Abstract

Anaerobic digestion of bioplastics’ wastes may represent a valuable disposal strategy for these important substitutes for plastics, in order to reduce their leakage into the environment and to produce bioenergy in the form of biomethane, contributing positively to the sustainability of the entire bioplastics’ life chain. Only lab-scale data about bioplastics’ anaerobic degradability and biomethane production have been produced until now, using approaches far from full-scale conditions. This paper presents a novel approach in studying the potential of bioplastics to produce biomethane because a pioneering methodology was adopted, allowing acquisition of full-scale data that can be useful to further attest environmental and economic sustainability in recovering bioplastics’ wastes. A simple and replicable experimental approach was proposed to studying bioplastic degradability during anaerobic digestion, which consisted of placing the bioplastics into a full-scale digester within perforated steel boxes. Three different bioplastics items (one starch-based and two polylactic-acid based) were co-digested with organic wastes in a real anaerobic digestion plant using the real process parameters (thermophilic temperature for 30 days of hydraulic retention time followed by 30 days of mesophilic maturation). The experiments were replicated at laboratory-scale to evaluate the potential for biomethane recovery from bioplastics degradation.

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 and a different degradation mechanism for the two types studied was found through Fourier Transform-InfraRed spectroscopy (FTIR) analysis. Starch-based bioplastics showed a quick consumption of the starch component, followed by a slow degradation of the polyester fraction. Polylactic acid bioplastics were degraded without chemical changes to their composition. In both laboratory-scale and full-scale experiments, temperature was a key parameter affecting bioplastics' degradation, i.e., thermophilic temperatures were needed to obtain a significant degradation. Biomethane potential determination of the bioplastics (135±23 $NLCH_4$ kg Volatile Solids^{-1} as average at the end of the thermophilic digestion) proved that anaerobic digestion of bioplastics may be a sustainable approach, reducing bioplastic leakage and producing bioenergy (biogas), respecting Circular Economy principles. Anaerobic digestion may represent a valorisation treatment for bioplastics' wastes contributing positively to the sustainability of the entire bioplastics' life chain.

Keywords

Biogas; Circular Economy; Polylactic acid; Starch-based bioplastics; Waste management

1. Introduction

In 2018, the European Commission adopted the European Strategy for Plastics in a Circular Economy, i.e., plastic wastes are circulated through a new supply chain and are consumed completely in the final stage (European Commission, 2018a). The strategy is part of a wider plan to develop a Circular Economy and it comes from the 2015 Circular Economy Action Plan that identified plastics as a priority area (European Commission, 2015). Plastics have changed human life and they are found in almost every item ranging from food and household packaging materials, construction materials, fabrics, agriculture tools and automobile parts (PlasticsEurope, 2020). The excessive use of plastics, their long half-life, and the improper management of plastics' wastes have caused the accumulation of plastics and microplastics in almost all environments, particularly in the sea (Bellasi et al., 2020). 90% of all plastics are derived from virgin fossil feedstocks and account for about 6% of global oil consumption, and it is expected that plastics production will account for 20% of oil consumption by 2050 (Matthews et al., 2021). Plastics recycling is still facing important technical limits (i.e., the presence of contaminants that limits their reuse in food packaging) (Geueke et al., 2018).

Bioplastics emerged as potential substitutes for fossil-fuel derived plastics, since they are defined as biodegradable and/or bio-based materials (European bioplastic, 2020). The biodegradation process is defined as polymer degradation by biological microorganisms into carbon dioxide, water, biomass and methane by composting, soil biodegradation, marine biodegradation, or other biodegradation processes (Meereboer et al., 2020). It is also termed biotic degradation and can be enhanced or started after some initial abiotic degradation processes occur, and finally it leads to a reduced environmental persistence of bioplastics in comparison with plastics (Emadian et al., 2017). Bioplastics are expected to degrade in natural environments in a few years (Cucina et al., 2021a), in comparison to other plastics that will require hundreds/thousands of years (Chamas et al., 2020). However, recent literature revealed that the claims of biodegradability can be ambiguous if further information regarding the timeframe, the conditions necessary for biodegradation and the level of biodegradation are not provided (Bátori et al., 2018). For example, Cucina et al. (2021b) showed that bioplastics items certified as biodegradable did not undergo a complete degradation during mesophilic biological treatments, partly due to the high concentration of this material in organic wastes tested. Ruggero et al. (2020) and Zhang et al. (2018) proved that bioplastics need a long time to degrade under composting and anaerobic digestion conditions, respectively. Degree of biodegradability can be evaluated through different methodologies, i.e., mass loss, evolution of carbon dioxide or methane (for aerobic and anaerobic degradation, respectively) or by means of FT-IR investigation (Ruggero et al., 2019).

Being bio-based means that bioplastics are obtained from renewable resources (Emadian et al., 2017), decreasing their carbon footprint and increasing their sustainability (Bátori et al., 2018). Starch-based and polylactic acid (PLA)-based bioplastics are the most common biodegradable bio-based bioplastics, and their market is expected to grow fast in the coming years (European bioplastic, 2020). PLA is obtained through the microbial fermentation of carbohydrates to lactic acid and its subsequent polymerization, and it is mainly used to produce durable and disposable goods. Starch-based blends are made of starch obtained from different crops processed with plasticizers and find their main applications in the production of films, carrier bags and waste-collection bags. Bioplastics certified as compostable are collected within biowaste and their disposal follows waste management strategies used for organic wastes (i.e., anaerobic digestion and/or composting). Compostability is defined by four conditions which need to be respected: biodegradability, disintegrability, no toxicity towards composting microorganisms and no adverse effects on compost quality (Emadian et al., 2017). Bioplastics end-of-life through composting have been recently investigated from a sustainability point of view by Zhao et al. (2020), who showed that bioplastics had a high global warming potential if industrial composting and disposal transportation were considered as their end-of-life strategy using a life cycle assessment analysis. This was mainly because composting and related operations, i.e., transportation, are energy-requiring processes. In the light of this, other strategies should be investigated for bioplastics end-of-life treatments in order to meet the principles of the Circular Economy, i.e., material recovery and/or bioenergy production (Cucina et al., 2021a). Anaerobic digestion (AD), which is the anaerobic conversion of organic matter into an energy-rich gas mixture (biogas) and an organic fertilizer (digestate) appears to be a promising disposal strategy for bioplastics. AD would not only reduce bioplastics leakage into the environment through their degradation, but could convert bioplastics into biomethane, increasing their circularity.

According to recent literature, anaerobic degradability of bioplastics depends mainly on bioplastics composition (i.e., polyhydroxyalkanoates degrade better than PLA-based and starch-based bioplastics) (Cucina et al., 2021) and on the operative parameters of AD (i.e., temperature) (Abraham et al., 2021). The main findings of recent literature dealing with anaerobic degradability of bioplastics are summarized in Table 1.

Thermophilic temperatures (55-58°C) are mandatory to obtain an effective degradation of bioplastics during AD due to the need to reach the glass transition temperature (Cucina et al., 2021a). Mesophilic conditions do not reach the glass transition temperature (Cucina et al., 2021b) which is needed to increase bioplastics hydrophilicity, as reported by Yagi et al. (2013 and 2014) studying the mesophilic and thermophilic degradation of PLA. Cazaudehore et al. (2021) showed that thermophilic conditions were needed to improve anaerobic degradation of bioplastics-based coffee capsules, with mesophilic conditions being almost ineffective.

The topic of anaerobic biodegradability of bioplastics and the potential production of biomethane from their degradation still presents some serious research gaps:

- No data are yet available from full-scale studies.
- All the knowledge on bioplastics degradation during AD has come from laboratory-scale experiments, often using process conditions established by standard methods, which can be far from full-scale conditions. For instance, when Cucina et al. (2021b) used bioplastics concentrations higher than those prescribed by standard methods to test their degradation during AD and composting, the results were far from the complete degradation expected.
- Lab-scale experiments usually did not test bioplastics degradability using continuous conditions that better simulate real scale anaerobic digestion processes. Batch tests are generally carried out, as described by Cazaudehore et al. (2021) to test anaerobic biodegradability of coffee capsules made of different bioplastics.
- Laboratory-scale data may not agree with full-scale data, as described by Bolzonella et al. (2005), who reported significant differences between full-scale and laboratory-scale results from activated sludge anaerobic digestion.

Considering all these research gaps and that bioplastics are expected to represent a considerable fraction of organic wastes in the coming years (Cucina et al., 2021b), the need for full-scale studies on anaerobic degradability of bioplastics is evident.

The aim of the research presented here was to assess the anaerobic degradability of three different bioplastics (one starch-based and two PLA-based items) using full-scale AD operating under continuous conditions. This approach was possible using an innovative and replicable experimental system that allowed the co-digestion of the bioplastics with organic wastes in a full-scale thermophilic AD facility using the process parameters of the plant (feeding, hydraulic retention time, temperature). The experiment was replicated at laboratory-scale to evaluate the potential for biomethane recovery from bioplastics degradation. Quantitative and qualitative degradation were evaluated through mass loss determination and FT-IR spectroscopy, respectively, to attest the validity of using biogas production at pilot scale in comparison to the full-scale approach. This expedient was important because for the first-time biogas production of bioplastic at full-scale was reported, these data being important for further study, attesting environmental and economic sustainability in recovering waste (Sarkar et al., 2022a) in the framework of the circular economy (Sarkar et al., 2022b).

