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Energy recovery from co-digestion of complex substrate mixtures: An overview, and case study of biogas production modeling

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ABSTRACT

Anaerobic digestion processes represent one of the mainstream technologies for indirect energy recovery from waste streams, currently supported in many Countries by governmental incentives to promote renewable sources such as biogas production. The intrinsic variability of supply modes, composition and physical-biological characteristics of the feed mixtures, significantly affect the stability of this type of processes, and may negatively impact on the actual rates of biogas production, affecting the financial feasibility of this type of projects. Composition and pH the generated biogas are important indicators of process performance, and the availability of buffer alkalinity plays an key role in the sustainability methane-generating process steps. In this paper, a theoretical analysis of requirements and applications aimed at addressing critical issues arising during process operation in full-scale plants are reported. The aim of the paper is to support proper assessment methods to identify adequate design and operating conditions of co-digestion systems for the treatment of solid/liquid mixtures of biodegradable waste while maximizing biogas recovery. In order to appropriately assess the influence of organic load on the process, a case study was included to represent possible process conditions typical of Mediterranean Countries.

KEYWORDS

Anaerobic processes;
Biogas;
Co-digestion;
Energy;
Substrate mixtures.

INTRODUCTION

Anaerobic treatment of organic substrates represents at the moment one of the mainstream technologies for indirect energy recovery from waste streams, and must be carefully evaluated from both technical and economic feasibility in the presence of existing production centers of such waste streams, such as food conversion industries, cattle/pig farms, dairy farms, or crop-producing farms with land set to growing energetic crops[1].

In particular, anaerobic digestion is a widely used solution for wet residues treatment with the aim of water pollution reduction and energy recovery. The process converts a large part of COD into biogas (composed by methane) or bio-hydrogen thanks to its high

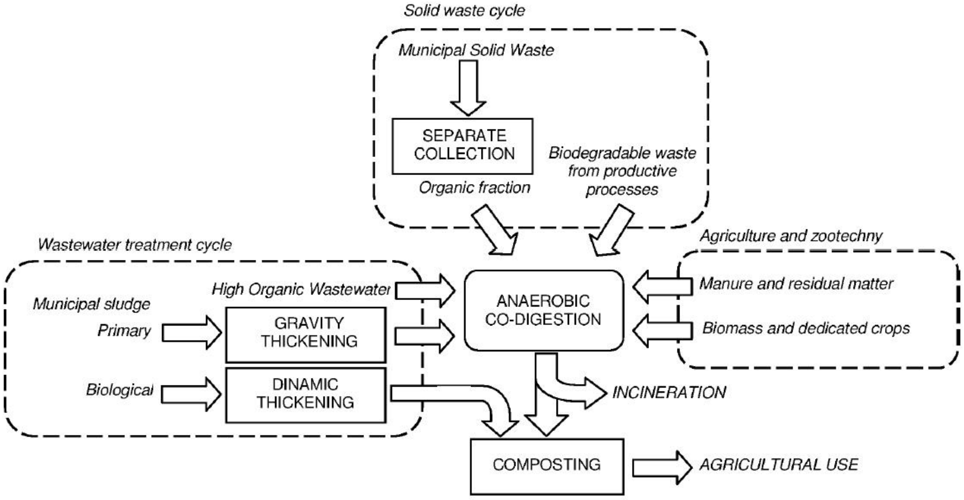


Figure 1 : Role of anaerobic co-digestion in an integrated wastewater/waste management system.

removal efficiency. The anaerobic process has been studied over the years from many points of view. As a result, both conventional and unconventional aspects of such a process are adequately. In order to obtain a good removal of organic matter during anaerobic digestion, it is necessary to properly select the system to be implemented[2-5].

Centralized facilities may allow better recovery yields, in terms of biogas production, better energetic efficiency due to scale effects, and the reduction of operating and investment costs, even if such solutions may more complex to manage from an administrative/organizational aspect. Disadvantages, both in economic and sustainability terms, may be related to the need for transport of biomasses to the facility from greater distances, however, integration of such facilities into the regional water and waste cycles may ensure the consistent availability of adequate organic loads to the process.

