Article

Pilot-Scale Anaerobic Co-Digestion of Food Waste and Polylactic Acid

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Abstract: Bioplastics are frequently utilized in daily life, particularly for food packaging and carrier bags. They can be delivered to biogas plants through a separate collection of the organic fraction of municipal waste (OFMSW). The increased demand for and use of bioplastics aimed at mitigating plastic pollution raises significant questions concerning their life cycle and compatibility with waste management units. Anaerobic digestion (AD) in OFMSW is a valuable resource for biogas production. In this work, the valorization of poly-L-lactic acid (PLLA) composed of food waste within the Biowaste to Bioplastic (B2B) Project framework was studied in laboratory and pilot-scale anaerobic liquid conditions. Taking into account that the addition of PLLA to biowaste can increase biogas production, we performed laboratory-scale anaerobic tests on food waste enriched with different molecular-weight PLLAs produced from food waste or commercial PLLA at a mesophilic temperature of 37 °C. PLLA with the highest molecular weight was subjected to AD on the pilot scale to further validate our findings. The addition of PLLA increased biogas production and had no apparent negative impact on the operation of the reactors used in the laboratory or on the pilot scale. Biogas production was higher when using PLLA with the lowest molecular weight. In the pilot-scale experiments, co-digestion of FW with PLLA increased biogas production by 1.1 times. When PLLA was added to the feed, biomethane was 8% higher, while volatile solids (VS) and total chemical oxygen demand (TCOD) removal were almost the same. Importantly, no effect was observed in the operation of the digesters.

Keywords: biogas; anaerobic digestion; food waste; PLLA

1. Introduction

Plastic pollution has overtaken global warming as the biggest environmental issue in the world, with the annual plastic environmental burden accounting for more than 8 Mt, most of which ends up in the oceans. With the European Union’s Single Use Plastics Directive (2019/904) banning single-use plastics entering into force in the summer of 2021, states and organizations are firmly fighting plastic pollution. Plastics made from petroleum could be replaced by bioplastics. In Greece, non-biodegradable lightweight plastic carrier bags have been prohibited since 1 January 2018, with the imposition of an environmental tax. Biodegradable and compostable bags made from PLLA are excluded from this environmental tax. Therefore, compostable bags are increasingly being used in Greece. Additionally, the need for bioplastic manufacturing is rising due to the expanding use of bioplastics in food packaging applications [1].

During the last few decades, bioplastics have emerged as sustainable and ecologically acceptable alternatives to plastics made from fossil fuels. Bioplastic manufacturing has increased globally in recent years, especially in the EU, where single-use plastics were outlawed by new legislation in 2019 [2]. In contrast to traditional plastics, which will take
a few hundred or a few thousand years to disintegrate in natural environments, causing multifarious problems in the process, bioplastics are expected to degrade within a matter of years [3,4]. The characteristics of the materials themselves and the environment in which they are disposed of, such as pH, temperature, moisture, and oxygen content, have a significant impact on how quickly bioplastics degrade. There are numerous alternatives to post-use bioplastic disposal, including anaerobic digestion, composting, and recycling [5]. The biodegradability and compostability of bioplastics are certified using the best-defined conditions, which generally diverge from actual conditions in industrial biowaste treatment facilities [6].

Currently, the organic waste management system for the separate collection and recycling of the OFMSW is governed by the Waste Framework Directive (2008/98/EC). It mainly consists of biological treatments like composting or integrated anaerobic-composting digestion. However, AD and composting facilities will soon need to deal with the growth and adverse effects of novel polymeric organic fraction materials within the OFMSW. In this context, one of the main concerns is the integration of biodegradable bioplastics in the biowaste management chain and, consequently, in AD and composting facilities [4,5]. Due to the increasing need to process large quantities of bioplastics in facilities not intended or designed for processing bioplastics, the degradation of bioplastics in OFMSW treatment facilities could become a growing problem. Inefficient processing of bioplastics could result in the contamination of digestate and/or compost with significant amounts of non- or partially degraded bioplastics. Currently, during the initial stage of waste treatment (before the biological processes), plastics, including bioplastics, are mechanically separated and frequently disposed of in landfills.