Description of the materials, experimental design and methods used in the present study are given in Section 2.

Table 1. Recent findings in bioplastics' anaerobic degradability.

Bioplastic	**Process**	**Time (d)**	**Temperature (°C)**	**Degradation (% w/w)**	**Research gap**	**Reference**
Starch-based	Pilot-scale batch AD	35	35	29.5	No full-scale assessment; No continuous process	Cucina et al., 2021b
Starch-based	Lab-scale batch AD	100	35	12	No full-scale assessment; No continuous process	Cazaudehore et al., 2021
Starch-based	Lab-scale batch AD	100	55	47	No full-scale assessment; No continuous process	Cazaudehore et al., 2021
Starch-based	Lab-scale batch AD	40	50	> 71	No full-scale assessment; No continuous process	Dolci et al., 2022
Starch-based	Lab-scale semi-continuous AD	40	50	< 27	No full-scale assessment	Dolci et al., 2022
PLA	Pilot-scale batch AD	35	35	3.7	No full-scale assessment; No continuous process	Cucina et al., 2021b
PLA	Lab-scale batch AD	65	35	20	No full-scale assessment; No continuous process	Zhang et al., 2018
PLA	Lab-scale batch AD	65	55	18	No full-scale assessment; No continuous process	Zhang et al., 2018
PLA	Lab-scale batch 1AD	100	35	24	No full-scale assessment; No continuous process	Cazaudehore et al., 2021
PLA	Lab-scale batch AD	100	55	58	No full-scale assessment; No continuous process	Cazaudehore et al., 2021

2. Materials and methods

2.1 Bioplastics

The bioplastic items studied were starch-based shoppers (SBS), PLA-based cutlery and PLA-based dishes available at Italian supermarkets. All the products were labelled as compostable by TÜV Austria (Austria) and the Italian Consortium of Composters (CIC) (Italy). Before the full-scale and laboratory-scale experiments, the bioplastics were cut into pieces of about 2.5 x 2.5 cm, which is the size used in the standard procedure to evaluate anaerobic degradability of bioplastics (ISO 14853, 2018). Full-scale and laboratory-scale experiments' design is fully described in the following subsections.

2.2 Full-scale experiment

2.2.1 Anaerobic digestion plant

The high-solid thermophilic anaerobic digestion (THSAD) plant used for the full-scale experiment is located in the Lombardy Region (northern Italy). A detailed description of the plant can be found in Pigoli et al. (2021). Briefly, the plant transforms organic wastes (i.e., sewage sludge) into an organic fertilizer (digestate), an N-based mineral fertilizer (ammonium sulphate) and energy, through a combined heat and power unit fed with biogas. The plant consists of three thermophilic digesters in series operating in thermophilic conditions (55°C) treating up to 120 kton year^{-1} of high-solid (about 18.5%) feedstock. The feedstock is represented by 85% of municipal sewage sludge and the remainder by the liquid fraction of food waste from separate collection and agro-food industry sewage sludge (Table S1). The feedstock is heated by steam injection to 55°C and mixed with digestate coming from the third digester and water to reach a solid content of about 140 g kg^{-1} fresh weight before being fed to the first digester. Digestate mixing in the reactors is guaranteed by continuous circulation through external pumps. The main operative parameters of the anaerobic digestion plant and digestate characteristics are shown in Table 2. An ammonia-stripping unit is used to control the total ammonium-N level in the digestate and allows the production of ammonium sulphate solution (Pigoli et al., 2021). At the end of the digestion process, the digestate is collected in a 53,000 m^3 storage tank prior to being used as fertilizer. The maturation stage is performed under environmental temperature (Pigoli et al., 2021) and no biogas recovery is carried out (Table 2) (Giordano et al., 2019). During the experiments, the temperature of the storage tank varied in a mesophilic range (31±3°C).

Feedstock and digestate were sampled and characterized before the beginning of the experiments (Table 2, Table S1).

Table 2. Main operative parameters of the anaerobic digestion plant and digestate characteristics.

	Parameter	**Unit**	**Value**
Thermophilic anaerobic digestion	HRT[a] of the sludge	Days	33
	OLR[b]	kgVS m^{-3} d^{-1}	3.2[c]±0.5
	Temperature	°C	55±2
Mesophilic maturation	HRT of the digestate	Days	37
	Temperature	°C	35±5
Digestate characteristics	Total solids	%	11±1.3
	Volatile solids	% d.m.[d]	60.4±2.4
	pH	pH unit	8.3±0.2
	Total organic C	g kg^{-1} d.m.	297±34
	Total N	g kg^{-1} d.m.	70±5
	C/N		4.2
	Ammonium-N	g kg^{-1} d.m.	36±5

[a] Hydraulic retention time of the sludge

[b] Organic loading rate

[c] Mean value ± Standard Deviation (n=3)

[d] Dry matter

2.2.2 Experimental design: full-scale experiment

After being cut into pieces of about 2.5x2.5 cm, bioplastics were placed in cube shaped boxes made of steel with sides of 10 cm square (1 L volume) (Figure 1a). The walls of the boxes were perforated to obtain a 2 mm mesh. This permitted the entrance of the digestate into the boxes and ensured a proper contact between digestate and bioplastics. In addition, 2 mm mesh was selected by taking into consideration standard procedures for bioplastic degradation assessment and Italian legislation concerning high quality fertilizers. Bioplastics are considered to have disintegrated when the size of the residues is under 2 mm (ISO 20200, 2016). From a fertilizer quality point of view, all inert materials (i.e., plastic, glass, metal, and bioplastic) with a size under 2 mm are not considered (EC, 2019).

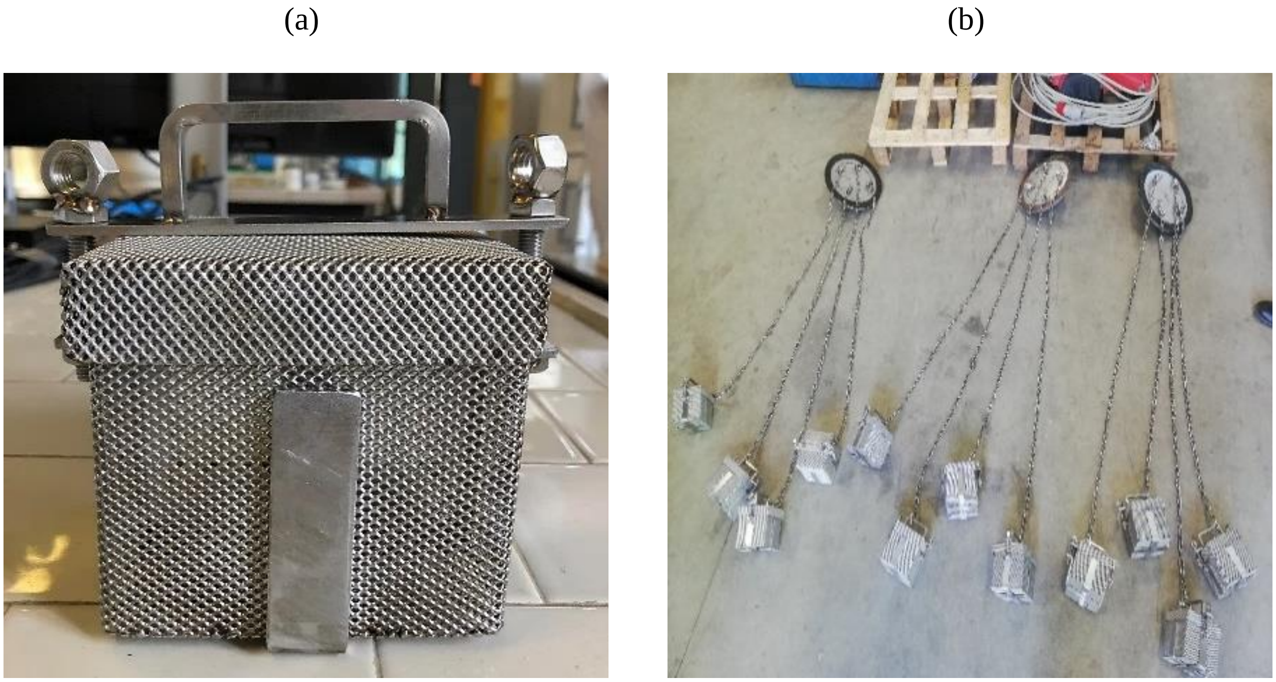

Figure 1. (a) Steel boxes used for sampling of bioplastics in the full-scale experiment; (b) System used to introduce the steel boxes inside the anaerobic digestor.

Twelve boxes were used for the experiment: 6 were filled with 10 g of starch-based shoppers (SBS) and the remaining boxes with 10 g of cutlery and 10 g of dishes. PLA-based items were placed in the same box since they were easily recognizable, whereas SBS were kept alone due to their high volume that completely filled the box at the beginning of the experiment. The boxes filled with bioplastics were inserted inside the first digester tank through a system of chains and grouped (2 SBS boxes + 2 PLA-based boxes) to allow three samplings (Figure 1b).

