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Numerical framework for anaerobic digestion and/or composting of bioplastics and organic waste performance evaluation under real-like large scale operating conditions

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A R T I C L E I N F O

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A B S T R A C T

Bioplastics are newly designed and developed plastic materials to meet the criteria of sustainability and sustainable development that present the following characteristics: biobased, biodegradable or both properties. They include a whole family of materials with different properties and applications. Bioplastics certified as biodegradable and/or compostable, according to the regulatory plan of Italy and other countries around the world, must be collected with organic waste and follow the same end-of-life path, i.e., organic recycling processes in anaerobic digestion and/or industrial composting plants. However, it should be noted that they are certified according to tests carried out under standardized optimal degradation conditions, that is, conditions that are not always the same as those found in the operation of anaerobic digestion and composting plants. Improper management of these materials can become an increasing problem as their continued widespread use will require large volumes of these materials to be treated in facilities that are not designed to biodegrade bioplastics, resulting in potential contamination of large volumes of undegraded bioplastics in digestate and/or compost produced in the treatment of conventional organic waste. The purpose of this work is to investigate the degradation of the most popular bioplastics currently collected with organic waste through separate collection under the actual operating conditions of waste recycling plants operating through biodegradation processes. Specifically, using both studies conducted at the laboratory scale and evidence of biodegradation plant configuration and operation at the real scale, the Aspen Pus V12 program was used to model and simulate an anaerobic digestion process integrated with a composting process. This evaluated the level of biodegradation, expressed as a percentage of degraded mass, as a function of the implemented process conditions such as temperature and hydraulic residence time (HRT). The bioplastics that have been focused on, certified as compostable, are PLA (rigid disposable containers: cups) and starch-based shopping bags (SBS). The results achieved show that through an anaerobic digestion process alone, after 28 days in thermophilic conditions, PLA can degrade by 42%, and SBS by 44%.

The integrated anaerobic/aerobic process provides a biodegradation performance allows 85% and 56% for SBS and PLA, respectively, in case the biopolymers are subjected to 28 days of thermophilic anaerobic digestion and 90 days of composting.

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1. Introduction

Bioplastics are biobased and/or biodegradable plastic materials newly designed and developed to meet the criteria of sustainability. They offer several advantages compared to some conventional plastics used for the same applications at lower impact (Lombardi et al., 2023) in terms of environmental pollution.

Biodegradable plastics are environment friendly, but they also came with some limitations such us high manufacturing cost and low mechanical tendency (Thakur et al., 2018). Despite this, the European Bioplastics forecast shows that up to 2.22 million tons of bioplastics were produced in 2022, and that the projection to 2027 will result in the production of 6.30 million tons of bioplastics (European Bioplastics, 2022b).

The European Bioplastics Agency classifies bioplastics into three categories: biodegradable and produced from renewable resources, biodegradable and produced from nonrenewable resources, and non-biodegradable but produced from renewable resources (European Bioplastics, 2022a).

Biodegradable bioplastics are generally designed to degrade via biological processes such as landfilling, composting, or in natural environments such as aquatic media and soils (Atiwesh et al., 2021) (Folino et al., 2020). Biodegradable plastics are plastics that can be degraded by microorganisms into H_2O and CO_2 under aerobic conditions, or into methane and carbon dioxide (biogas) under anaerobic conditions (Dilkes-Hoffman et al., 2019)(Abraham et al., 2021).

In particular, the most popular biodegradable bioplastics include PLA (polylactic acid) and starch-based bioplastics (European Bioplastics, 2022b). The widespread use of these biopolymers highlights the need to analyze and evaluate their end-of-life as waste. Starch is one of the first biopolymers used for the development of sustainable materials to replace petroleum-based synthetic plastic polymers. As a results of their low cost, renewability, and inherent biodegradability, starch-based polymers are high-potential feedstocks for the large-scale production of bioplastic films.

Polylactic acid or polylactide (PLA) is a biobased, biodegradable, widely used bioplastic. It is a linear thermoplastic aliphatic polyester synthesized from lactic acid molecules (Fredi and Dorigato, 2021)(Di Bartolo et al., 2021). Lactic acid is a chiral molecule that can exist in three different stereochemical forms: L-lactide (PLLA), D-lactide (PDLA) and D-L-lactide (or meso lactide) (PDLLA). The ratio utilized for these isomers determines the overall properties of future synthetized PLA (Di Bartolo et al., 2021).

Starch granules consist almost entirely of two main polysaccharides, namely amylopectin, accounting for 70–85% of total starch and amylose, present for the remaining 15–30%. Starch granules consist of a semi-crystalline structure with a central amorphous region, mainly composed of amylose, and a circumferential repetition of alternating crystalline and amorphous lamellae. Due to its high brittleness and poor mechanical properties, native starch cannot be directly processed as thermoplastic material. Plasticizers are generally used to increase the capability of processing starch-based biopolymers (Bartolucci et al., 2023)(Bertoft, 2017).

As defined in the standards UNI EN 13432:2002 for packaging items (UNI EN 13432:2002) and UNI EN 14495:2007 for other materials (UNI EN 14995:2007), biodegradability is the breakdown of an organic chemical compound by microorganisms in the presence of oxygen to carbon dioxide, water and mineral salts, or any other elements present and new biomass (aerobic degradation), or in the absence of oxygen to carbon dioxide, methane, mineral salts, and new biomass (anaerobic degradation). A plastic material is considered aerobically biodegradable if at least 90% of its C content is converted to CO_2 during a composting test carried out for a maximum period of 6 months at 58 ± 2 °C (UNI EN 13432:2002), following the method reported in (ISO 14855–1:2012), and anaerobically biodegradable if the percentage of biodegradation based on biogas production is greater than 50% (UNI EN 13432:2002). In the latter case, the test is carried out at 35 °C for a maximum of 90 days (ISO 14853: 2016).