AD is one of the most efficient processes for treating various kinds of raw biomasses and converting them into clean energy. The main goal of such processes is to efficiently convert waste into renewable biofuel or biogas. By converting bioplastics into biomethane, AD could increase the circularity of bioplastics by passing bioplastic leaking into the environment through natural deterioration. A practical method for recovering organic energy is the AD of organic waste, which has been thoroughly studied and approved for commercial use [7]. AD is one of the most popular ways of handling OFMSW because contamination risks are minimized and biogas, a renewable energy source, is produced in a controlled process that benefits the environment. More than 20,000 biogas plants have been built worldwide [8,9]. On the other hand, it is worth adding that digestate sludge from an agricultural biogas plant can also be reused to produce biodegradable materials. Ekielski et al. (2021) [10] showed that adding digestate to thermoplastic starch enabled the production of homogeneous biocomposites with a structure resembling a sheet or thick film. Furthermore, thermoplastic starch has emerged as a readily biodegradable and cheap biomaterial that can replace traditional plastics in applications such as food service and packaging [11]. According to Cazaudehore et al. (2022) [11], thermoplastic starch can reach near-complete mineralization in a relatively short period under mesophilic and thermophilic conditions. These materials could be treated in biogas plants without altering the hydrolytic retention time. Moreover, Russo et al. (2009) [12] investigated the degradability of thermoplastic starch under anaerobic conditions to simulate the most common disposal environment for household wastes. The extent of solubilization data has shown that starch is almost entirely degraded.

According to Abraham et al. 2021 [5], the anaerobic degradability of bioplastics is primarily influenced by their composition (e.g., polyhydroxyalkanoates degrade more effectively than PLA- and starch-based bioplastics), their physicochemical features (e.g., the degradation rate of PLLA is significantly slower than PDLLA), and the key operating parameters (such as temperature). At the same time, bioplastic degradation is also heavily influenced by microbial diversity and the environmental and operational conditions involved. It must be stressed that not all types of bioplastics can be efficiently managed by AD.
A common ingredient in the food, pharmaceutical, and chemical industries, lactic acid (2-hydroxypropanoic acid), can be polymerized to create the biodegradable and compostable polymer polyactic acid (PLA). This has the potential to replace polymers made from petroleum. The primary application for PLA is the production of both durable and disposable items. PLA is created through the microbial fermentation of carbohydrates into lactic acid and its subsequent polymerization.

The anaerobic biodegradability of bioplastics and the potential production of biomethane from their degradation presents some serious research gaps. One crucial gap is probably the fact that no data are currently available from pilot-scale studies. Equally important, laboratory experiments mostly present the properties of bioplastics produced from commercially available monomers or bioplastics. In this context, the aim and innovation of the work presented in this manuscript were to evaluate the biodegradability of PLLA produced from FW on a pilot scale and to examine for the first time the compatibility of such biodegradable materials with the current industrial management scheme for biowaste. The increasing use of bioplastics and their disposal within the organic fraction of municipal solid waste represent a critical component for the future of waste management. At the same time, the production of PLLAs with low molecular weight can be validated to produce materials useful in composting and absorbing harmful and persistent contaminants. Their biodegradation was studied during AD to extract useful information on this aspect.

The goal of the present paper was to examine the anaerobic decomposition of PLLA produced in the B2B Project. In the course of the B2B project, PLLA was synthesized from food waste with the aim of optimizing bioplastic production in the framework of the circular economy. PLLA produced by these means could substitute conventional, petrochemical-based plastics and find application in typical full-scale anaerobic units. The purpose of the research presented herein was to evaluate how food-waste-derived PLLA with different molecular characteristics would affect anaerobic digestion and biogas production. To evaluate the synergistic effect of co-digestion of PLLA with FW, mono- and co-digestion experiments were performed in lab-scale and pilot-scale bioreactors.