Sampling times were chosen in accordance with the operative parameters of the AD plant (i.e., hydraulic retention time, HRT).

The first two samplings were carried out at 15 and 30 days. At day 30, the remaining four boxes were transferred to the storage tank, where the mesophilic maturation phase took place for another 30 days (for a total duration of 60 days). After each sampling, bioplastics were recovered, washed to remove the digestate and dried in a ventilated oven at 37 °C until a constant weight was reached. Dried bioplastics were weighed to determine the percentage of degradation according to the equation:

$$\%\ degradation\ = \left(\frac{mass\ t0 - mass\ t}{mass\ t0}\right)\ x\ 100$$

Where *mass* t_0 is the initial mass of bioplastics, whereas *mass t* is the residual mass of bioplastics at the sampling time.

2.3 Laboratory experiment

Biochemical methane potential (BMP) assays were carried out by modifying the procedure described in Schievano et al. (2008). The inoculum was the digestate collected from the same digester of the AD plant where full-scale degradation was assessed. The three bioplastics items were studied separately in the laboratory experiments.

In 500-mL serum bottles, 3 g of bioplastic were added to 300 mL of inoculum. The batch tests were carried out with 300 mL of material and 200 mL of headspace. Control blanks were prepared using 300 mL of inoculum without bioplastics and the positive control was performed with 3 g of cellulose.

All batches were sealed with Teflon hermetic caps, flushed with a N_2 atmosphere, and incubated at 55±2°C for 30 days. Then the bottles were transferred into a second oven maintained at 35±2°C until the end of the experiment (60 days) to replicate the full-scale experiment.

Assay bottles were periodically analysed for both quantitative and qualitative determination of biogas production. Quantitative biogas production was estimated by withdrawing extra-pressure gas with a 100-mL syringe. Biogas production of blank control batches was subtracted from biogas production of every sample. Qualitative characterization of biogas was performed by a gas chromatograph (Carlo Erba Megaseries 5300, capillary column 25-m x 0.32-mm diameter and flame ionization detector -FID) to determine CH_4–CO_2 ratio in the biogas. The carrier gas was nitrogen at 20 kPa pressure and temperatures of injector and FID were 130 and 150°C, respectively. Comparison of obtained peak areas was carried out with a standard gas mixture of 30-70 CH_4-CO_2. The only difference between the full-scale and laboratory-scale experiments was that in the laboratory, the strict anaerobic conditions were maintained in order to evaluate the amount of biomethane produced during maturation.

In addition to the bottles used for BMP determination, a series of bottles were set up to allow destructive sampling at 15 and 30 days. These samples were used for degradation evaluation through mass loss determination. To do this, bioplastics were removed, rinsed, dried and weighed to quantify the amount of substrate degraded following the same procedure described for the full-scale experiment.

Degradation of bioplastics in the laboratory-scale experiment was also evaluated by calculating it from the biomethane potential. The quantity of bioplastic degraded was estimated taking into account the amount and composition of biogas at each sampling time and performing stoichiometric calculations.

2.4 Analytical procedures

2.4.1 Chemical analysis

Chemical analyses on feedstock and digestate fresh samples were carried out following standard procedures. Total solids (TS) and Volatile solids (VS) were determined according to standard procedures of the American Public Health Association (APHA, 2017). Total organic carbon (TOC) determination was carried out by the wet acid oxidation method following standard methods (APHA, 2017). pH was determined in aqueous solution using a 1:2.5 sample/water ratio and a pH probe (US Department of Agriculture - US Composting Council, 2002). Total N and ammonium-N were determined according to the analytical method for wastewater sludges (APHA, 2017).

2.4.2 Spectroscopic analysis

Bioplastics were characterized by spectroscopic investigation, using the FT-IR spectra, which were collected in total reflectance mode (ATR) with a Shimadzu IRAffinity-1S equipped with a Miracle Pike ATR device (Shimadzu Italia srl, Milano, Italy). The investigated wavenumber range was of 4,000–500 cm^{-1} and the resolution was of 2 cm^{-1}. Bioplastic samples were dried, cleaned and gently brushed by tooth-brush to remove all the deposits formed on their surfaces before spectroscopic analysis.

Peak areas were determined using Shimadzu LabSolutions IR software (Shimadzu Italia srl, Milano, Italy).

2.5 Statistics

All experiments and analyses were replicated three times except for the full-scale experiments, which were duplicated. Mean and standard deviation values were calculated according to standard procedures (Microsoft Excel Software). Determination of significant differences among the parameters analyzed at a level of significance of $P<0.05$ was carried out by analysis of variance (ANOVA) and Tukey's test (Microsoft Excel Solver 2013).

3. Results and discussion

3.1 Characterization of bioplastics

FT-IR investigation of bioplastic materials before the experiment showed that SBS were bio-composites, probably constituted by starch and a biodegradable polyester (Elfehri Borchani et al., 2015) (Figure 2a). Four diagnostic peaks were visible in the spectrum (Table S2). The presence of absorption bands at around 3,400 cm^{-1} and 1,050 cm^{-1} were attributed to O-H and C-O functional groups of starch as suggested by Elfehri Borchani et al. (2015). The other two bands were assigned to the polyester component (1,730 cm^{-1} and 730 cm^{-1}) (Ruggero et al., 2020).

The spectra of PLA-based cutlery and dishes are reported in Figures 2b and 2c, respectively. PLA-based cutlery was found to be mainly composed of polylactic acid. The spectra were characterized by four diagnostic peaks that are commonly referred to polylactic acid (3,000, 1,750, 1,400 and 1,100 cm^{-1}) (Table S2). PLA-based cutlery presented a more homogeneous composition than the PLA-based dishes. PLA-based dishes showed an intense peak at around 1100 cm^{-1} which may have covered the diagnostic peak of the ether group of PLA. According to the literature, PLA-based dishes may be blended with poly(butylene succinate) (PBS) or cellulose, which are polymers that absorb IR in that region (de Matos Costa et al., 2020).

Starting from the characterization of the bioplastics studied, the results of bioplastics' degradation at the end of the experiments in both quantitative and quality terms, are reported in Sections 3.2 and 3.3.

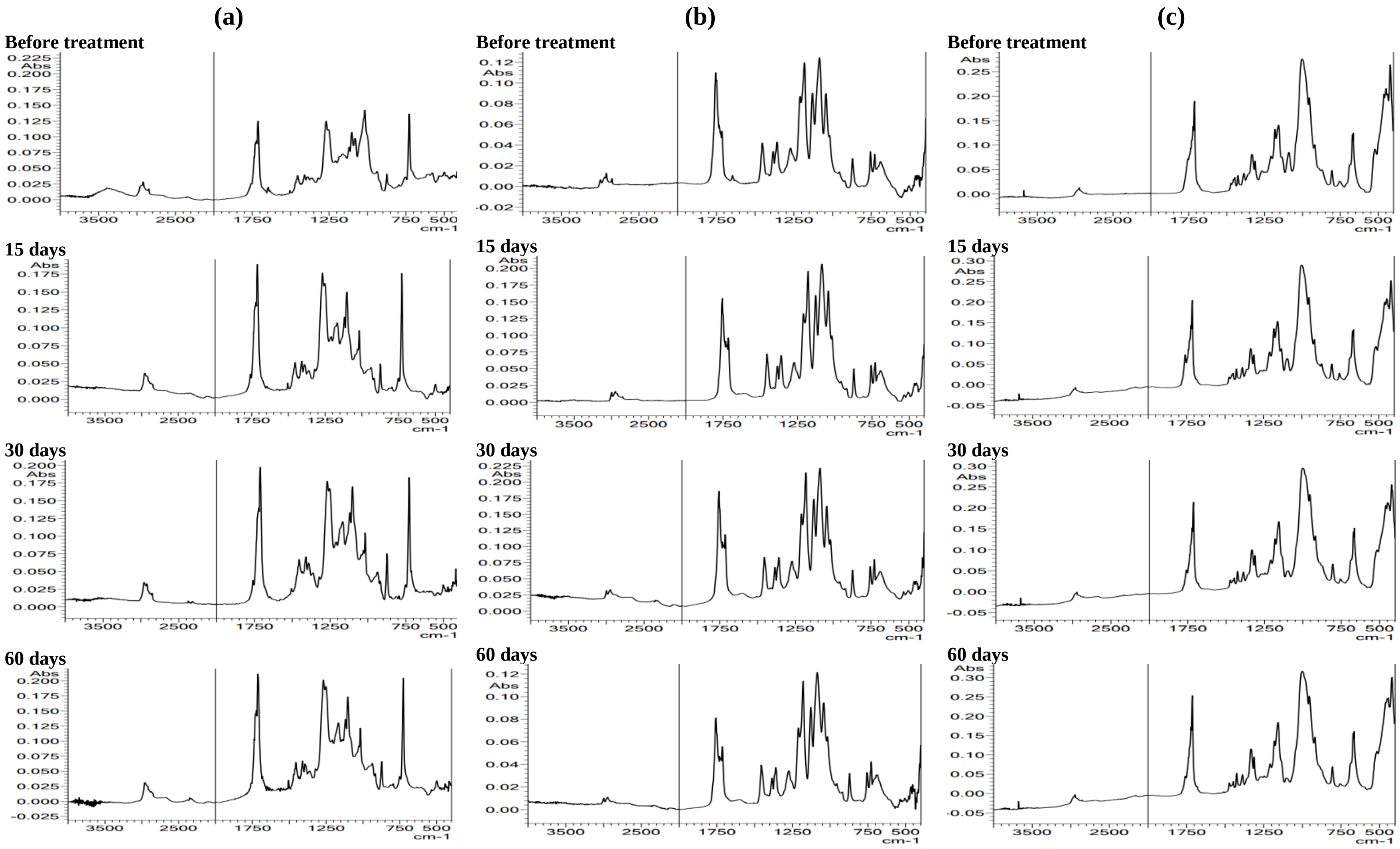

Figure 2. FT-IR spectra of bioplastics during the full-scale experiment. (a) Starch-based shoppers, (b) PLA-based cutlery, (c) PLA-based dishes.