The biodegradability of polymers depends on their chemical structures, polymer chains, complexities, crystallinities, and glass transition temperatures. Polymers with short polymer chains, simple formula and an amorphous part are degraded more easily. Factors, such as temperature, hydraulic residence time (HRT), pH, moisture and oxygen content, also affect the degradation of the polymers (Massardier-Nageotte et al., 2006)(Emadian et al., 2017)(Kale et al., 2007) as well as the microbial population involved in the degradation media (Bátori et al., 2018) (Quecholac-Piña et al., 2020).

Usually, the real scale processing conditions used for the treatment of organic waste are different from those prescribed by standards, in terms of temperature, bioplastic concentration, and time (Assessing bioplastics biodegradability by standard and research methods: Current trends and open issues). The incomplete degradation of bioplastics, achieved at the end of biological treatment, results in contamination of digestate and/or compost, which must be mechanically separated in the final step (or before the biological processes) (Cucina et al., 2021a). The contamination of digestate and/or compost by bioplastic fragments doesn't allow to be used it in agriculture, resulting in significant environmental burden (Gadaleta et al., 2023). In fact, these bioplastic fragments and pesticide residues used in agriculture can potentially cause unpredictable damage to the environment due to their persistent bioaccumulation (Tran et al., 2023)(Tourinho et al., 2019)(Gadaleta et al., 2023).

However, the real applicability of biological processes for the treatment of used bioplastics is mainly based on their biodegradability that depends on the complexity of the bioplastics structure and on the type of raw materials used, so that potentially different waste streams should be adopted according to the characteristics of the bioplastics. Now, the scientific literature reports few examples of full-scale plants destined specifically to bioplastics treatment (Folino et al., 2023). Laboratory methods possess the advantage of being able to set and keep control of the experimental conditions (temperature, humidity, pH, oxygen supply, and test duration), but cannot exactly reproduce the conditions present in the multitude of natural and industrial environments (Folino et al., 2023).

Papa et al. showed the percentage of degradation and constant degradation kinetics (mgC/gC*d) of tableware made of PLA and starch blend polymers. They are subjected to different anaerobic digestion conditions (thermophilic and mesophilic), and the most relevant results show a percentage of degradation of 20 ± 0.9 for PLA and 21 ± 3.4 for starch blend for an anaerobic digestion performed at 37 °C for 30 days with which is associated a kinetic constant of 1.87 mgC/gC*d for PLA and 2.67 ± 0.21 for starch blend bioplastic; a percentage of degradation of 28 ± 1.4 for the PLA and 20 ± 1.8 for the starch blend and a kinetic constant of 2.87 ± 0.15 mgC/gC*d for PLA and 1.91 ± 0.18 mgC/gC*d for starch blend; after 30-day anaerobic digestion at 55 °C; a percentage degradation of 39 ± 1.2 for PLA and 20 ± 0.9 and a kinetic constant of 4.19 mgC/gC d for PLA and 1.86 mgC/gC*d for starch blend bioplastic after a 60-day anaerobic digestion at 55 °C (Papa et al., 2023).

Calabrò et al. tested some biodegradable polymers under mesophilic or thermophilic conditions. After 15 days of digestion, a dry mass reduction of 22.8 ± 6.2% and 27.6 ± 14.0% for mesophilic and thermophilic tests, respectively, for starch blend bioplastics (Calabro' et al., 2020).

Cafiero et al. investigated the performance of small-scale composting. Starch blend bioplastics were composted in 600L static home composters, and a 1m^3 electromechanical composter. Six months of residence time in static home composters resulted in 90–96wt% degradation depending on the management approach adopted, and two months in the electromechanical composter achieved 90 by weight (Cafiero et al., 2021a).

Cucina et al. discovers that, studying an AD with thermophilic temperature for 30 days of hydraulic retention time followed by 30 days of mesophilic maturation, a laboratory-scale data did not differ from the data coming from the full-scale experiment, i.e., bioplastic degradation was not affected by the reactor volume. Bioplastics showed an average degradation of 27 ± 5% on a weight basis (Cucina et al., 2022).

It is clear that at a bibliographic level there is discussion and comparison on the actual biodegradation of biopolymers during biological processes. It arises from the difference in the modes of operation of the tests performed, but also in the polymers used. Indeed, although the PLA polymer is unique, it can be produced with different degrees of crystallinity, different polymer chain lengths, different geometries, or different additives. These operational diversities may lead to nonunique determination of the biodegradation behavior of biopolymers. In addition, the operational difficulty in measuring biodegradation and disintegration on a large scale gives rise to the need for a tool that assesses the true biodegradability of rigid and flexible bioplastics and examines compatibility with current management of the organic waste fraction, in order to provide useful information to plant operators.

In this study, we explored the biodegradation of two distinct bioplastics, namely PLA (found in disposable cups) and starch-based bioplastic (commonly used in shopping bags), alongside the typical waste present in the organic fraction. To address the challenges associated with assessing the actual biodegradation of bioplastics in real-world scenarios, a simulation model for an integrated anaerobic/aerobic system using Aspen Plus V12 software has been developed.

Key characteristics of the biopolymers and the organic fraction were sourced from relevant literature and incorporated into the simulation. Additionally, parameters related to biochemical reactions and reaction kinetics crucial to the process were obtained from literature sources and applied in the simulation model. By utilizing waste characteristics, biochemical reactions, and kinetics documented in existing literature, we successfully modeled aerobic, anaerobic, and integrated biodegradation processes under conditions mirroring those of full-scale plants.