2. Materials and Methods

2.1. Raw Materials, Substrate and Inoculum

Liquid pig manure (LPM) was collected from a local pig farm raising 70 sows (Voutes, Crete, Greece). Food waste (FW) was gathered from the student restaurant at the Hellenic Mediterranean University in Heraklion, Crete, and used in the current investigation. The FW composition was 30% vegetables and salads, 10% bread and bakery, and 60% cooked meals (on a wet weight basis). A mechanical mixer was used to homogenize FW (approximately 4.0 mm). The anaerobic digester of the Heraklion sewage treatment facility (STP) was used to harvest inoculum for this experiment (population: about 200,000). The obtained materials were refrigerated at 4 ± 2 °C before the experimental or pilot trials. Three different batches of PLLA were synthesized through direct azeotropic polycondensation of L-lactic acid produced using FW gathered from the Municipality of Heraklion in the context of the B2B Project (PLLAl, PLLA2, and PLLA3). For the production of lactic acid, the approach suggested by Sakai and colleagues [13] (Sakai et al., 2003) was followed and optimized for the creation of a scalable synthetic process. All samples of PLLA were obtained from polymerization as a white powder with a particle size < 3 mm and were used without pretreatment. Table 1 summarizes the average raw FW composition and LPM. Table 2 provides a summary of the characteristics of synthetic and commercial PLLAs used in the current study. The three batches of synthetic PLLA (PLLAl, PLLA2, and PLLA3) and the commercial PLLA were used in biochemical methane potential (BMP) experiments. The highest molecular weight of PLLA (PLLAl) was used for experiments in pilot AD reactors.
Table 1. Composition of Food Waste (FW) and Liquid Pig Manure (LPM).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>LPM</th>
<th>FW</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>7.7 ± 0.1</td>
<td>4.5 ± 0.0</td>
</tr>
<tr>
<td>TS (g/kg)</td>
<td>9.4 ± 9.2</td>
<td>256.3 ± 2.0</td>
</tr>
<tr>
<td>VS (g/kg)</td>
<td>5.7 ± 5.3</td>
<td>242.7 ± 5.0</td>
</tr>
<tr>
<td>TCOD (g/L)</td>
<td>11.8 ± 5.2</td>
<td>151.3 ± 10.0</td>
</tr>
<tr>
<td>TN (g/L)</td>
<td>0.2 ± 0.1</td>
<td>2.5 ± 0.0</td>
</tr>
</tbody>
</table>

Table 2. Characteristics of produced and commercial PLLAs.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Commercial PLLA&lt;sub&gt;c&lt;/sub&gt;</th>
<th>PLLA&lt;sub&gt;1&lt;/sub&gt;</th>
<th>PLLA&lt;sub&gt;2&lt;/sub&gt;</th>
<th>PLLA&lt;sub&gt;3&lt;/sub&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>Weight Average Molecular Weight (M&lt;sub&gt;w&lt;/sub&gt;)</td>
<td>153,373</td>
<td>149,452</td>
<td>27,987</td>
<td>55,663</td>
</tr>
<tr>
<td>Number Average Molecular Weight (M&lt;sub&gt;n&lt;/sub&gt;)</td>
<td>76,640</td>
<td>81,146</td>
<td>13,552</td>
<td>26,332</td>
</tr>
<tr>
<td>Polydispersity D</td>
<td>2.00</td>
<td>1.84</td>
<td>1.84</td>
<td>1.55</td>
</tr>
</tbody>
</table>

2.2. Biochemical Methane Potential Experiments

At mesophilic conditions (37 °C), batch studies were conducted in triplicates to ascertain the impact of the molecular weight of the different PLLAs on the methane potential of fresh food wastes. Commercial PLLA was also used for comparison. Different substrates (FW, FW and PLLA<sub>c</sub>, FW and PLLA<sub>1</sub>, FW and PLLA<sub>2</sub>, and FW and PLLA<sub>3</sub>) were assessed to identify which were the most successful in achieving the highest methane yield. The experiments were performed using a technique based on Angelidaki and Sanders (2004) [14]. A total of 0.5 g of each PLLA sample and 2 g of FW were added to batch reactors. One inoculum-to-substrate ratio (ISR) was examined at 1:1 (VS). Reactors made out of 120 mL serum bottles were used for this research. Immediately after adding the inoculum and substrates, the reactors were flushed with a gas combination of 70% nitrogen and 30% carbon dioxide to create anaerobic conditions. Rubber septa and aluminum crimph closures were then used to seal the serum vials. For more than four months (about 125 days), the output and content of biogas were monitored in each bottle at regular time intervals. The results are presented as the amount of methane produced from the breakdown of the organic substrate in milliliters per kilogram of volatile solids loaded (mL/g VS) and the degradation rate.