Anaerobic energy recovery plants need significant initial investments, and their effluent streams need specific finishing treatment, that can be carried out by biological processes. Integration of both processes could imply advantages, related to the improvement of the overall energy balance of such a plant, and to the reduction of odor emissions[6], that is a critical aspect to be considered under the environmental point of view and regarding the stakeholder conflicts[7].

Possible process layouts, integrating anaerobic methane-generation processes with subsequent aerobic composting of residuals, are very interesting for the recovery of organics and nutrients in agriculture. Reuse possibilities consist of spreading the treated organic waste directly on agricultural soil, or its use as a constituent of high quality compost. An issue of great concern involves the mixing of substrates with waste containing undesired substances, such as inorganic and microbiological pollutants.

MATERIALS AND METHODS

Co-digestion process planning

In the larger landscape of biological waste treatment systems, anaerobic processes present a wide application area. Organic waste materials, produced in urban, agricultural and industrial activities, may come from the following cycles:

- wastewater treatment: high strength organic wastewater, mixed sludges, primary and biological sludges;
- MSW: organics from separate collection;
- agro-food industry and animal husbandry activities: liquid and solid waste, sludges and high-strength wastewaters.

Figure 1 shows a possible intervention scenario for the integrated treatment of organic biodegradable waste.

The quality and quantity of sludge produced during municipal and industrial wastewater treatment depend on wastewater characteristics and on the processes adopted. Sludge treatment is generally carried out *in situ* (at the plant), and is usually represented by thickening, stabilization, dewatering and drying. Depending on the amounts, aerobic or anaerobic biological treatments are generally used for organic material stabilization; the former are usually adopted in medium-low potentiality facilities (less than 30.000÷40.000 PE). In the anaerobic plants, energy recovery through biogas generation is usually foreseen, although with efficiencies that are much lower than that possible in integrated systems.

A basic hypothesis in a sludge management cycle, is whether to separate primary sludges management from that of biological ones: primary sludge is suitable to be easily thickened by gravity, and can subsequently show greater energy returns than the latter when treated. It also may contain undesired constituents making it alone, or the resulting mixed sludge, not suitable, in terms of quality, for use as an organic fertilizer or for compost production. Biological sludge, on the other hand, can generally find proper use in agriculture, due to its high nutrient content, although studies have shown that these nutrients (especially P), are not readily available to crops[8]. Nutrient recovery in mineralized form (e.g. *struvite*), has recently become a quite active research topic, in view of the ever increasing commercial prices of mineral P[9].

Wastewater and wastes produced by cattle and pig farms, dairy farms and other agro-food industries (processing of olives, tomatoes, wine, canned food and preserves) are ideal candidates for this type of treatment. This usually takes place in local dedicated facilities[10-12]. Animal husbandry activities' (cattle and pig farming, poultry farms and aviculture) residues essentially consist of animal dejections, both liquid and solid, often disposed by spreading over agricultural soils in loosely controlled manner. They may however constitute a good substrate to integrate in a co-digestion processes[13,14].

The recently introduced practice of separate collection/selection of MSW produces flows of organic material with different features. Separate collection of the organic fraction mainly operates on waste coming from large users and, only partly, from domestic users; its quality features can be quite variable. Mechanical selection of "raw" matter allows to obtain a high organic contents and good biodegradability characteristics, that can be suited almost ideally to a biological digestion process[15].

Quality features of the organic fraction are, nevertheless, strongly influenced by the composition of the original material and by the degree of the treatment: volatile solids content of "under-sieve" material does not usually rise above 50% of total solids. In addition, a high content of inert matter contributes to lower the quality of the mixtures so that it does not appear suitable for such co-digestion processes.

Innovation in organic waste treatment and disposal can be achieved through several technological interventions differentiated on the type of plant, and through managerial options at different levels of complexity. Initial criteria guiding in the choice of a specific facility layout planning, must take into account several objectives, such as process efficiency and environmental impact. Often, the main constraints, related to the features of the intervention area, are constituted by the availability of organic materials and by the nature and composition of organic matrixes. The quality and quantity of organic matter available for treatment depends

TABLE 1: Characteristics of biodegradable waste and wastewater.