3.2 Degradation of bioplastics in the full-scale experiment

Before evaluating the degradability of bioplastics, visual analyses were carried out after each sampling (15, 30 and 60 days) to highlight any possible change in terms of consistency, thickness, discolouring and erosion of the material (Ruggero et al., 2019). Examples of visual inspection of bioplastics at the end of the full-scale experiment (60 days) are shown in Figure S1.

SBS appeared intact and they did not show any significant changes in their appearance. Once dried, SBS were less transparent and from a tactile point of view, they had become less elastic than the initial SBS. Conversely, PLA-based items showed some visual modifications i.e., they were crumbly. Furthermore, PLA-based dishes presented a surface covered by bubbles (Figure S1). Similar results were reported by Zhang et al. (2018), who investigating the degradation of compostable bioplastics during mesophilic anaerobic digestion of food wastes, reporting that both starch-based and PLA-based bioplastics showed few changes after anaerobic digestion, i.e., slight changes in color, deformation and/or fragmentation.

The results of degradation of bioplastics during the full-scale experiment are reported in Table 3. After 60 days of treatment (thermophilic AD and mesophilic maturation stage), SBS was more degraded in comparison to PLA-based items. SBS degraded by about 30.9% on a weight basis, while PLA-based cutlery and PLA-based dishes degraded by 27.9% and by 21.4% on weight basis, respectively.

Table 3. Degradation of bioplastics during full-scale and laboratory-scale experiments.

		Starch-based shoppers		**PLA-based cutlery**		**PLA-based dish**	
Degradation[a]	*Unit*	*Full-scale*	*Lab-scale*	*Full-scale*	*Lab-scale*	*Full-scale*	*Lab-scale*
15 days	%[b]	23.6±0.5[c]	21.7±1.9[d]	6.7±0.1	6±0.5	7.9±1.5	7.8±1.2
30 days	%	26.4±2.2	28.7±0.9	21.6±3.2	23.3±0.0	18.3±2.3	19.7±0.5
60 days	%	30.9±1.3	30±1	27.9±2.2	29.2±1.2	21.4±0.7	24.2±1.2

[a] Determined from mass loss

[b] % weight basis

[c] Mean value ± Standard Deviation (n=2)

[d] Mean value ± Standard Deviation (n=3)

The sampling of bioplastic items at different times (15, 30 and 60 days) also allowed us to evaluate the kinetics of degradation. SBS degradation mainly occurred in the first 15 days, after which a plateau was reached (Table 3). Since SBS FT-IR spectrum proved the presence of two different components, probably in the first period (15 days) the starch was preferentially degraded, while the polyester component needed more time (Cucina et al., 2021b). This is in accordance with Ruggero et al. (2020), who reported that the starch amount in SBS can range from 20 to 60 % on a weight basis and that it is preferentially degraded with respect to polyesters and additives during biological treatments. PLA-based cutlery and dishes showed a different trend of degradation. The kinetics of PLA-based items in the 30 days of thermophilic anaerobic digestion occurred following a linear kinetic model, in accordance with literature findings (Chinaglia et al., 2018). For instance, Cucina et al. (2021b) reported a linear kinetic degradation for PLA based items during mesophilic biological treatments, suggesting that this material was degraded with a kinetic that was not affected by the total amount of bioplastic, but it was by the available substrate.

After 30 days of thermophilic anaerobic digestion, the bioplastic items were transferred into the digestate storage tank under mesophilic conditions, and this slowed down bioplastics' degradation. Bioplastics continued to degrade but a reduced kinetic was observed, confirming the fundamental role of temperature in degradation (Cucina et al., 2021a). It is well known that reaching the glass transition temperature of bioplastics (about 55-60°C) is necessary in order to modify the structure of the polymer from the crystalline to the amorphous structure. The latter was found to be more hydrophilic than the crystalline structure, which also resulted in an improved biodegradability of the polymer (Gil-Castell et al., 2020). Even if the decrease of degradation kinetic during the mesophilic maturation of digestate was expected, these data represent a valuable and unique result because there are no data in the literature dealing with bioplastics' degradation during digestate maturation.

Digestate storage in mesophilic conditions is now a well-established AD-post treatment (Lu and Xu, 2021). Therefore, getting information on bioplastics degradation during maturation becomes important. Data indicated that bioplastics did not undergo significant degradation at the maturation stage, and so, that it is mandatory to improve bioplastic degradation during the AD process to get full results, i.e., degradation and biogas production.

Bioplastics' degradation during the full-scale experiment was also evaluated at qualitative level by using FT-IR analysis. Spectroscopic analysis is commonly used as a tool to study how chemical structures have been modified by microorganisms during the degradation process (Ruggero et al., 2019). The evolution of SBS spectra during the full-scale experiment (Figure 2a) confirmed the previous findings. Preferential consumption of the starch in comparison with the polyester component was found. In the spectrum of bioplastic sampled at 15, 30 and 60 days (Figure 2a) the diagnostic peak of starch at 3400 cm^{-1} almost disappeared.

The spectra of PLA-based items during full-scale anaerobic digestion (Figures 2b and 2c) did not differ from the initial spectra before treatments. Although PLA-based cutlery and PLA-based dishes degraded by about 27.9% and 21.4% respectively, no changes in chemical structure were observed from their FT-IR spectra. This result confirmed the polymeric nature of PLA, which seems to degrade following a “take away” mechanism, as suggested by Cucina et al. (2021b). Following this mechanism, PLA-based bioplastics were hydrolyzed into monomers that can enter the microbial cell and then can be metabolized, leading to a slow but constant degradation. The PLA-based polymer did not undergo chemical changes with time. Since PLA-based dishes were probably composed by PLA blended with PBS or cellulose and no variations were observed in the spectra, it is likely that the two components were degraded at the same rate. These findings were validated by calculating the ratio between the areas of the diagnostic peaks (Cucina et al., 2021b) (Table 4). For SBS, two peaks were selected: 1050 cm^{-1} (starch) and 1730 cm^{-1} (polyester). The ratio between polyester and starch peaks area increased from 1.4 to 3.5 in the first 15 days. The ratio remained constant after that, ranging from 3.4 to 3.6, similarly to a report in the literature (Cucina et al., 2021b). No differences were observed when calculating the ratio between diagnostic peaks’ areas for PLA-based items.

The ratios remained constant over time, ranging from 0.11 to 0.07 for cutlery and from 0.09 to 0.08 for dishes. This confirmed that PLA degradation followed a “take away” mechanism and that other possible additives, (i.e., PBS or cellulose), degraded with a similar kinetic and mechanism.

Table 4. Ratio between diagnostic peaks areas in the full-scale and laboratory-scale experiments.

		Starch-based shoppers	**PLA-based cutlery**	**PLA-based dish**
	Time (days)	*1730 cm^{-1} / 1050 cm^{-1}*	*3000 cm^{-1} / 1750 cm^{-1}*	*3000 cm^{-1} / 1750 cm^{-1}*
Full-scale	0	1.4±0.0[a]	0.11±0.02	0.09±0.01
	15	3.5±0.1	0.09±0.00	0.08±0.01
	30	3.6±0.2	0.07±0.01	0.09±0.00
	60	3.4±0.0	0.08±0.02	0.08±0.00
Laboratory-scale	0	1.4±0.2[b]	0.11±0.02	0.09±0.01
	15	3.3±0.2	0.08±0.00	0.09±0.00
	30	3.4±0.0	0.07±0.02	0.10±0.01
	60	3.7±0.2	0.09±0.01	0.09±0.00

[a] Mean value ± Standard Deviation (n=2)

[b] Mean value ± Standard Deviation (n=3)

3.3 Degradation of bioplastics in the laboratory-scale experiment

The laboratory-scale experiment was conducted in parallel to the full-scale one in order to evaluate the amount of biomethane that could potentially be recovered from bioplastics degradation. It was necessary to evaluate in parallel the bioplastic degradation at both full-scale and laboratory-scale, since biomethane production could have been extended to full-scale if the degradation pathway were to be the same in the two experiments. The same conditions used as those in the full-scale plant were maintained during the laboratory-scale experiment and degradation and biomethane production were evaluated.

Trends of degradation in the full-scale plant were well reproduced at laboratory-scale (Table 3). Statistical analysis confirmed that no significant differences were found between quantitative degradation in full-scale and laboratory-scale experiments ($P<0.05$). At the end of the laboratory-scale experiment, SBS, PLA-based cutlery and PLA-based dishes degraded by about 30, 29 and 24% on a weight basis, respectively. These results were in accordance with recent literature dealing with bioplastics degradation in the laboratory under thermophilic anaerobic conditions. Yagi et al. (2013) studied the anaerobic biodegradation of four biodegradable polyester at 55°C and reported a degradation of PLA by about 24% in 30 days, 43% in 40 days and 68% in 60 days. Calabro' et al. (2020) studied the degradation of SBS under both mesophilic and thermophilic temperatures, showing an average degradation of 30.5% on a weight basis after 60 days. Under our laboratory conditions, SBS showed the fastest degradation kinetic in the first 15 days, followed by a significant reduction of speed of degradation in the remaining part of the experiment. PLA-based items however showed a linear kinetic during the 30 days under thermophilic conditions. In the final part of the experiment, the mesophilic temperatures used to simulate the maturation phase of the full-scale plant led to a reduction in degradation rates for both SBS and PLA-based items in the laboratory-scale experiment.

The analysis of FT-IR spectra confirmed that the degradation which occurred in the full-scale plant was well replicated by the laboratory-scale results from a qualitative point of view. Figure 3a represents the FT-IR spectra of SBS during the laboratory-scale experiment. The spectra showed a reduction of starch compared to the polyester component, resulting in an increased ratio between polyester and starch peaks areas (Table 4), similarly to data obtained in the full-scale experiment. PLA-based cutlery (Figure 3b) and PLA-based dishes (Figure 3c) did not show any significant modifications of the diagnostic peaks area. In fact, the ratio between diagnostic peaks areas remained unvaried. All these data supported the thesis that full-scale degradation may be well reproduced at laboratory-scale.

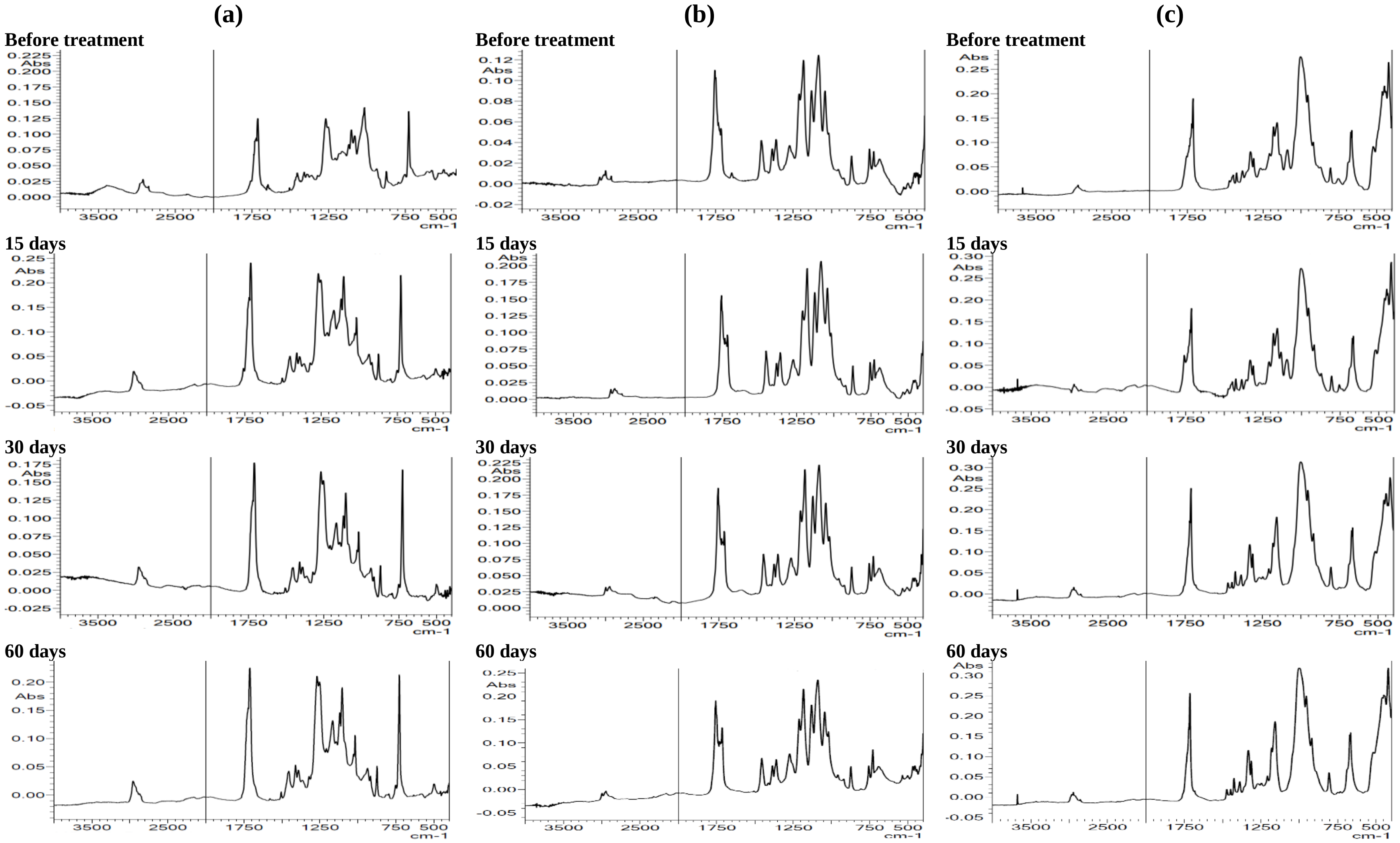

Figure 3. FT-IR spectra of bioplastics during the laboratory experiment. (a) Starch-based shoppers, (b) PLA-based cutlery, (c) PLA-based dishes.

It can be concluded that bioplastics degraded similarly at both full and lab-scale, so that data acquired for biogas production at lab-scale can be extended to the full-scale, as discussed in the following section.

3.4 Potential for biomethane recovery from bioplastics degradation

3.4.1 Biochemical methane potentials

During the laboratory experiment, biomethane production was measured to evaluate the amount of biomethane that can be produced from the degradation of bioplastics (Table 5).

Table 5. Biochemical methane potential (BMP) of bioplastics and degradation calculated from BMP.

Parameter	**Unit**	**Starch-based shoppers**	**PLA-based cutlery**	**PLA-based dish**
BMP – 15 days	NLCH_4 $kgVS^{-1}$	95[a]±12	56±25	44±22
BMP – 30 days	NLCH_4 $kgVS^{-1}$	139±24	154±31	108±27
BMP – 60 days	NLCH_4 $kgVS^{-1}$	165±28	168±11	123±14
Degradation[b] – 15 days	%	22.5	6.6	6.1
Degradation – 30 days	%	25.5	21.5	19.1
Degradation – 60 days	%	29.2	29.8	24.9

[a] Mean value ± Standard Deviation (n=3)

[b] Calculated from BMP

Biomethane production from bioplastics degradation was found to be higher under thermophilic conditions in comparison with mesophilic ones, in accordance with recent literature (Cazaudehore et al., 2021). Considering the total amount of biomethane measured during the 60 days incubation (30 days under thermophilic conditions plus 30 days under mesophilic conditions), about 84%, 92% and 88% of the biomethane was produced in the thermophilic stage for SBS, PLA-based cutlery and PLA-based dishes, respectively. At the end of the thermophilic anaerobic digestion (30 days), SBS produced 139 $NLCH_4$ $kgVS^{-1}$, while PLA-based cutlery and PLA-based dishes produced 154 $NLCH_4$ $kgVS^{-1}$ and 108 $NLCH_4$ $kgVS^{-1}$, respectively. This was probably due to changes in the mechanical properties of bioplastics that can occur only under thermophilic conditions. For instance, PLA-based bioplastics degradation under thermophilic AD was reported to be enhanced by reaching their glass transition temperature (55-60°C) that makes them more hydrophilic and accessible for microbial hydrolysis (Marek and Verney, 2016). Hamad et al. (2015) also reported that elevated temperatures (>58°C) are needed to reduce the molecular weight of PLA blends and to start their biodegradation. With reference to SBS, the higher biomethane yield in the thermophilic stage could be ascribed to the rapid anaerobic degradation of starch (Cucina et al., 2021). Since starch contributes about 30% in weight to the total SBS (Elfehri Borchani et al., 2015) and that it has a BMB of 400 $NLCH_4$ $kgVS^{-1}$ (Cucina et al., 2021c), it was calculated that there was a total theoretical contribution to BMP of SBS of 120 $NLCH_4$ $kgVS^{-1}$ that was similar to that measured during the experiment, i.e., 139 $NLCH_4$ $kgVS^{-1}$. Thermophilic conditions have also been reported to contribute to enhancing SBS degradation during AD (Gil-Castell et al., 2020). These values agreed with Cazaudehore et al. (2021) who evaluated the biomethane production from coffee capsules made of biodegradable plastics under mesophilic and thermophilic conditions. Starch-based capsules showed a biomethane potential of 160 $NLCH_4$ $kgVS^{-1}$ at 58 °C and 10 $NLCH_4$ $kgVS^{-1}$ at 38°C, and those values were not far from those obtained in this work. Other capsules made of a mixture of PLA and a fossil-based biodegradable polymer showed a biomethane potential of 200 $NLCH_4$ $kgVS^{-1}$ at 58°C and 20 $NLCH_4$ $kgVS^{-1}$ at 38°C. The biomethane potentials obtained from bioplastics in the present research at the end of the thermophilic anaerobic digestion stage (30 days) were not far from those reported in literature for other biomasses commonly used as feed in anaerobic digestion. For instance, Gunaseelan (1997) reported a biomethane potential of 190 LCH_4 $kgVS^{-1}$ for the hand-sorted organic fraction of municipal solid wastes (HS-OFMSW), whereas biomethane potentials of 180, 223 and 188 LCH_4 $kgVS^{-1}$ were reported for OFMSW, food wastes and livestock manure, respectively (Mao et al., 2015).