2.3. Experimental Procedure and Pilot-Scale Basic Anaerobic Digester

To investigate the biodegradability of PLLA in anaerobic digestion, two different types of influent feedstock were used: (A) a mixture of FW and LPM and (B) a mixture of FW, LPM, and PLLA. About 180 L of a mixture of 20% v/v FW, 50% v/v LPM, and 30% v/v inoculum was added to the digester, which was then sealed. After 50 days of AD, the digester was opened to extract the digestate. The second type of influent feedstock was a mixture of 20% v/v FW, 50% v/v LPM, 30% v/v inoculum, and 1 kg PLLA. Prior to analysis, representative samples of digestate were gathered and stored at 4 °C. A simple anaerobic digester was utilized to test the biogas production by co-digestion of food waste with PLLA. The anaerobic digester had a double wall and was made of stainless steel. Agitation was achieved by a motor drive unit mounted on top of the reactor. The reactor had a 220 L capacity and was operated in a mesophilic environment (37 ± 2 °C). The digester had a top-mounted, three-bladed mixer with a nominal shaft speed of 100 rpm (Figure 1). The motor was equipped with an on/off timer to maintain 2 h of daily intermittent mixing. The amount of biogas generated was measured each day using a Ritter TG5 model 5 gas meter in the form of a drum. The digesters were not run in parallel, so the FW and LPM originated at different times. The principal operational and output variables for the pilot anaerobic digestion process are summarized in Table 3.
Table 3. Principal operational and output variables for the pilot anaerobic digestion process.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit</th>
<th>A</th>
<th>B</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Operational parameters</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Operating volume</td>
<td>L</td>
<td>180</td>
<td>180</td>
</tr>
<tr>
<td>Retention time</td>
<td>days</td>
<td>50</td>
<td>50</td>
</tr>
<tr>
<td><strong>Production Parameters</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total Biogas</td>
<td>L</td>
<td>1.710</td>
<td>1.889</td>
</tr>
<tr>
<td>Total Biogas L/kg VS</td>
<td></td>
<td>185.5</td>
<td>197.6</td>
</tr>
<tr>
<td>CH₄ %</td>
<td></td>
<td>64 ± 4.7</td>
<td>65 ± 3.4</td>
</tr>
<tr>
<td>Total Biomethane</td>
<td>L</td>
<td>1.094</td>
<td>1.228</td>
</tr>
<tr>
<td>Total Biomethane L/kg VS</td>
<td></td>
<td>118.7</td>
<td>128.5</td>
</tr>
<tr>
<td><strong>Characteristics of Mixture and digestate</strong></td>
<td></td>
<td>Mixture</td>
<td>Digestate</td>
</tr>
<tr>
<td>VS g/L</td>
<td></td>
<td>51.2 ± 3.2</td>
<td>19.2 ± 1.2</td>
</tr>
<tr>
<td>TCOD g/L</td>
<td></td>
<td>38.7 ± 3.1</td>
<td>23.7 ± 1.5</td>
</tr>
</tbody>
</table>

Figure 1. Pilot-scale anaerobic digester with 220 L capacity.

2.4. Analytical Methods

A pH meter was used to measure the pH (Crison, GLP 21, Barcelona, Spain). Total nitrogen (TN) was also measured using the Semi-Micro-Kjeldahl Method. Total chemical oxygen demand (TCOD) was measured using the Closed Reflux Colorimetric Method. Additionally, total solids (TS) and volatile (VS) solids were determined using standard methods [15]. A gas chromatography (GC) instrument was used to evaluate the composition of the biogas (Agilent 6890N GC System, Santa Clara, CA, USA). After sampling with gas-tight syringes, the needle was sealed with a butyl rubber stopper. Twenty microliters of gas samples were introduced to the GC to analyze methane and carbon dioxide.
A capillary column (GS Carbonplot, 30 m × 0.32 mm, 3 lm) and a thermal conductivity detector (TCD) were tandemly utilized. The detector port and the column were operated isothermally at 150 °C and 80 °C, respectively. Helium was used as the carrier gas with a 15 mL/min flow rate. All individual sample analyses were performed in triplicate. The study data and findings were statistically analyzed (average values, variance, and standard deviation) using Origin 9 (OriginLab, Northampton, MA, USA). PLLAs were characterized with nuclear magnetic resonance (NMR) and infrared spectroscopy (FTIR) and analyzed with gel permeation chromatography (GPC). GPC was performed on a modular chromatography system (Shimadzu, Kyoto, Japan). This system comprised a CBM-20A system controller (Shimadzu, Kyoto, Japan), LC-20AD pump (Shimadzu, Kyoto, Japan), SIL-20A automatic injector (Shimadzu, Kyoto, Japan), RID-20A differential refractive index detector (Shimadzu, Kyoto, Japan) and, CTO20AC oven (Shimadzu, Kyoto, Japan). SFC was performed on a modular chromatography system (Shimadzu, Kyoto, Japan). This system comprised a CBM-20A system controller (Shimadzu, Kyoto, Japan), LC-20AD pump (Shimadzu, Kyoto, Japan), SIL-20A automatic injector (Shimadzu, Kyoto, Japan), RID-20A differential refractive index detector (Shimadzu, Kyoto, