Production VFA [%]	183.87	68.60
Production CH_4 [L]	0.16	7.95
Production CO_2 [L]	0.58	3.44
Production H_2 [L]	0.25	1.57
Production LCH_4/L_{react}	0.09	4.68
Methanogenic stage		
	Average M	Average T
Time [days]	29	18
DOC removal [%]	60.19	42.55
CODs removal [%]	76.96	58.88
VS removal [%]	49.53	41.76
VFA removal [%]	97.62	83.23
CH_4 [L]	7.17	1.31
CO_2 [L]	1.63	0.37
H_2 [L]	0.00	0.00
L CH_4/g DOC_{rem}	4.99	1.40
L CH_4/g $CODs_{rem}$	1.53	0.41
L CH_4/g VFA_{rem}	1.95	0.85
L CH_4/g VS_{rem}	0.10	0.68
LCH_4/L_{dig}	4.22	0.77

process compared to that used in the mesophilic process, 17.3 mg $CaCO_3$/L vs. 14.0 mg $CaCO_3$/L. This difference determines the evolution of the pH values.

It can be observed that there is an increase of the hydrolysis rate with temperature, and the results agree with other results obtained by different authors[17, 18]. Furthermore, the heterogeneity of the waste fractions of different biodegradability[19] can explain the evolution of the solubility of the organic matter that would warrant the extra time needed for solubilisation of certain fractions contained in the solid waste to liquid waste. In this sense, there is a fraction that is highly refractory (cellulose, hemicelluloses and lignin) which come from vegetables waste and paper[20]. Therefore, the acclimatisation of inoculum-substrate and the rapid growth of the microbial hydrolytic in thermophilic conditions lead to rapid waste hydrolysis. But, also, it should be noted that the real evolution of organic matter parameters may not have been recorded with the sampling conditions that were established at the beginning of the experiment.

The data suggest that the generation of volatile fatty acids in this case is attached, at least in part. In this sense, their degradation is not seen as a significant contributor to increases in acidity in the both the T and M processes. In any case, it is likely that with an increased sampling frequency more pronounced changes could have been detected. It should be noted that all values of the acidity/alkalinity ratio recorded in this study are within the appropriate limits for the proper development of anaerobic digestion processes[21].

In general, the evolution of the variables that represent the organic content shows the heterogeneity of the waste, with fractions of different biodegradability[19], as it has mentioned above. In addition, an increase in the hydrolysis rate with the temperature is observed, in agreement with the results previously obtained by different authors working with different wastes[17, 22].

Analysing the hydrolysis step

The evolution of the hydrolysis and acidogenesis stages is used to characterise these stages and to predict the behaviour of systems in successive stages. The difference between the start of both processes is distinguished, and the detailed study of the hydrolysis provides information on the anaerobic digestion in both of the temperature ranges.

The duration of the hydrolysis and acidogenesis stages together has shown to be between 7 and 9 days in the thermophilic range, from the starting time of the methanogenesis stage, in which the products generated in the previous stages are consumed to form methane and carbon dioxide. Figure 10 shows a histogram with the percentage increases obtained for the parameters related to the organic content in the reactor: CODs, DOC and VFA (measured as the total acidity). These values have been calculated as the percentage increase over the value of the initial day. The CODs progressively increase until day 11 of the experiment. From day 7, we can see that the hydrolysis rate decreases. The DOC and VFA increase up to day 7, but from this time we can see a decrease, which involves the progressive consumption of the organic matter measured as the DOC and volatile acidity.

The hydrolysis-acidogenesis period in the mesophilic range shows an increase in the parameters (CODs, DOC and VFA) compared to the initial day of testing (Fig. 11).

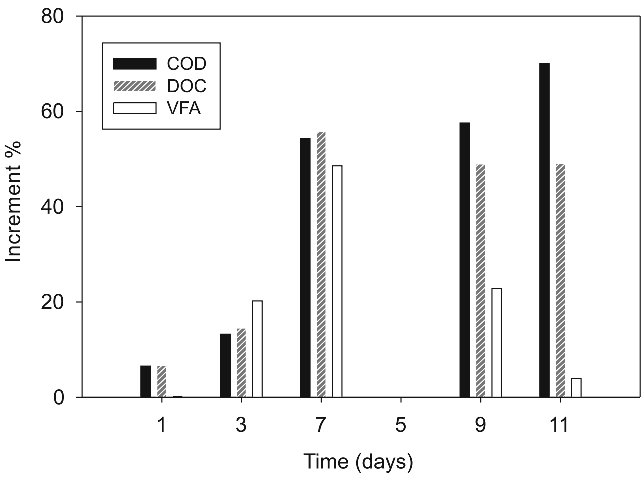

Figure 10. Percentage increases over the initial values for the CODs, DOC and VFA (measured as volatile acidity) for the hydrolysis-acidogenesis during the thermophilic OFMSW

These parameters show the highest rate of increase between 14 and 21 days. The DOC and VFA detect the largest increase on day 14, while for the CODs the largest increase occurs on day 21. In CODs two stages of attack and solubilisation of the waste are observed, where one of them reaches its maximum around day 9 of testing and the second and more pronounced stage is detected on day 21. From these data, it can be seen from the consumption of organic matter solubilised, CODs, DOC and VFA, lead gradually to the methanogenic stage.