Degradation calculated from BMP data in laboratory experiments replicated the degradation determined from mass loss (both in full- and laboratory-scales) (Table 3, Table 5). At the end of the experiment, a degradation of 30.9% on a weight basis was found for SBS from mass loss, compared to 29.2% calculated from BMP data. Similar results were obtained for PLA-based items. This result was important to prove that bioplastics degradation was mainly due to biochemical processes. The bioplastics were likely converted to biogas, resulting in the probable absence of bioplastic residues in the digestate. From an environmental point of view, this means a reduction of potential bioplastic leakage into the environment following digestate application in agriculture.

Since quantitative and qualitative degradation of bioplastics in full-scale and laboratory-scale experiments were proved to be similar, the biomethane potentials determined may be correctly extended from the laboratory-scale to the full-scale. An attempt to evaluate the potential enhancement of biomethane production of the full-scale plant studied in this work due to bioplastics degradation is presented in the following paragraph.

3.4.2 Potential impact of biomethane production from bioplastics on the full-scale anaerobic digestion plant

Energy production in the form of biomethane can produce a benefit to bioplastics degradation during anaerobic digestion in a circular economy perspective. Bioplastics are currently collected with the OFMSW (Pagga, 1998). Their concentration in the OFMSW accounted for 3.7% in 2020 in Italy (ISPRA, 2020) and it could increase up to 10% by 2030, as estimated by Cucina et al. (2021b). Nowadays, plastics, including bioplastics, are mechanically separated at an early stage in waste treatment facilities, and are often disposed of in landfill. This appears to contradict the circular economy principles that establish a waste hierarchy which should apply as a priority order in waste prevention and management legislation and policy (i.e., prevention, reuse, recycling, other recovery, and disposal) (European Commission, 2018b). A sorted collection of bioplastics to be treated in anaerobic digestion plants as co-digestion feedstock cannot be excluded in the future.

An attempt was made to evaluate the potential for biomethane recovery from bioplastics in the full-scale anaerobic digestion plant used for the degradation assessment. This attempt was possible since degradation results obtained in the laboratory experiment did not differ from those obtained in the full-scale experiment.

Assumptions and results of this evaluation are reported in Table 6. A specific biomethane production of 200 $NLCH_4$ $kgVS^{-1}$ and a volatile solids content of 12.2% fresh weight in the feedstock were considered (Scenario 0% BP) (Pigoli et al., 2021). For the bioplastics, a 100% fresh weight content of volatile solids and a concentration of 10% fresh weight of bioplastics in the feedstock were assumed in the Scenario 10% BP. An average biomethane potential from bioplastics of 135 $NLCH_4$ $kgVS^{-1}$ (Table 6) was used in the calculations.

As reported in Table 6, bioplastics co-digested with sewage sludge in the concentration of 10% fresh weight, may significantly increase the biomethane production of the anaerobic digestion that was studied in the research plant (+45%). This biomethane increase (from 2,440 to 3,550 $NLCH_4$ $100kg^{-1}_{feedstock}$) (Table 6) may lead to a more sustainable life cycle of bioplastics. Biomethane production represents a valuable by-product of bioplastics degradation which could cover the increased cost of separate collection of bioplastics and their transport. Composting of bioplastics appears as a less sustainable strategy due to the mineralization of bioplastics to carbon dioxide without energy recovery. The production of biomethane from bioplastics degradation could reduce the utilization of fossil-derived energy sources, increasing the circularity of these promising plastics' substitutes.

Table 6. Potential for biomethane production enhancement at full-scale.

		Scenario 0% BP[a]		**Scenario 10% BP[b]**	
Parameter	*Unit*	*Bioplastics*	*Sludge*	*Bioplastics*	*Sludge*
Concentration	% fw[c]	0	100	10	90
Volatile solids (VS)	% fw	100	12.2	100	12.2
VS contained in 100 kg of feedstock	kg 100 kg$^{-1}_{feedstock}$	0	12.2	10	11.0
Biomethane potential	NLCH$_4$ kgVS^{-1}	135	200	135	200
Biomethane production	NLCH$_4$ 100kg$^{-1}_{feedstock}$	0	2,440	1,350	2,200
Total biomethane production	NLCH$_4$ 100kg$^{-1}_{feedstock}$	2,440		3,550	
Increase	%	-		**45.5**	

[a] Actual scenario: no bioplastics in the feedstock

[b] 10% weight basis bioplastics in the feedstock

[c] Fresh weight

Future research should also evaluate these aspects through a life-cycle assessment (LCA) approach. Bioplastics have been proved to have a higher global warming potential if industrial composting and disposal transportation are considered as their end-of-life strategy (Zhao et al., 2020). AD or AD coupled to digestate composting, should represent preferred treatments for bioplastics treatment (Wainaina et al., 2020). From an environmental point of view, AD based systems should be preferred as reported by Edwards et al. (2018), who showed that AD based systems were shown to outperform composting based systems for global warming potential in a LCA study of different technologies for organic wastes treatment.

3.5 Sustainability of the approach, managerial insights, and future perspectives

Considering the results obtained from this full-scale assessment of bioplastics degradation during anaerobic digestion, a few considerations may deserve discussion.

A linear degradation was observed for PLA-based items in the 30 days thermophilic anaerobic digestion, whereas SBS already seemed to reach a plateau after the first sampling (15 days). The different degradation kinetics of starch-based and PLA-based bioplastics should be evaluated using longer retention times (i.e., 60 and 90 days of thermophilic anaerobic digestion). If the different kinetics were to be confirmed using longer retention times, different management strategies for the two bioplastics should be evaluated. Supposing that the anaerobic degradation (and biomethane production) of SBS occurred mainly in the first 15-30 days of thermophilic anaerobic digestion, it would seem useless to keep these products in the digester for a longer time. Since SBS have proved to degrade better under aerobic conditions than in anaerobic conditions (Cafiero et al., 2021), these bioplastics could be preferentially treated in OFMSW treatment facilities, where integrated AD and composting systems are widespread (Lin et al., 2018). If the linear degradation of PLA-based bioplastics could be confirmed with longer retention times, strict anaerobic treatments should be preferred for these products. Linear degradation would mean a complete degradation and the recovery of the total biomethane potential within 90–150 days. These are retention times longer than the commonly used HRT (30-45 days), but they can be obtained by separating the bioplastics into the digester and working with a solid retention time (SRT) of bioplastics different from the HRT. The experimental data obtainedsuggest that different management systems for different bioplastics might enhance bioplastics' degradation and methane production, increasing sustainability of the entire bioplastics' chain (i.e., production, use, recycle). This is in accordance with Sarkar and Seo (2021) who showed that pollution due to waste can be reduced using an energy conversion policy (energy supply chain management). The data obtained in this study may serve as inputs to feed models commonly used to develop sustainable strategies for waste management in a circular framework such as those reported in Sarkar et al. (2022a) and Sarkar et al. (2022b).

Another point to be addressed in the future is the presence of bioplastics residues in the digestate. Digestate is commonly used as organic fertilizer in agriculture for organic matter and plant nutrients recovery (Scaglia et al., 2018). Bioplastics residues in digestate can leak into the soil and understanding the fate of bioplastics in the soil becomes an urgent matter. Little is known about the degradation of bioplastics in natural environments (soil, freshwater, and seawater). Recent studies have reported that bioplastics degrade in these environments with slower kinetics than those reported for engineered environments mainly due to less favourable conditions, i.e., low temperature (Beltrán-Sanahuja et al., 2020) and moisture (Accinelli et al., 2012) or low microorganisms' concentration (Rapisarda et al., 2019). Estimated times of degradation in soil of 1.5 and 4.5 years were reported for starch-based and PLA-based bioplastics (Cucina et al., 2021b). Although these times are longer than those reported for bioplastics degradation during anaerobic digestion and composting, they are much shorter than the times required for degradation of petroleum-derived plastics in soil (1,000–5,000 years) (Chamas et al., 2020). Accumulation of large amounts of bioplastics in the environment appears to be unlikely, and it may be appropriate to consider bioplastics in the soil along with other biopolymers (i.e., cellulose, lignin). More studies are needed to evaluate the degradation of bioplastics in soil: there are no studies dealing with the evaluation of bioplastics degradation in soil in the presence of digestate/compost. This topic should be further addressed since bioplastics may leak into the soil through digestate/compost application and the potential effect of additional N on bioplastics degradation should be evaluated.

Summarising, the present study aimed to evaluate the degradability of different bioplastics items under real-scale anaerobic digestion. Apart from the potential recovery of energy, the sustainability of this approach is also proved by the reduction of potential bioplastics' leakage into the environment. As previously described (Cucina et al., 2021a) the larger the amount of bioplastics degraded during waste managment, the less their leakage into the environment. This fact, coupled with biomethane production, proved that AD can represent a sustainable approach to manage bioplastics.

Improving biomethane production from bioplastics provides the basic pillars of sustainability:

- Economical sustainability because there is an increase of the total plant biomethane production increasing total renewable energy production.
- Environmental sustainability because there is both a reduction of bioplastic leakage and of GHGs produced (fossil fuel substitution).
- Social sustainability because the reuse of bioplastic producing biomethane in a circular economy frame increases the positive perception of the society for bioplastic.

All these aspects need to be further better investigated and quantified by adopting correct approaches (Sarkar et al., 2022a).

4. Conclusions

Anaerobic digestion of bioplastics is gaining interest as a sustainable waste management method because it can provide both energy production and reduction of bioplastic leakage. Until now only laboratory-scale batch or semi-continuous studies were reported, and the literature included no data from full-scale anaerobic digestion processes.

The main results obtained in this study were as follows:

- The starch component of the starch-based shoppers degraded quickly, whereas the polyester component required longer time; PLA-based items degraded without chemical modifications.
- Temperature is a key parameter for bioplastics' degradation at full-scale AD performed under continuous conditions.
- Full-scale degradation was well replicated by laboratory-scale degradation from both quantitative and qualitative points of view, so that potential biomethane production from the pilot could be adapted to the full-scale plant.
- Biomethane production from bioplastics at full-scale improved total biomethane production of plants, making bioplastic management more sustainable.
- Full-scale data produced can be useful for further studies dealing with environmental and economic sustainability.

Future research needs to clarify the effect of other variables on bioplastics' anaerobic degradation, such as i. bioplastics' composition (Abraham et al., 2021); ii. process parameters (e.g., type of feedstock, moisture regime) (Shrestha et al., 2020); iii. the effect of lengthening bioplastics retention time in the reactor (Battista et al., 2021); and iv. to assess the fate of bioplastics in soil in the case of bioplastics-rich digestate spread on agricultural soils (Cucina et al., 2021b).

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References

Abraham, A., Park, H., Choi, O., Sang, B.I., 2021. Anaerobic co-digestion of bioplastics as a sustainable mode of waste management with improved energy production – A review. Bioresource Technology. https://doi.org/10.1016/j.biortech.2020.124537

Accinelli, C., Saccà, M.L., Mencarelli, M., Vicari, A., 2012. Deterioration of bioplastic carrier bags in the environment and assessment of a new recycling alternative. Chemosphere 89. https://doi.org/10.1016/j.chemosphere.2012.05.028

APHA, 2017, 2017. APHA/AWWA/WEF (2017) Standard Methods for the Examination of Water and Wastewater.23rd Edition, American Public Health Association. American Water Works Association Water Environment FederationStable.

Bátori, V., Åkesson, D., Zamani, A., Taherzadeh, M.J., Sárvári Horváth, I., 2018. Anaerobic degradation of bioplastics: A review. Waste Management 80. https://doi.org/10.1016/j.wasman.2018.09.040

Battista, F., Frison, N., Bolzonella, D., 2021. Can bioplastics be treated in conventional anaerobic digesters for food waste treatment?. Environmental Technology & Innovation, 22, 101393.https://doi.org/10.1016/j.eti.2021.101393

Bellasi, A., Binda, G., Pozzi, A., Galafassi, S., Volta, P., Bettinetti, R., 2020. Microplastic contamination in freshwater environments: A review, focusing on interactions with sediments and benthic organisms. Environments - MDPI. https://doi.org/10.3390/environments7040030

Beltrán-Sanahuja, A., Casado-Coy, N., Simó-Cabrera, L., Sanz-Lázaro, C., 2020. Monitoring polymer degradation under different conditions in the marine environment. Environmental Pollution 259. https://doi.org/10.1016/j.envpol.2019.113836

Bolzonella, D., Pavan, P., Battistoni, P., Cecchi, F., 2005. Mesophilic anaerobic digestion of waste activated sludge: Influence of the solid retention time in the wastewater treatment process. Process Biochemistry 40. https://doi.org/10.1016/j.procbio.2004.06.036

Cafiero, L.M., Canditelli, M., Musmeci, F., Sagnotti, G., Tuffi, R., 2021. Assessment of disintegration of compostable bioplastic bags by management of electromechanical and static home composters. Sustainability (Switzerland) 13. https://doi.org/10.3390/su13010263

Calabro', P. S., Folino, A., Fazzino, F., Komilis, D., 2020. Preliminary evaluation of the anaerobic biodegradability of three biobased materials used for the production of disposable plastics. Journal of Hazardous Materials 390. https://doi.org/10.1016/j.jhazmat.2019.121653

Calabro', P.S., Folino, A., Fazzino, F., Komilis, D., 2020. Preliminary evaluation of the anaerobic biodegradability of three biobased materials used for the production of disposable plastics. Journal of Hazardous Materials 390. https://doi.org/10.1016/j.jhazmat.2019.121653

Cazaudehore, G., Monlau, F., Gassie, C., Lallement, A., Guyoneaud, R., 2021. Methane production and active microbial communities during anaerobic digestion of three commercial biodegradable coffee capsules under mesophilic and thermophilic conditions. Science of The Total Environment 784. https://doi.org/10.1016/j.scitotenv.2021.146972

Chamas, A., Moon, H., Zheng, J., Qiu, Y., Tabassum, T., Jang, J.H., Abu-Omar, M., Scott, S.L., Suh, S., 2020. Degradation Rates of Plastics in the Environment. ACS Sustainable Chemistry and Engineering 8. https://doi.org/10.1021/acssuschemeng.9b06635

Chinaglia, S., Tosin, M., Degli-Innocenti, F., 2018. Biodegradation rate of biodegradable plastics at molecular level. Polymer Degradation and Stability 147. https://doi.org/10.1016/j.polymdegradstab.2017.12.011

Cucina, M., de Nisi, P., Tambone, F., Adani, F., 2021a. The role of waste management in reducing bioplastics’ leakage into the environment: A review. Bioresource Technology. https://doi.org/10.1016/j.biortech.2021.125459

Cucina, M., de Nisi, P., Trombino, L., Tambone, F., Adani, F., 2021b. Degradation of bioplastics in organic waste by mesophilic anaerobic digestion, composting and soil incubation. Waste Management 134. https://doi.org/10.1016/j.wasman.2021.08.016

Cucina, M., Pezzolla, D., Tacconi, C., Gigliotti, G., 2021c. Anaerobic co-digestion of a lignocellulosic residue with different organic wastes: Relationship between biomethane yield, soluble organic matter and process stability. Biomass and Bioenergy 153. https://doi.org/10.1016/j.biombioe.2021.106209

de Matos Costa, A.R., Crocitti, A., de Carvalho, L.H., Carroccio, S.C., Cerruti, P., Santagata, G., 2020. Properties of biodegradable films based on poly(Butylene succinate) (pbs) and poly(butylene adipate-co-terephthalate) (pbat) blends. Polymers 12. https://doi.org/10.3390/polym12102317

Decreto Legislativo 29 Aprile 2010, n. 75, 2010. Riordino e revisione della disciplina in materia di fertilizzanti, a norma dell’articolo 13 della Legge 7 Luglio 2009 n. 88. Gazzetta Ufficiale n. 121 - Supplemento Ordinario n.106, Roma.

Dolci, G., Venturelli, V., Catenacci, A., Ciapponi, R., Malpei, F., Turri, S.E.R., Grosso, M., 2022. Evaluation of the anaerobic degradation of food waste collection bags made of paper or bioplastic. Journal of environmental management, 305, 114331. https://doi.org/10.1016/j.jenvman.2021.114331

Edwards, J., Othman, M., Crossin, E., Burn, S., 2018. Life cycle assessment to compare the environmental impact of seven contemporary food waste management systems. Bioresource Technology 248. https://doi.org/10.1016/j.biortech.2017.06.070

Elfehri Borchani, K., Carrot, C., Jaziri, M., 2015. Biocomposites of Alfa fibers dispersed in the Mater-Bi® type bioplastic: Morphology, mechanical and thermal properties. Composites Part A: Applied Science and Manufacturing 78. https://doi.org/10.1016/j.compositesa.2015.08.023

Emadian, S.M., Onay, T.T., Demirel, B., 2017. Biodegradation of bioplastics in natural environments. Waste Management. https://doi.org/10.1016/j.wasman.2016.10.006

European bioplastic, 2020. Market update 2020: Bioplastics continue to become mainstream as the global bioplastics market is set to grow by 36 percent over the next 5 years [WWW Document]. https://www.european-bioplastics.org/market-update-2020-bioplastics-continue-to-become-mainstream-as-the-global-bioplastics-market-is-set-to-grow-by-36-percent-over-the-next-5-years/.