The simulations conducted allowed for an exploration of actual biodegradation and biogas production as a function of residence time and temperature, providing insights into the performance under operational conditions resembling those of large-scale plants. This approach addresses practical challenges faced in plant operations related to assessing the actual biodegradation of bioplastics.

The primary objective is to offer a valuable tool for evaluating the biodegradation of widely-used biopolymers in conjunction with organic waste. This is achieved through the simulation of real-scale conditions using the Aspen Plus V12 software, thus overcoming challenges posed by ambiguities in the literature (mainly focus on lab-scale analysis). The novelty of this work lies in its ability to provide useful insights into a critical aspect—biodegradation of biopolymers—especially when subjected to treatment in large-scale plants. Ultimately, this tool serves as a valuable resource for assessing the practical feasibility and performance of strategies aimed at recovering bioplastics from organic waste under conditions closely resembling real-world operations.

2. Materials and methods

In the Aspen Plus V12 simulation environment, data characterizing the waste entering the combined anaerobic-composting process were used as input data and it was necessary to process the characteristic data of the chemical reactions regulating the process and related reaction rates, again of the waste at input, to allow simulation using the prepared model.

2.1. Aspen Plus modeling

Fig. 1 shows a sketch of the numerical model developed in Aspen Plus V12 to represent the integrated anaerobic/aerobic plant. The

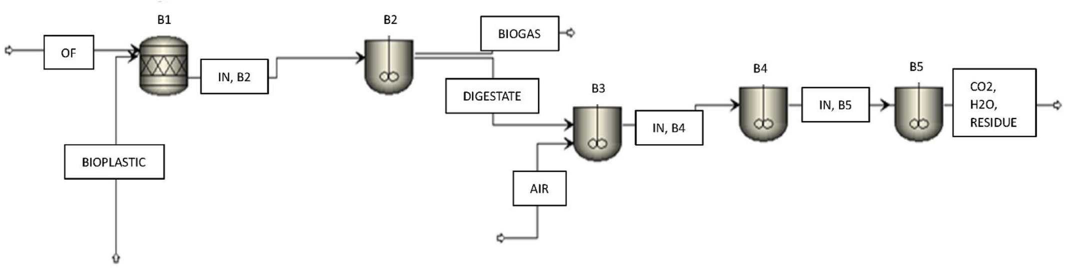

Fig. 1. Integrated plant layout.

model is composed by an anaerobic digestion section (B1 and B2), and a composting section (B3, B4 and B5). Organic fraction (OF) and bioplastics enter first anaerobic digestion reactor. In particular, the first phase of the anaerobic digestion was described in two steps, the first one related to the hydrolysis phase of the organic fraction using a model from literature (Anaya Menacho et al., 2022) (Hoa Huu Nguyen, 2014) (Serrano et al) and the second related to the reactions of the organic fraction and bioplastics after hydrolysis. Within the second AD reactor, the chemical reactions of the organic fraction and biopolymers (explained in section 2.3) with their respective reaction kinetics (explained in section 2.4) were included, returning biogas and digestate (where the bioplastics that did not complete the AD reaction are present).

For the simulation of AD, two temperature regimes were considered separately: mesophilic and thermophilic, and three different hydraulic retention times: 14, 21, and 28 days. At the end of the anaerobic phase, the resulting digestate (IN, B2) was taken as input of the aerobic phase, schematized using three continuous stirred tank reactors (CSTR) in series: one for the sanitization phase (B3, operating in of high thermophilic temperature, approximately 55 °C), one for the stabilization phase (B4, operating in a medium-low thermophilic temperature regime, approximately 37 °C) and one for the maturation phase (B5, operating in a mesophilic temperature regime, approximately 25 °C). On leaving the last composting reactor there is carbon dioxide, water and residue (undegraded bioplastics). For the three reactors considered, process times of 10, 20 and 60 days were assumed, respectively (ANPA. et al., 2002) (Misra R.V et al., 2003). Within the 3 composting reactors, the same two reactions were included for PLA and SBS, respectively (shown in Section 2.3). Differences within the reactors are the reaction kinetics (illustrated in section 2.4), which decrease in value as the reaction temperature decreases. At the end of the three composting reactors, there is production of CO_2, water, and residual bioplastics.

For AD, the degradation of both the OFMSW (or OF for simplicity), and the biopolymers considered was evaluated to estimate the contribution of the biogas that can be produced from each of these fractions if processed together. For composting, attention was focused on the degradation of the biopolymers in order to evaluate the completion of the degradation processes within the operating conditions usually adopted for OFMSW composting.

The main assumptions made can be summarized as follows: i) the bioplastic and the organic fraction enter the plant under environmental conditions; ii) the mass composition of the incoming waste is 97% as organic fraction and 3% as bioplastics; iii) bioplastics are made up of 50% PLA and 50% SBS, both 100% volatile; iv) the AD process considered is of the semi-dry type: dry content equal to 15% by mass.

2.2. Feedstock data collecting

For the composition of the OFMSW used as input to the process, the characterization results obtained by thermogravimetric analysis were taken as reference (Rajendran et al., 2014) (Anaya Menacho et al., 2022; Luz et al., 2021)and the resulting degradation kinetics.

For the composition of the biopolymers considered and constituting the bioplastics present in the incoming waste stream, given their high availability on the market, the chemical characteristics of the PLA cups and starch-based shopper (SBS) were taken as reference. compostable certificates according to EN 13432:2002 and labeled by CIC (Italian Consortium of Composters) and TuV Austria. obtained in a previous work (Lombardi et al., 2023). Since the biopolymers considered are unconventional compounds in Aspen Plus, their composition has been schematized in terms of proximate (Table 1) and elemental analysis (Table 2).