Substrate	COD		Solids		Macro-nutrients		pH
	total	soluble	total	volatile	Nitrogen	Phosphorus	
	gO_2/l	%	g/kg	%	gN/kg	gP/kg	unit
Dairy effluent	15÷70	40÷70	0.2÷0.5	80÷90	7÷10	0.6÷1.0	3÷6
Olive oil mill wastewater	2÷82	40÷60	0.5÷25	70÷90	0.3÷0.5	0.2÷0.3	4.0÷5.5
Winery effluent	10÷92	40÷70	6÷80	70÷90	0.02÷0.25	0.002÷0.47	3.0÷5.3
Slaughterhouse wastewater	1÷5	30÷50	1÷3	70÷80	0.1÷0.2	0.001÷0.002	6.0÷7.5
Municipal primary sludge	10÷25	40÷60	20÷70	60÷80	0.3÷2.8	0.2÷2.0	5÷8
Cow Manure	3÷5	20÷30	90÷150	65÷75	0.15÷0.18	0.2÷0.27	6÷7
Pig Manure	10÷30	30÷50	10÷20	40÷70	2÷4	0.1÷0.2	6÷7
Fish waste	60÷420	-	300÷400	50÷80	15÷35	3÷8	-
MSW organic fraction	15÷210	-	100÷250	80 ÷ 90	1÷12	0.7÷2.5	-

on the intensity of activities within the territory, climatic factors, number and potential of waste production centers, and disposal modalities. TABLE 1 summarizes the main characteristics of a typical biodegradable waste.

Planning and design choices must guarantee processes efficiency and environmental sustainability and impact minimization. The two most common possible solutions, in relation to the treatment options, are the following: in situ treatment at facilities serving a single farm, or centralized treatment, whether at existing municipal WWTPs, or at new dedicated facilities servicing a consortium of users, public and private. Integrated systems have to be carefully considered, both for the construction of new facilities, and for the upgrade of already existing ones. Systems' feasibility must be verified through a technical-economic analysis of the intervention scenarious on the specific territory, evaluating existing waste stream production and plants location, selecting the most appropriate technologies, analyzing product destination, and assessing law constraints and a sustainable costs level.

Process influences and operational issues

The choice of a specific process derives from the analysis of the actual feed mixture (density, settling properties, solids tendency towards coalescence or adsorption), of the organic load physical characteristics, dissolved and particulate, and of the biomass affinity towards the substrate. Following are some indications on process and operational parameters that could be adopted to optimize a digestion process. Depending on several factors and considering the variability of technical solutions and operating conditions, it is interesting considering the value due to the extension of the different ranges.

Optimization of operative conditions can, in general, be achieved adopting a feeding rate as uniform as possible. Composition, physical-biological conditions of feed mixture and their variability significantly affect the stability of the process; preprocess/homogenization storage can induce significant variations of the substrate quality, caused by the development of natural fermentation processes.

In the digestion of poorly degradable waste, the hydrolytic phase is the limiting step of the whole process. In cattle manure anaerobic digestion, hydrolysis and acid production play the main roles: the development of two different enzymatic processes, one is controlled by native microorganisms already present in the manure, the other by external enzymes[16,17]is needed for optimal efficiency. The hydrolysis process is controlled by pH, temperature and by the total or dissociated concentration of volatile fatty acids (VFA)[18]. SRT can also be a limiting factor for a complete substrate hydrolyzation to occur[19].

The feed may contain impurities, such as sand, glass, metals, plastics, wooden residuals, terrain, straw. These must be eliminated as far as possible prior to digestion since, once in the reactor, they can determine malfunctions and loss of process volume[20,21]. Highly diluted mixtures can be opportunely treated by fine sieving, sand collection and thickening.