European Commission, 2018a. Plastics in a circular economy [WWW Document]. https://ec.europa.eu/info/research-and-innovation/research-area/environment/circular-economy/plastics-circular-economy_en.

European Commission, 2018b. Directive (EU) 2018/851 of the European Parliament and of the Council of 30 May 2018 amending Directive 2008/98/EC on waste (Text with EEA relevance). Official Journal of the European Union.

European Commission, 2015. Closing the Loop - An EU action plan for the Circular Economy - (ANNEX 1). Communication from the Commission to the European Parliament, the Council, the Europena Economic and Social Committee and the Committee of the Regions.

Geueke, B., Groh, K., Muncke, J., 2018. Food packaging in the circular economy: Overview of chemical safety aspects for commonly used materials. Journal of Cleaner Production. https://doi.org/10.1016/j.jclepro.2018.05.005

Gil-Castell, O., Andres-Puche, R., Dominguez, E., Verdejo, E., Monreal, L., Ribes-Greus, A., 2020. Influence of substrate and temperature on the biodegradation of polyester-based materials: Polylactide and poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) as model cases. Polymer Degradation and Stability 180. https://doi.org/10.1016/j.polymdegradstab.2020.109288

Giordano, A., di Capua, F., Esposito, G., Pirozzi, F., 2019. Long-term biogas desulfurization under different microaerobic conditions in full-scale thermophilic digesters co-digesting high-solid sewage sludge. International Biodeterioration and Biodegradation 142. https://doi.org/10.1016/j.ibiod.2019.05.017

Gunaseelan, V.N., 1997. Anaerobic digestion of biomass for methane production: A review. Biomass and Bioenergy 13. https://doi.org/10.1016/S0961-9534(97)00020-2

Hamad, K., Kaseem, M., Yang, H.W., Deri, F., Ko, Y.G., 2015. Properties and medical applications of polylactic acid: A review. Express Polymer Letters 9. https://doi.org/10.3144/expresspolymlett.2015.42

ISO, 2018. 14853: Plastics - Determination of the ultimate anaerobic biodegradation of plastic materials in an aqueous system - Method by measurement of biogas production.

ISO 20200, 2016. ISO 20200. Determination of the degree of disintegration of plastic materials under simulated composting conditions in a laboratory-scale test.

ISPRA, 2020. Rapporto rifiuti urbani (edizione 2020). ISPRA - Area Comunicazione.

Lin, L., Xu, F., Ge, X., Li, Y., 2018. Improving the sustainability of organic waste management practices in the food-energy-water nexus: A comparative review of anaerobic digestion and composting. Renewable and Sustainable Energy Reviews. https://doi.org/10.1016/j.rser.2018.03.025

Lu, J., Xu, S., 2021. Post-treatment of food waste digestate towards land application: a review. Journal of Cleaner Production, 303, 127033. https://doi.org/10.1016/j.jclepro.2021.127033

Mao, C., Feng, Y., Wang, X., Ren, G., 2015. Review on research achievements of biogas from anaerobic digestion. Renewable and Sustainable Energy Reviews. https://doi.org/10.1016/j.rser.2015.02.032

Marek, A.A., Verney, V., 2016. Photochemical reactivity of PLA at the vicinity of glass transition temperature. the photo-rheology method. European Polymer Journal 81. https://doi.org/10.1016/j.eurpolymj.2016.06.016

Matthews, C., Moran, F., Jaiswal, A.K., 2021. A review on European Union's strategy for plastics in a circular economy and its impact on food safety. Journal of Cleaner Production. https://doi.org/10.1016/j.jclepro.2020.125263

Meereboer, K.W., Misra, M., Mohanty, A. K., 2020. Review of recent advances in the biodegradability of polyhydroxyalkanoate (PHA) bioplastics and their composites. Green Chemistry 22(17), 5519-5558. 10.1039/D0GC01647K

Pagga, U., 1998. Biodegradability and compostability of polymeric materials in the context of the European packaging regulation. Polymer Degradation and Stability 59. https://doi.org/10.1016/s0141-3910(97)00192-4

Pigoli, A., Zilio, M., Tambone, F., Mazzini, S., Schepis, M., Meers, E., Schoumans, O., Giordano, A., Adani, F., 2021. Thermophilic anaerobic digestion as suitable bioprocess producing organic and chemical renewable fertilizers: A full-scale approach. Waste Management 124. https://doi.org/10.1016/j.wasman.2021.02.028

PlasticsEurope, 2020. Plastics - the Facts 2020. An analysis of European plastics production, demand and waste data.

Rapisarda, la Mantia, Ceraulo, Mistretta, Giuffrè, Pellegrino, Valenti, Rizzarelli, 2019. Photo-Oxidative and Soil Burial Degradation of Irrigation Tubes Based on Biodegradable Polymer Blends. Polymers 11. https://doi.org/10.3390/polym11091489

Ruggero, F., Carretti, E., Gori, R., Lotti, T., Lubello, C., 2020. Monitoring of degradation of starch-based biopolymer film under different composting conditions, using TGA, FTIR and SEM analysis. Chemosphere 246. https://doi.org/10.1016/j.chemosphere.2019.125770

Ruggero, F., Gori, R., Lubello, C., 2019. Methodologies to assess biodegradation of bioplastics during aerobic composting and anaerobic digestion: A review. Waste Management and Research. https://doi.org/10.1177/0734242X19854127

Sarkar, B., Debnath, A., Chiu, A. S., Ahmed, W., 2022a. Circular economy-driven two-stage supply chain management for nullifying waste. Journal of Cleaner Production, 130513. https://doi.org/10.1016/j.jclepro.2022.130513

Sarkar, B., Ullah, M., Sarkar, M., 2022b. Environmental and economic sustainability through innovative green products by remanufacturing. Journal of Cleaner Production, 332, 129813. https://doi.org/10.1016/j.jclepro.2021.129813

Sarkar, M., Seo, Y.W., 2021. Renewable energy supply chain management with flexibility and automation in a production system. Journal of Cleaner Production, 324, 129149. https://doi.org/10.1016/j.jclepro.2021.129149

Scaglia, B., Tambone, F., Corno, L., Orzi, V., Lazzarini, Y., Garuti, G., Adani, F., 2018. Potential agronomic and environmental properties of thermophilic anaerobically digested municipal sewage sludge measured by an unsupervised and a supervised chemometric approach. Science of the Total Environment 637–638. https://doi.org/10.1016/j.scitotenv.2018.04.426

Schievano, A., Pognani, M., D’Imporzano, G., Adani, F., 2008. Predicting anaerobic biogasification potential of ingestates and digestates of a full-scale biogas plant using chemical and biological parameters. Bioresource Technology 99. https://doi.org/10.1016/j.biortech.2008.03.030

Shrestha, A., van-Eerten Jansen, M. C., Acharya, B., 2020. Biodegradation of bioplastic using anaerobic digestion at retention time as per industrial biogas plant and international norms. Sustainability, 12(10), 4231. https://doi.org/10.3390/su12104231

US Department of Agricolture - US Composting Council, 2002. Test Methods for the Examination of Composting and Compost (TMECC).

Wainaina, S., Awasthi, M.K., Sarsaiya, S., Chen, H., Singh, E., Kumar, A., Ravindran, B., Awasthi, S.K., Liu, T., Duan, Y., Kumar, S., Zhang, Z., Taherzadeh, M.J., 2020. Resource recovery and circular economy from organic solid waste using aerobic and anaerobic digestion technologies. Bioresource Technology. https://doi.org/10.1016/j.biortech.2020.122778

Yagi, H., Ninomiya, F., Funabashi, M., Kunioka, M., 2014. Mesophilic anaerobic biodegradation test and analysis of eubacteria and archaea involved in anaerobic biodegradation of four specified biodegradable polyesters. Polymer Degradation and Stability 110. https://doi.org/10.1016/j.polymdegradstab.2014.08.031

Yagi, H., Ninomiya, F., Funabashi, M., Kunioka, M., 2013. Thermophilic anaerobic biodegradation test and analysis of eubacteria involved in anaerobic biodegradation of four specified biodegradable polyesters. Polymer Degradation and Stability 98. https://doi.org/10.1016/j.polymdegradstab.2013.03.010

Zhang, W., Heaven, S., Banks, C.J., 2018. Degradation of some EN13432 compliant plastics in simulated mesophilic anaerobic digestion of food waste. Polymer Degradation and Stability 147. https://doi.org/10.1016/j.polymdegradstab.2017.11.005

Zhao, X., Cornish, K., Vodovotz, Y., 2020. Narrowing the Gap for Bioplastic Use in Food Packaging: An Update. Environmental science & technology. https://doi.org/10.1021/acs.est.9b03755