These assumptions were necessary both to evaluate the anaerobic biodegradation of the organic fraction of municipal solid waste and bioplastics, and to evaluate the contribution to the quantity of biogas that can be produced not only from OFMSW but also from bioplastics if collected jointly and sent to a complete cycle of combined anaerobic digestion and composting treatment.

By multiplying the percentage of each element by the dry weight of the corresponding bioplastics (volatile matter), the mass quantities are obtained:

$$P_{i,j}(g) = P_{SV,i} \bullet [xi, j]/100 \quad (1)$$

Where:

- i is the index identifying the bioplastics (PLA and SBS);
- j indicates the C, H, O, N, and S content of the i-th bioplastic;
- Pi,j is the weight of the j-th element related to the i-th bioplastic;
- Psv,i is the weight of the volatile substance of the i-th bioplastic;
- [xi,j] is the mass fraction of the i-th element;

Neglecting the presence of sulfur for PLA and nitrogen for SBS, the moles of the various elements can be obtained by dividing $P_{i,j}$ by the corresponding molecular weight ($PM_{i,j}$), to then obtain the corresponding approximate chemical formula (Table 3).

$$n_{i,j}(moles) = Pi, j/PM_{i,j} \quad (2)$$

Table 1
Bioplastics Thermogravimetric data for bioplastics.

	Moisture (%)	Volatile metter (%)	Ash (%)	Fixed carbon (%)
PLA	2.52	97.35	0.08	0.05
SBS	2	92.62	0.25	5.13

Table 2
Bioplastics elemental data.

	C (%)	H (%)	O (%)	N (%)	S (%)
PLA	52.74	4.04	43.17	0.03	0.02
SBS	59.53	5.15	34.81	0.03	0.48

Table 3
Bioplastics chemical formula.

Polylactic Acid (PLA)	$C_{1237}H_{1120}O_{752}N$
Starch blend shopper (SBS)	$C_{319}H_{318}O_{139}S$

2.3. *Chemical model*

The general anaerobic digestion and composting reactions of the two biopolymers have been schematized as shown in (Kolstad et al., 2012). Specifically, the following reactions are used:

PLA anaerobic digestion:	$C_{1237}H_{1120}O_{752}N + 582\ H_2O \rightarrow 667\ CO_2 + 570\ CH_4 + NH_3$
SBS anaerobic digestion:	$C_{319}H_{318}O_{139}S + 170\ H_2O \rightarrow 155\ CO_2 + 164\ CH_4 + H_2S$
PLA composting:	$C_{1237}H_{1120}O_{752}N + 1140\ O_2 \rightarrow 1237\ CO_2 + 559\ H_2O + NH_3$
SBS composting:	$C_{319}H_{318}O_{139}S + 330\ O_2 \rightarrow 319\ CO_2 + 159\ H_2O + SO_2$

Thanks to the Aspen Plus model it was possible to introduce, for each operating condition considered, the most appropriate reaction kinetics to then evaluate the actual biodegradation of both the OFMSW and the bioplastic, in each of the simulated steps.

2.4. *Kinetic model*

In this section, the calculation of the reaction kinetic constant of the main reaction steps of an anaerobic digestion or composting plant has been carried out. Determining the reaction rate as a function of time and temperature is a necessary requirement for the implementation of the kinetic biodegradation model for bioplastics. Pseudo-zero order kinetics was assumed for the biodegradation reactions of the two bioplastics, both during anaerobic digestion and composting. This assumption was made because these polymers exhibit linear biodegradation over time, for certain time intervals (Cucina et al., 2022). Such fragmented linearity indicates that the reaction rate, being of pseudo-zero order, is independent of the amount of bioplastic and proportional to the kinetic constant considered in the specific time interval (Cucina et al., 2022).

The determination of the kinetic constants of the two biopolymers was performed using different methods depending on the availability of the data in the literature. In each case, the kinetic constants of the individual processes, that is, anaerobic digestion and then composting, were calculated separately. The sources from which the reference data for the calculations were taken are all laboratory-scale studies.

In the case of AD and PLA composting, biodegradation curves from laboratory studies, under varying hydraulic residence time, in mesophilic and thermophilic conditions, can be found in the literature. Using these curves, it was possible to perform linear interpolations and extrapolate the angular coefficient of the half lines to obtain the kinetic constant of degradation. In the case of composting, no biodegradation curves could be found; only mass losses corresponding to hydraulic residence times are available (point values). So, for the reconstruction of biodegradation half lines, these point values were taken and merged. By manipulations described in the respective sections, it was possible to obtain the values of the biodegradation kinetic constants under the different conditions.

The kinetic reaction during the OFMSW anaerobic process is taken from literature (Rajendran et al., 2014b)(Anaya Menacho et al., 2022).

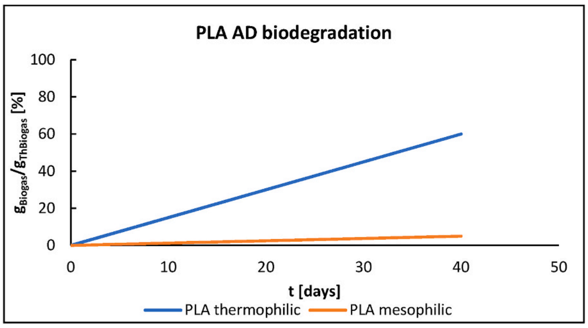

Fig. 2. Interpolated mass loss curve during PLA AD.

Table 4
kinetic constant value for PLA AD.

Temperature (°C)	k (d^{-1})
52	$15*10^{-3}$
37	$1.2*10^{-3}$

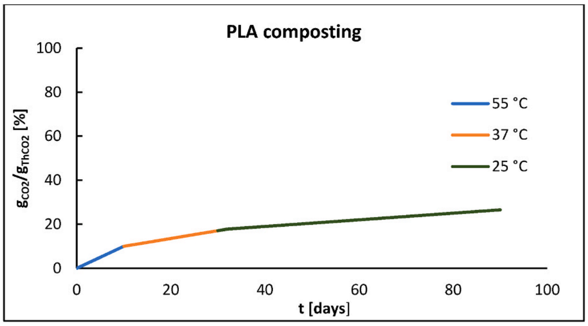

Fig. 3. Interpolated biodegradation curve during PLA composting.

2.4.1. *PLA reaction kinetics during anaerobic digestion*

Determination of kinetic constants useful for simulation of anaerobic digestion of PLA was done by interpolating polymer biodegradation curves during tests taken from literature (Itävaara et al., 2002). In the considered paper, the biogas generated during the anaerobic digestion of the organic fraction of mixed municipal solid waste together with PLA is measured. This method is designed to simulate typical anaerobic digestion conditions for the organic fraction of mixed municipal solid waste. The test method is designed to provide the percentage of carbon in the test material and its conversion rate to developed carbon dioxide and methane (biogas). The test material is exposed in a laboratory test to a methanogenic inoculum derived from anaerobic digesters operating only on pretreated municipal solid waste (ASTM D5511). The trend of the biogas curve produced is shown in Fig. 2, as a function of the temperature regime applied and of hydraulic residence time.

Taking into account thermophilic conditions, there is a biogas release of 60% in 40 days. With a linear biodegradation regime, the value of the kinetic constant, represented by the angle coefficient of the biodegradation line, can be determined. Observing the biogas release curve during the test carried out under mesophilic conditions (37 °C), a degradation of 5% is observed in 40 days. The reaction kinetic constants for the two different temperature regimes are summarized in Table 4.

2.4.2. *PLA composting*

Literature data useful for extrapolating kinetic constants refer to the time course of the evolved gas, in this case CO_2, using a method for determining the ultimate aerobic biodegradability of organic compound-based plastics under controlled composting conditions (Itävaara et al., 2002.) (ASTM D5338).

To find a linear trend for biodegradation at intervals, the curves available in the literature for different operating temperatures were approximated to straight line segments Therefore, the value of the kinetic constant (k) can be calculated by determining the angle coefficient of the straight lines. It was then possible to express the degradation kinetics used for time intervals (10, 20 and 60 days) at different temperatures (55, 37, and 25 °C respectively) in terms of actual process conditions (Fig. 3). It is shown that in each time interval considered (Fig. 3), colored lines the temperature remains constant and then varies from interval to interval.

Thus, it was possible to determine the three values of the kinetic constants as a function of the temperature regime and the number of associated days, as shown in Table 5.

2.4.3. *SBS anaerobic digestion*

Compared with the available literature on the biodegradation of polylactic acid, there are no degradation curves available for starch blends. Therefore, the values have been taken as a biodegradation reference (Calabro' et al., 2020). The studied samples are exactly the starch-based shoppers found in Italian supermarkets, with a composition of 60% starch and 40% PBAT, and the tests were carried out under both mesophilic and thermophilic conditions. The values are shown in Table 6.

Mass loss is observed to occur mainly during the first two weeks, under both mesophilic and thermophilic conditions (see Table 6 and Fig. 4). Since this type of SBS consists of two polymers, namely starch and polyester, it is inferred that starch was rapidly degraded

Table 5
Kinetic constant value for PLA composting.

Temperature (°C)	HRT (days)	k (d^{-1})
55	10	$1.0*10^{-2}$
37	20	$0.35*10^{-2}$
25	60	$0.15*10^{-2}$

Table 6
Mass loss value for SBS AD.

Condition	Mass loss (%)	
	15 days	30 days
Mesophilic	24.4	28.7
Thermophilic	36.5	44.7

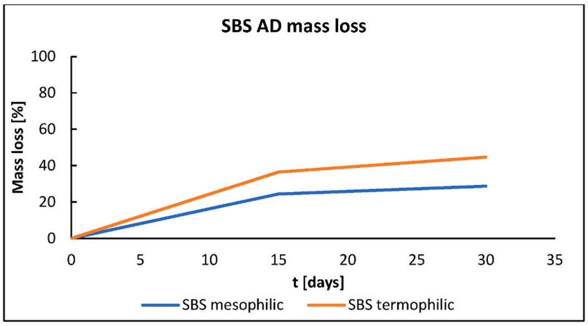

Fig. 4. Interpolated mass loss curve during SBS AD.

in the first 15 days. Thereafter, it is PBAT that degrades with slower kinetics (Calabro' et al., 2020) (Cucina et al., 2022) (Ruggero et al., 2021) (Papa et al., 2023).

Using this methodology for the assessment of biodegradation, linear kinetics was imposed, which allowed for easy determination of the constant values for both operating regimes (see Table 7).

AD is simulated for an HRT of 14, 21, and 28 days. For a proper analysis, the 21day simulation is carried out considering, in addition to the first reactor with an HRT of 14 d and characterized by its own kinetics (k_1), a second reactor in series, operating under the same T conditions but with an HRT of 7 days and characterized by reaction kinetics equal to those found in the second section. For Step 28, such a second reactor will have an HRT of 14 days and the imposed kinetic constant is k_2.