A suitable mixing and a heating to optimal process temperature should be foreseen to promote good biodegradation of the mixtures. High sludge water contents imply increasing costs for sludge heating, not compensated by higher energy recovery. Mixtures with a too high solids density affect process internal mixing dynamics, and interfere with mobilization of sludges. It is advisable to plan solids feeding at the front head of the heat exchangers, or at recycle pump location, and an automatic removal of foams and floating substances from within the reactor.

Advanced degradation of the solid fraction, allows the reduction of the amount of waste sludge to be sent to a post-treatment and, eventually, final disposal. To improve anaerobic digestion yield also in this sense, it is possible to enhance the hydrolyzation phase through enzymatic, thermal and chemical processes[13].

The presence of toxic/inhibiting compounds in the substrate has to be carefully evaluated, considering the acclimatization capability of microorganisms as well; antibiotics, disinfectants and pesticides can also be present and may inhibit partly or in full biological degradation. Methanogenic inhibitors, such as fatty acids, hydrogen sulfide and ammonia, that are also generated within the process itself, are toxic if present in unionized forms, which depends on pH values. Fatty acids with short chain represent a relevant product of the digestion process; during digestion of rapidly hydrolyzable wastes, an abnormal increase in fatty acids can point out an organic overload situation. Glucose fermentation is inhibited when total VFA concentration in the reactor is higher than 4 gL^{-1}[22]. Both propionic and butyric (fatty) acids are methanogenic bacteria inhibitors. It has been shown that a concentration higher than 3 gL^{-1} of propionic acid, leads to process failure[23,24]. Acetic acid is usually present at concentrations higher than that of other fatty acids[25]. Ammonia is toxic at pH values greater than 7, volatile fatty acids and hydrogen sulfide at values lower than 7[26].

All these factors may reflect negatively on biogas production rates; pH and biogas composition are important indicators of the stability of the ongoing process, and the availability of residual alkalinity plays an important role for the maintenance of methanogenesis. In an unstable process phase, the following operative parameters can be manipulated for control purposes: hydraulic retention time, solids residence time, applied organic load, sludge temperature. Buffer capacity and pH should be monitored, as well as fatty acids concentration.

PH stabilization is essential to achieve high conversion rates of volatile acids into biogas, minimizing their accumulation in the digester. This in fact will lead to pH decrease, and to a reduction of the bicarbonate buffer, thus inhibiting methanogenic activity. The most effective and fast way to adjust pH to close to neutrality values, is the addition of alkaline solutions (sodium and calcium hydroxides, sodium carbonate and bicarbonate) to the feed. The use of strong bases, such as lime or soda, on the other hand, implies drawbacks related to the variation of carbon dioxide concentration in the gas. If the rate of CO_2 absorption, both in gaseous form and in solution, is temporarily higher than the production of biogas, the digester can go into depression, with danger of air infiltrating into the reactor, and the subsequent formation of explosive mixtures.

Buffer capacity can also be increased quickly by addition of strong bases or carbonate salts, obtaining the removal of carbon dioxide in the gas phase and its conversion to bicarbonate. The balance of the gas phase can, however, proceed slowly and lead to overdosing. In a more suitable operation, bicarbonate can then be added directly[27].

Reduction of the ratio between organic and nutrient loads has been shown to be useful in pH control[28]. In co-digestion of MSW processes, a critical value of the COD/N rate was determined at about 50[29]. In digestion of animal husbandry waste, biomass acclimation allowed minimization of the inhibitory action due to high ammonia concentrations[30].

Process control

Process modelling techniques have now evolved into a mature simulation tool and are now one of the most efficient methods for defining project strategies and actuating process control. Among the mainstream models, the Anaerobic Digestion Model No.1 (ADM1) is perhaps the most used, as it describes the three well-known biochemical intra-cellular stages of acidogenesis (fermentation), acetogenesis (oxidation of organic acids) and methanogenesis and the two extra-cellular phases of disintegration and hydrolysis through parallel reactions through a first-order kinetics[31]. TABLE 2 summarizes the kinetic parameters applicable to disintegration and hydrolysis. The biological processes are represented by means of Monod-type reactions. Figure 2 shows the range of applicable parameters as reported in the technical literature[32,33].

TABLE 2 : Modelling parameters of disintegration and hydrolysis of biodegradable waste solids: first order constant rate for mesophilic process (d^{-1})

Substrate	Disintegration	Hydrolisis	Reference
Cheese whey	-	0.13 ÷ 0.24	[34]
OOMW	-	0.19 ÷ 0.35	[34]
Municipal primary sludge	0.25	0.10 ÷ 0.40	[35]
Cattle manure	0.13	-	[34]
Pig manure	0.01	0.28 ÷ 0.68	[34,36]
Solid waste	0.41	0.03 ÷ 0.40	[18,37]

A case study was carried out in order to assess the possibility of co-digesting specific waste streams generated in an Italian city, the operational results were then used to build a model of the process to simulate operating conditions and process design alternatives.

CASE STUDY OF MIXED WASTE CO-DIGESTION

Experimental setup

In order to assess co-digestion possibilities of a municipal sludge and a biodegradable waste, an experi-

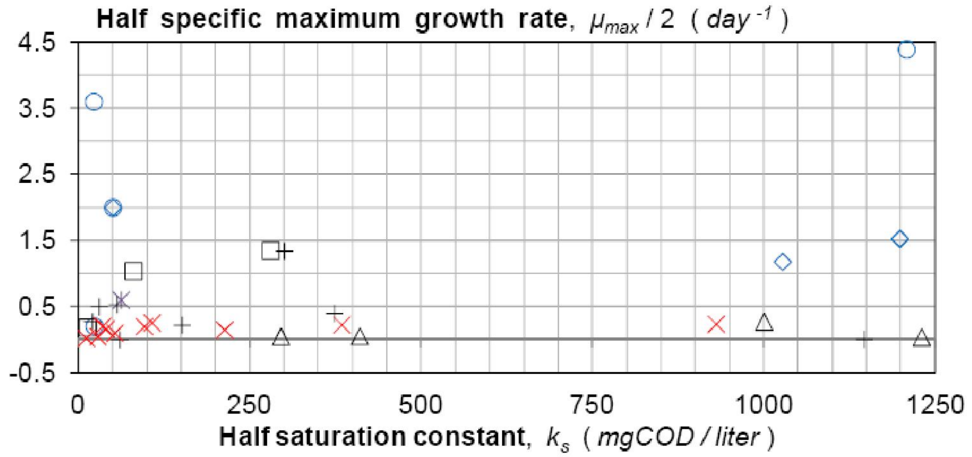


Figure 2 : Monod kinetics parameters for mesophilic anaerobic digestion of organic substrates. *Legend*: acidogenesis of monosaccharides (O) and amino acids (◇); acetogenesis of long chain fatty acids (Δ), valerate (∗), butyrate (□) and propionate (+); acetoclastic methanogenesis (X)

mental test was carried out in order to determine the anaerobic biodegradability of the mixture (Figure 3).

Tests were carried out at a mesophilic temperature of 35°C. This is the most common temperature for this type of processes, as it generally optimizes heating requirements of the digested mass and energy recovery yields. The inoculum was constituted by stabilized municipal sludge. Monitored parameters were COD, total (TSS) and volatile (VSS) suspended solids, total and ammonia nitrogen and pH. Biogas production was measured by volumetric displacement; a beaker, filled with 2M, NaOH solution, connected to a tank allowed to measure biogas volumes, already depurated from carbon dioxide.

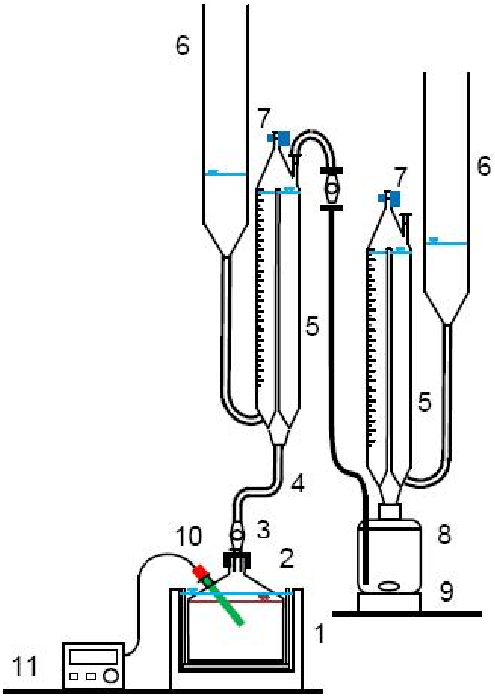


Figure 3 : Lab-scale batch anaerobic reactor. *Legenda*: (1) 1 liter glass bottle; (2) stirred thermostatic water bath; (3) no-return valves; (4) plastic tube; (5) eudiometric tube; (6) expansion tank; (7) gas discharge; (8) CO_2 trap beaker, containing magnetic stirring bar; (9) magnetic stirrer; (10) pH probe; (11) recording unit

In the batch test, organic substrate concentration varied in the range 5÷7 g COD L^{-1}. Biomass and substrate were added in function of organic load, which varied in the range 1÷2 g COD/g VSS. Retention time varied in the range 500÷600 hours (20.8-25 days).

Anaerobic process modeling

A modeling study was carried out to simulate the long-term behavior of the co-digestion process and verify the adequacy of process design parameters. Simulations were carried out by implementing the ADM1 model[33] on the tentative design and observed mixed wastes characteristics.

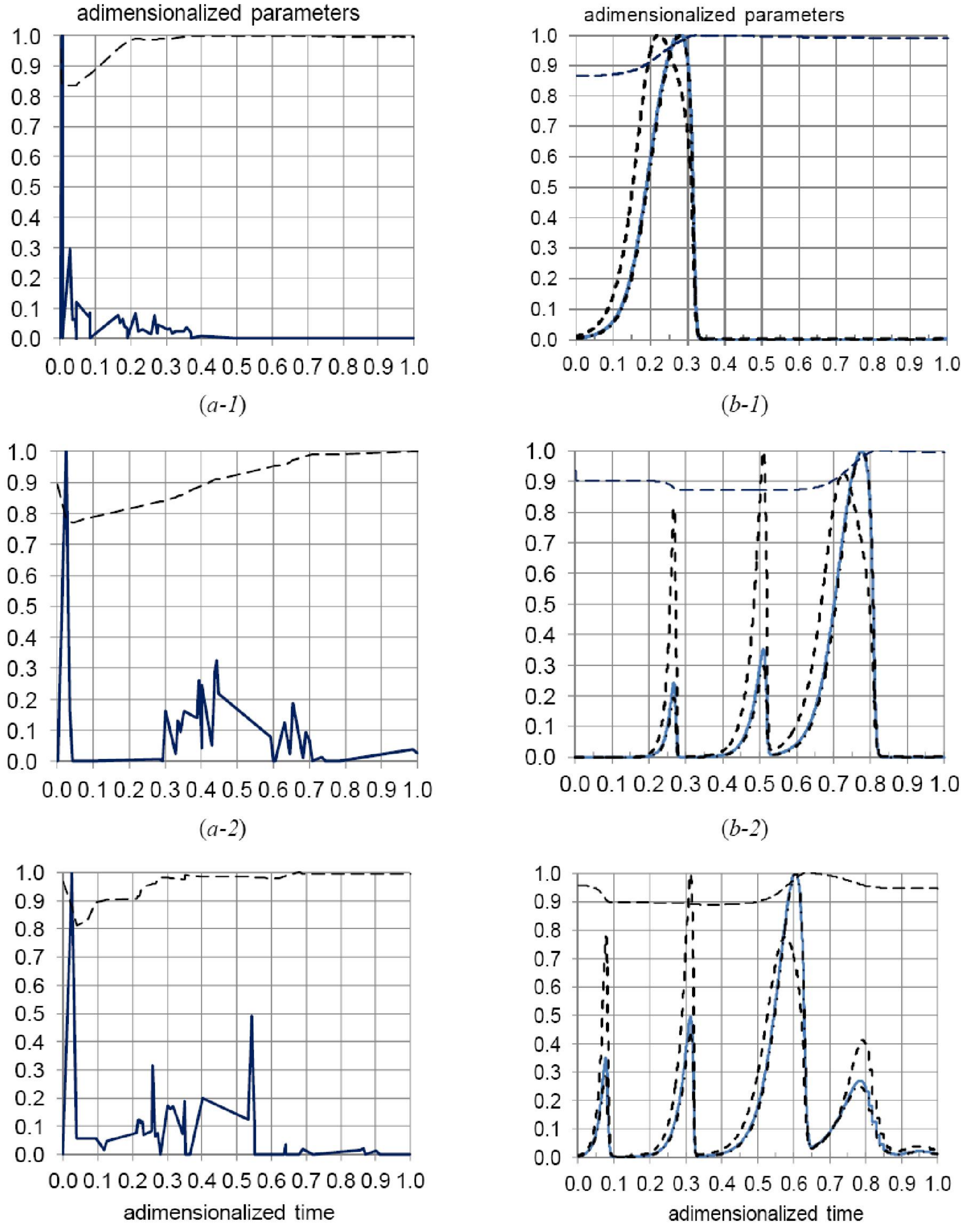
Three sets of simulation were carried out, each of them characterized by a different composition of or-

TABLE 3 : Composition of simulated organic substrate

Substrate	Unit	Test (ID)		
		A	B	C
Acetate	g L^{-1}	5.0	2.5	0.5
Propionate	g L^{-1}	-	1.25	0.5
Butirrate	g L^{-1}	-	1.25	0.5
Valerate	g L^{-1}	-	-	0.5
LCFA	g L^{-1}	-	-	0.5

ganic substrate fed to the digester: these were constituted by acetate only, by a mixture of VFAs (acetate, propionate and butirrate), and last, by a mixture of VFA, valerate and LCFA. TABLE 3 reports the composition of simulated organic substrate used in the test.

Organic substrate concentration varied in the range 3÷7 g COD L^{-1}. Biomass varied as a function of organic load in the range 1÷2 g COD $gSSV^{-1}$. A retention



(*a-1*) (*b-1*)

(*a-2*) (*b-2*)

(*a-3*) (*b-3*)

Figure 4 : Biogas production and pH in anaerobic assays: observed data in biodegradability tests (a-1, a-2, a-3) and simulated data in tests A (b-1), B (b-2) and C (b-3). *Legenda*: — (blue line) biogas; -·-·- methane; ---- carbon dioxide; — — — pH

time of 100 days was assumed. The inorganic fraction was characterized by alkalinity, cations and anions, in the ranges 1500÷5000, 10÷8000 and 10÷100 mg $CaCO_3$ L^{-1}, respectively. Nitrogen compounds were assumed to be present in negligible quantity.

Results were elaborated in adimensional form, as the ratio between the actual parameter value and its maximum value observed over the time, thus varying in a range 0÷1.

RESULTS

Results observed during the three assays in bench tests, were elaborated as a function of biogas production and pH range. In particular, the results of each assay "A", "B", "C" were reported in charts a-1, a-2 e a-3 and the results of relative simulations were reported in charts b-1, b-2 e b-3 (Figure 4).

In the assay A, an initial peak of biogas production is associated with the release of carbon dioxide, following which the batch process develops biogas at variable rates. The results of this test show that the biogas is mainly developed in the early stages of the process, and is accompanied by a rapid increase of pH, following closely a rapid phase of acidification.

In the assay B, the production of biogas is developed in an intermediate phase, and the pH, after a rapid phase of acidification, increases slowly to a stable value.

In the assay C, the production develops during most of the test duration, accompanied by a first phase of acidification followed by a marked process of alkalinization.

Simulation of assay A shows the production of biogas concentrated in one single peak, corresponding to the degradation of acetate, constituting in this case the only one organic substrate. The pH increases markedly as long as the methanogenic phase continues.

Simulation of assay B, biogas production develops through three subsequent peaks corresponding to the degradation of the three organic substrates contained in the influent, and consisting of VFA's (acetate, propionate and butyrate). The trend of pH shows now a moderate acidification phase, promptly followed by alkalinization which supports methane production.

Simulation of assay C, in which available organic substrate is a mixture of VFA, valerate and LCFA, more pronounced peaks of biogas production are evident. After a first stage of acidification, the pH remains constant while the production of methane starts; a following increase of pH value supports a major phase of methane production. A moderate pH decrease, and a final, less intense

Among the main results, batch assays allow to get time and maximum rates of biogas and methane and some information on the process of acidification by measuring the pH and the availability of alkalinity. Results observed and calculated during the three assays in bench tests, were elaborated as a function of biogas production and pH range (TABLES 4 and 5).

Some simulations show that the peaks of biogas flow correspond with the relative maximum of methane production. This aspect is certainly representative in the degradation of soluble substrates, quickly convertible into methane. This condition can detect a limit in the modeling of the process in batch process of substrates slowly acidifying, also present in particulate form, when the carbon dioxide production is predominant, in particular in the first phase of the process.

Considered the approximation of data used for base parameters of kinetic processes, the experimental assays present highest values of biogas rate production compared with data simulated, but the compared overall production rate is rather satisfactory, considering that the process of methane production develops during the simulation time in a more long time. The modelling study represents the different process as a sequential series and this is not widely confirmed in the observed assays. Anyway the global performance of anaerobic process

TABLE 4 : Results observed in anaerobic biodegradability assays

Test	Biogas maximum flow rate (mL h^{-1})	pH range
A	42.9	7.4 ÷ 8.5
B	73.8	7.4 ÷ 8.4
C	232.6	7.2 ÷ 8.1

TABLE 5 : Results of simulated anaerobic biodegradability assays

Test	Maximum Flow rate (mL h^{-1})			pH range
	Biogas	Carbon dioxide	Methane	
A	13.8	0.6	13.4	7.4 ÷ 8.5
B	13.9	1.0	13.2	7.4 ÷ 8.4
C	12.2	1.1	11.5	7.2 ÷ 8.1

is substantially reproduced.

CONCLUSIONS

Anaerobic treatment of organic substrates represents one of the possible technologies for indirect energy recovery from selected waste streams, and must be carefully evaluated for both technical and economic feasibility, in the presence of existing production centers of such wastes. Satisfactory application of treatment and disposal of organic waste with an energy recovery goals, needs an optimization of management procedures, assurance of adequate waste flows and specifications, and proper facilities layout and operation, in order to obtain a good integration of all the different phases that can take into account the innovations arising from current research and best technical practices.

The evolution of national/international regulations, and the constant costs increases for waste treatment and disposal, will soon determine new scenarious, where positive utilization of anaerobic, or other emerging processes, for energy recovery would be considered an obligatory solution, as demonstrated by the increasing number of already existing or planned full-scale applications.

Although the process itself is relatively stable, some problems that can influence or limit its design potential and yield cannot be neglected; among these, a balanced composition of organic mixtures, possible toxic effects of substances therein contained, requirement of specialized biomasses at process inception. Among the main factors influencing operation of anaerobic processes, composition and supply modalities of waste mixture can be recognized as significantly affecting the stability of the process. The hydrolytic phase itself is strongly influenced by pH, temperature and concentration of total or dissociated volatile fatty acids, representing a limit for the optimal development of the process.

The representation of the anaerobic digestion process is very difficult, particularly in the presence of complex substrates. In real reactors, in which the mixing conditions are generally not perfectly homogeneous, the development of different processes is strongly influenced by the local availability of substrate and the presence of suitable biomass. Regards to the batch assays which were carried out, a qualitative representation of the process was obtained (very important aspect, difficult to represent, and the content of methane in the biogas).

In order to obtain an efficient representation of the anaerobic process, further investigations are necessarily required for the assessment of both the methanogenic potential of complex substrate finalized to monitor the process and in particular to control the conditions of pH and available alkalinity.

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