Analysing the biogas

In the thermophilic process, a high production of biogas takes place from the first days. This result may be determined by the kind of inoculum used. The meso-

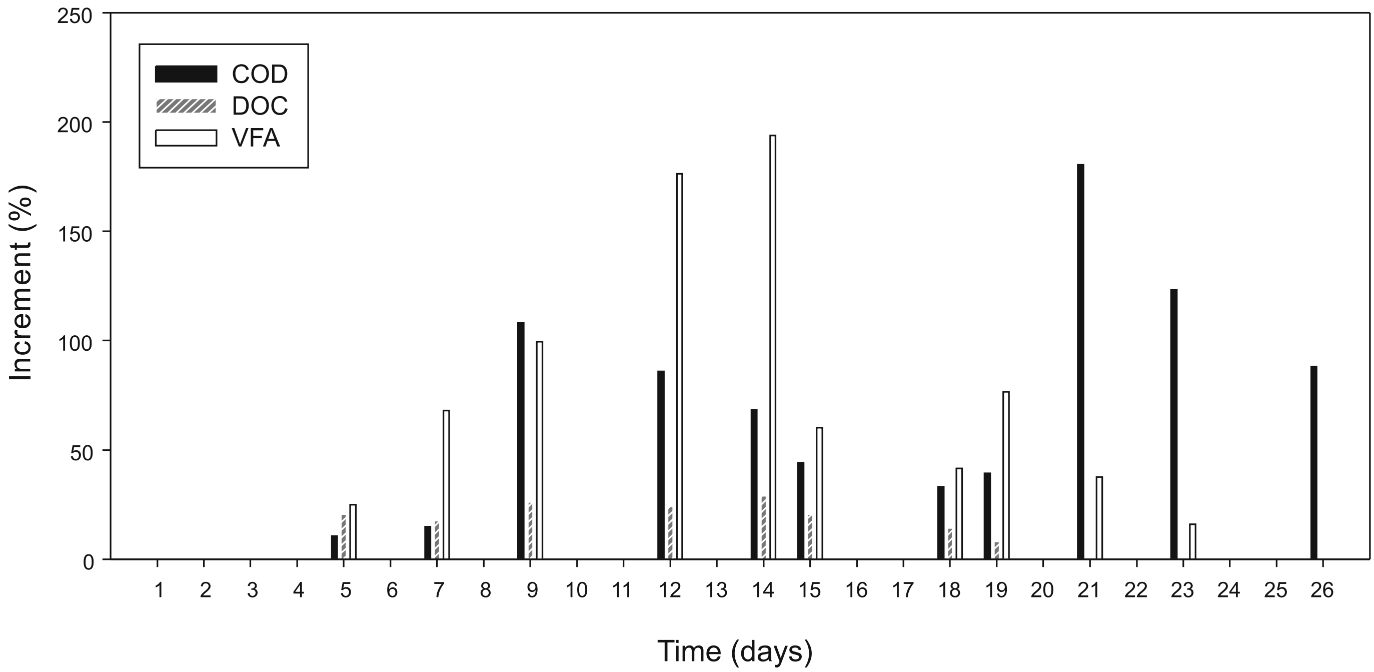

Figure 11. Percentage increases over the initial values of CODs, DOC and VFA (measured as volatile acidity) for the hydrolysis-acidogenesis during the OFMSW mesophilic

philic inoculum was adapted to the degradation of fresh sludge while the thermophilic inoculum was adapted to the degradation of OFMSW.

The good adaptation of the inoculum used in the thermophilic process may have involved the rapid production and consumption of VFA, which is manifested in the high biogas production.

The methane generated by the hydrolytic and acidogenic phases is due to the microbial activity of H_2users because this compound fails to appear in the biogas after this phase. After the hydrolysis-acidogenesis stage, the methanogenic stage begins. The beginning of this stage is later in the case of the M reactor, and as the inoculum is not acclimated to the new waste, the thermophilic process is faster. The beginning of the methanogenic stage can change depending on the selected temperature range.

These results contrast with what has been obtained by other authors[23], who obtained a methane content between 62 and 65 in a digestion between OFMSW and restaurant waste in the mesophilic range.

In the process of methane accumulation, different stages of production rates can be observed. These different stages can correspond to the different fractions of the waste with different biodegradability, as discussed above. Thus, the organisms begin to attack the most biodegradable fractions first, delaying the consumption of the less biodegradable fractions. Related to this, in the M reactor, a turning point in the trend towards day 20 can be observed.