As anaerobic digestion in Aspen Plus V12 is represented by a single reactor as seen below, the implementation of an average kinetic constant (k_m) that considers the different trends of mass loss during this process as a function of time is necessary.

$$k_m = \frac{k_1 \bullet N_1 + k_2 \bullet N_2}{N_{tot}} \tag{3}$$

Where:

- N_1 is the number of days spent at constant slope k_1 which is 14 days;
- N_2 is the number of days spent at constant slope k_2 and is between 14 and 28 days.

2.4.4. SBS composting

The determination of the first kinetic constant k_1, which is needed to describe the reaction during the sanitization phase, takes into account the biodegradation values under thermophilic conditions (Bastioli, 1998). From the percent biodegradation of the generated experimental curve of SBS, in thermophilic conditions, it was possible to break it into two segments as can be seen in Fig. 5: a slope for a duration of 10 days (assumed as k_1), and a slope for a duration over the 10 days (not considered in the evaluated process) (Fig. 5).

For the calculation of kinetic constants (k_2, k_3) under mesophilic conditions (at 37 °C, next step – mineralization - of the evaluated process) and at temperature condition (25 °C, last step – maturation - of the evaluated process) reference is made to the average value of $k_m = 5.3*10^{-3}\ d^{-1}$ during aerobic degradation over a temperature range of 25–45 °C (Cafiero et al., 2021b), whereas no other bibliographic references that could be used for this purpose have been found.

Staring to this value and assuming that in mineralization and maturation phase SBS and PLA have a similar behavior, and k_2 (referred to 20 days for the mesophilic phase process) can be assumed (Cucina et al., 2021b) equal to 2 times k_3 (where k_3 is referred to 60 days of maturation phase at 25 °C) using eq. (3), the values of the three kinetic constants is determined.

The assumed three values of the kinetic constants as a function of the temperature regime and the number of associated days, are

Table 7
Kinetic constant value for SBS AD.

Condition	k_1 (d^{-1})	k_2 (d^{-1})
	15 days	30 days
Mesophilic	$1.63*10^{-2}$	$0.28*10^{-2}$
Thermophilic	$2.40*10^{-2}$	$0.58*10^{-2}$

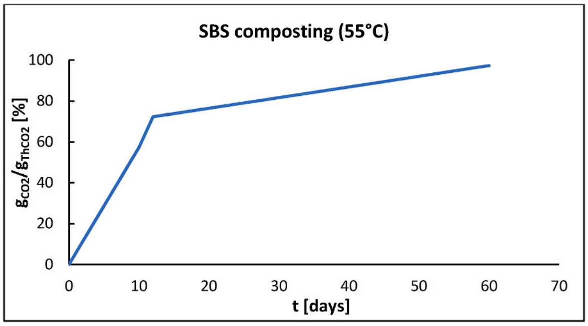

Fig. 5. Interpolated biodegradation curve during SBS composting in thermophilic condition.

shown in Table 8.

3. Results and discussion

The simulation results return the PLA and SBS flow rates and the biogas volume after the process. From the latter, note the input flow rate, the percentage of biodegradation of the respective bioplastics is determined by applying the equation:

$$\text{Biodegradation } (\%) = \frac{(M_i\text{-}M_f)}{M_i} \bullet 100 \quad (4)$$

The results are shown in three different sections: the first on the anaerobic process only, the second on composting only, and the third on integrated treatment. In this way, it is possible to identify possible criticalities of some subprocesses.

3.1. Anaerobic digestion

Fig. 6 shows the results of the percentage of biodegradation for anaerobic digestion only under mesophilic conditions, focusing only on biopolymers.

Observing the biodegradation, it becomes evident that:

- PLA exhibits minimal degradation, reaching a maximum of 4% even after 28 days of HRT.
- SBS, on the other hand, demonstrates a relatively higher percentage of biodegradation but still not very substantial, with the highest value being 28%.

These observed values align closely with those documented in the literature, indicating 4% biodegradation for PLA after 30 days and 27% after 30 days for SBS (Papa et al., 2023). This underscores the unsuitability of a mesophilic anaerobic digestion process for such types of bioplastics, suggesting that alternative collection and treatment methods may be more effective. Table 9 illustrates the methane production contribution from the three matrices entering the plant, namely the organic fraction, PLA, and SBS. It is noteworthy that, in the case of SBS, the gas volumes produced, considering an input flow of 100 kg/h at 85% moisture (97% OFMSW and 3% bioplastics), are not negligible compared to those produced by the organic fraction alone, especially for SBS. This is attributed to the significantly higher volatile content of bioplastics compared to the organic fraction alone.

Fig. 7 illustrates the performance of biopolymers over time under thermophilic conditions. These elevated temperatures contribute to higher mass loss values for both bioplastics:

- PLA exhibits approximately ten times more degradation compared to the mesophilic case. Specifically, while PLA achieves a maximum biodegradation of 4% at 37 °C, it reaches up to 42% under thermophilic conditions.
- SBS experiences a mass loss of 44%, indicating that the increase in temperature has a comparatively lesser impact on the biodegradation of SBS than that of PLA.

Similar findings are present in the literature, where a 25–35% biodegradation is reported following 20 days of anaerobic digestion in thermophilic conditions. However, discrepancies arise when evaluating after 30 days, with some studies reporting values around 50%. This variability is likely attributed to differences in sample sizes and crystallinity (Yagi et al., 2012). Despite the increased biodegradation rates compared to the mesophilic case, it remains evident that the current treatment conditions are not entirely suitable, suggesting the exploration of alternative recycling routes.

Table 8
Kinetic constant value for SBS composting.

Temperature (°C)	k (d^{-1})
55	$5.7*10^{-2}$
37	$0.84*10^{-2}$
25	$0.42*10^{-2}$

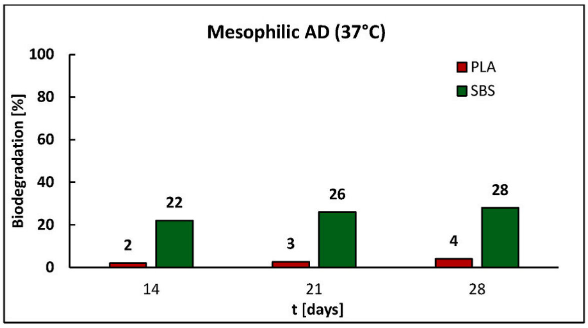

Fig. 6. Biodegradation percentages under mesophilic conditions.

Table 9
Biogas contributes to mesophilic conditions.

Temperature (°C)	HRT (d)	Total biogas (m^3/h)/(Nl/kg$_{SV}$)	OF biogas (m^3/h)/(Nl/kg$_{SV}$)	SBS biogas (m^3/h)/(Nl/kg$_{SV}$)	PLA biogas (m^3/h)/(Nl/kg$_{SV}$)
37	14	3.8/377.4	3.5/266.5	0.3/95.0	0.05/15.8
37	28	4.3/482.4	3.6/276.5	0.6/174.2	0.1/31.7

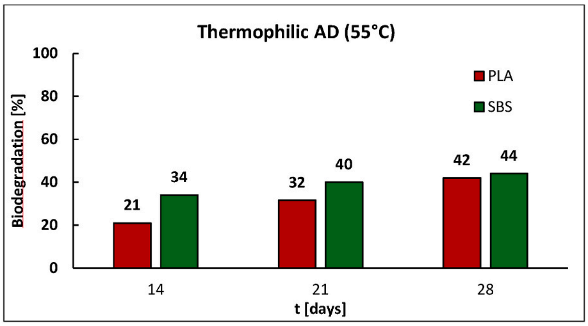

Fig. 7. Biodegradation percentages under thermophilic conditions.

Table 10 displays the methane production values for each of the three matrices under thermophilic conditions, considering an input flow of 100 kg/h at 85% moisture (97% OFMSW and 3% bioplastics):

- PLA demonstrates notable potential producibility values, reaching 294.5 Nl$_{Biogas}$/kg$_{SV}$. These results underscore an improvement in the process's performance on the organic fraction, as thermophilic conditions yield 294.5 Nl$_{Biogas}$/kg$_{SV}$ compared to the mesophilic case, which produces 31.7 Nl$_{Biogas}$/kg$_{SV}$.
- Additionally, it is observed that SBS exhibits more adaptable properties concerning degradation concerning temperature. Its kinetic constant of biodegradation during anaerobic digestion is less sensitive to temperature changes. However, the degradation profile is not linear; as HRT increases, the process slows down. This behavior is attributed to the composition of the material under assessment, comprising starch (easily biodegradable) and PBAT (biodegradable but with longer times). Therefore, a high reaction kinetics is observed in the initial two weeks due to starch, followed by a slowdown due to PBAT.

3.2. *Composting*

This section highlights the biodegradation behavior of bioplastics to evaluate their effective biodegradation.

In Fig. 8, bioplastic degradation levels can be observed as a function of the phase of the aerobic process to which they are subjected, applying the times and the temperature range (see paragraph 2.1) currently present in most plants.

The results show that SBS degrades better than PLA, especially in the first active phase. As mentioned above for the SBS, in the first phase, the rapid degradation of the starch-based component is highlighted. After only 10 days, the SBS reached a degradation of 57%. The results, compared to AD, showed that SBS degrades better under aerobic conditions; in fact, the reaction kinetics for SBS in aerobic processes is greater than observed in anaerobic processes, in line with the literature (Cucina et al., 2021). These results differ from the results presented by (Musioł et al., 2016). Specifically, their findings indicate that, for PLA, the mass loss is 30% after 7 days and approximately 43% after 21 days. Discrepancies in results could stem from various factors, including variations in operating temperatures. Notably, their investigation focuses on the composting process of PLA occurring at 70 °C. Analyzing the results, it suggests

Table 10
Biogas contributes under thermophilic conditions.

Temperature (°C)	HRT (d)	Total biogas (m^3/h)/(Nl/kg$_{SV}$)	Biogas from OF (m^3/h)/(Nl/kg$_{SV}$)	Biogas from SBS (m^3/h)/(Nl/kg$_{SV}$)	biogas from PLA (m^3/h)/(Nl/kg$_{SV}$)
55	14	5.7/711.3	4.56/350.3	0.7/221.7	0.44/139.3
55	28	6.6/965.1	4.69/360.3	0.98/310.3	0.93/294.5

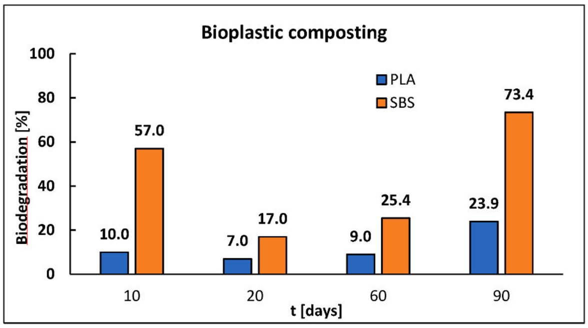

Fig. 8. Biodegradation percentages after composting section.

that BSS may exhibit potential suitability for composting, whereas PLA may require alternative recycling methods.

3.3. Integrated process

This section presents the results of the whole integrated process, with anaerobic digestion (mesophilic or thermophilic) as a function of time, in the range of 14–28 days, followed by the composting section where HRT, typical for current plants, is fixed at 10, 20 and 60 days for the sanitization, stabilization, and maturation phases, respectively. Figs. 9 and 10 show the mass loss rates of bioplastics subjected to the integrated process by applying actual plant conditions, and show the values related to anaerobic digestion alone to highlight the influence of the processes.

Figs. 9 and 10 reveal the following observations:

- For SBS, a marginal difference in degradation is observed when simulating the integrated process with the anaerobic digestion section under thermophilic and mesophilic conditions. This implies that the composting process predominates in the overall degradation process, with a difference of 4 percentage points—85% in the thermophilic case and 81% in the mesophilic case.
- PLA exhibits a biodegradation level of 56% in the most intense case (28 days of anaerobic digestion), more than double that of the mesophilic anaerobic digestion scenario. This underscores the heightened influence of the anaerobic process on PLA. In the case of mesophilic anaerobic digestion, PLA displays greater degradation during the composting phase, whereas in the simulation with anaerobic digestion in thermophilic conditions, a significantly larger proportion of degradation occurs in the anaerobic digestion section. Specifically, it accounts for 52% after 14 days of anaerobic digestion and rises to 75% with 28 days of anaerobic digestion. This is substantially higher comparedto the mosophilic anaerobic digestion scenario, where it constitutes only 15% of the process.

4. Conclusions

In this study, the goal is to provide a tool for evaluating the biodegradation of popular biopolymers collected with organic waste by simulating real plant conditions. The biodegradation of PLA and starch blend shopper (SBS) was simulated and evaluated in an

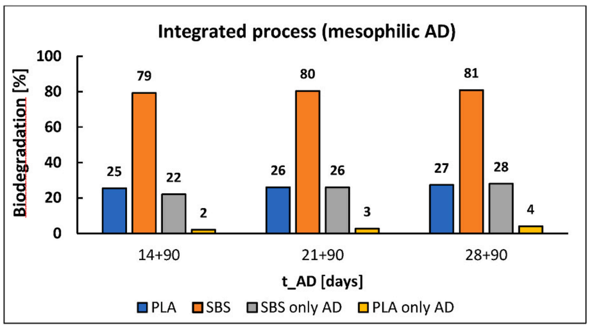

Fig. 9. Biodegradation percentages in integrated plants with mesophilic AD.

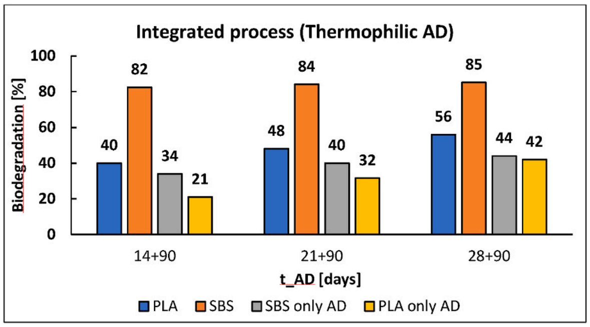

Fig. 10. Biodegradation percentages in integrated plants with mesophilic AD.

integrated anaerobic/aerobic plant using Aspen Plus software. In particular, the integrated process model allowed an in-depth study of the behavior of bioplastics when exposed to real plant conditions, whereas today biodegradability tests are performed on a small scale and under conditions far from real conditions (e.g., hydraulic residence time). The main objective is to analyze the biodegradation efficiency during the anaerobic, aerobic, and integrated real process to assess the compatibility between waste and the recycling method.

The main results can be summarized as follows:

- SBS presenting a maximum biodegradation of 85% with 28 days of AD if current plant conditions are applied operating by means of integrated process when both mesophilic and thermophilic anaerobic digestion is applied.
- SBS, during composting, reach 73.4% of biodegradation, while it resulted lower during anaerobic digestion reaching a maximum of 44% during a 28-day thermophilic AD.
- PLA shows its maximum biodegradation in the integrated process characterized by 28-day thermophilic anaerobic digestion, registering the 56% of biodegradation. In composting the highest observed biodegradation was 23.9%; in anaerobic digestion PLA biodegradation reached the 42% during a 28-day.
- Due to their high volatile matter content, bioplastics make up only 3% of the input stream, but contribute to biogas production for a 14.1% under mesophilic conditions and 28.3%, under thermophilic for duration of the anaerobic process is 28 days respect a total production of biogas from OFMSW and biopolymers AD.
- There is a need for the scientific community to assess the biodegradation kinetics of bioplastics under real conditions in biodegradation facilities to address current limitations, including reliance on kinetic data primarily derived from controlled laboratory tests. Consequently, one of our forthcoming endeavors involves evaluating the kinetic behavior of bioplastics through full-scale experiments.
- It appears that improvements in plant conditions, particularly in terms of hydraulic residence time, are essential for enhancing the performance of PLA artifacts. In the case of SBS, composting emerges as a suitable process, while anaerobic digestion may not be very suitable unless hydraulic residence time is increased. Clearly, these modifications in operating conditions will impact the requirements for operators.

The key determinant of success for these processes lies in the efficiency and effective management of the provided facilities. In the case of PLA, alternative forms of recycling, such as chemical recycling through pyrolysis, which enables the recovery of fundamental precursors like lactide and lactic acid, could be contemplated.

CRediT authorship contribution statement

D. Sorino: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Validation, Writing – original draft, Writing – review & editing. **L. Bartolucci:** Supervision, Writing – original draft, Writing – review & editing. **S. Cordiner:** Supervision, Writing – review & editing. **G. Costa:** Supervision, Writing – review & editing. **F. Lombardi:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Supervision, Validation, Writing – review & editing. **V. Mulone:** Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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