Table 3 shows the methane productivity as the maximum methane detected in the digestion process per gram of DOC and CODs consumed in the overall process, taking the hydrolytic-acidogenic and methanogenic stages together. It is observed that productivity is higher in the thermophilic process; the DOC_{rem} is 9.95 L CH_4/g in the thermophilic process and 5.16 L CH_4/g in the mesophilic process. In the thermophilic process, the productivity is approximately double that of the mesophilic process. When the productivity refers to the $CODs_{rem}$, it is higher in the T, 2.94 L CH_4/g for the T versus 1.65 L CH_4/g for the M; again, the value for the T reactor is approximately double than the M reactor.

Table 3. The global productivity of methane according to consumed amounts of organic matter

Global process		
	M	T
Production CH_4 [L]	7.31	9.26
L CH_4/g DOC_{rem}	5.16	9.95
L CH_4/g $CODs_{rem}$	1.65	2.94
L CH_4/g VS_{rem}	0.073	0.087
L CH_4/L_{react}	4.30	5.45

Analysing the overall process

Analysis of the overall process has resulted in extracting some interesting points. The biogas production during the hydrolytic stage shows higher increments of CH_4, H_2 and CO_2 in the thermophilic digester compares to the mesophilic digester. For the thermophilic process, the acidogenic phase is in conjunction with the methanogenic phase. The methane production in the methanogenic stage is considerably high. This shows that the hydrolysis step is more pronounced in the T process, despite not having been recorded in the parameters representing the organic matter inside the reactor. The hydrolysis step is favoured in thermophilic conditions compared to mesophilic conditions because temperature speeds up the reactions that take place in the anaerobic digestion processes. In addition, the acclimation of the thermophilic inoculum has allowed for a quick start and an efficient process, which has masked the hydrolysis and acidogenesis stages.

On the other hand, an increased consumption of organic matter (measured as DOC, vs. and VFA) in the methanogenic stage occurs at mesophilic conditions, taking into account that the values were determined until the complete biodegradation of the organic matter. When considering the same times for both operating temperature ranges, it is observed that the rate of degradation is of the same order in both temperature ranges. Additionally, the important advantage for the thermophilic process should be considered: the different stages involved are brought forward.

The methane productivity per gram of the organic matter consumed during the methanogenic stage is higher in the mesophilic process because most of the methane generated in the thermophilic process occurs during the hydrolytic stage, as already discussed above. The higher biogas production in the mesophilic process occurs during the methanogenic phase. The parameters L CH_4/g VS_{rem} and L CH_4/L_{react} also show differences in the thermophilic and mesophilic ranges, in both cases resulting in an increased productivity in the thermophilic range[24, 25].

CONCLUSIONS

The process in the thermophilic temperature range supposes an increase in the hydrolysis rate in anaerobic digestion of OFMSW, shortening the time periods of the hydrolytic and methanogenic phases. The duration of these stages fluctuates from 7–9 days under thermophilic conditions versus 14–21 days under mesophilic conditions. Additionally, the thermophilic process achieves high biogas productivity per organic matter consumed in the process; the productivity is almost double the amount that was obtained in the mesophilic process, 2.94 versus 1.65 L CH4/g CODsrem.

The process in the mesophilic range can obtain better quality in the final effluent compared with the process in the thermophilic range, with higher methane production but with a longer operating time, 29 versus 18 days.

The OFMSW is highly heterogeneous, which is manifested in the evolution of the parameters referring to the organic content. In this way, it shows irregular trends according to the composition of the fractions of different biodegradability.

The anaerobic digestion of OFMSW in the thermophilic and mesophilic conditions has shown high biogas production and high removal efficiencies of the organic content. Both ranges of temperature have operational advantages and an integrated system is possible. This integrated system would include an initial thermophilic phase that allows for a greater rate of hydrolysis, followed by another stage for the mesophilic process that allows for a more stable process and lower energy costs.

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GLOSSARY AND NOTATIONS

CODs: Chemical Oxygen Demand soluble, expressed as mg O_2/L,
DOC: Dissolved Organic Carbon, expressed as mg/L,
ERDF: European Regional Development Fund,
L CH_4/g COD_{srem}: L methane per gram of Soluble Chemical Oxygen Demand removed,
L CH_4/g DOC_{rem}: L methane per gram of Dissolved Organic Carbon removed,
L CH_4/g VS_{rem}: L methane per gram of Volatile Solid removed,
L CH_4/L_{react}: L methane per L reactor,
M: Mesophilic temperature,
OFMSW: Organic Fraction of Municipal Solid Waste,
RMB: Biodegradable Municipal Waste,
T: Thermophilic temperature,
TS: total solids, expressed as %,
VFA: Volatile Fatty Acids, expressed as mg/L,
VS: volatile solids, expressed as %,
WWTP: Waste Water Treatment Plant.

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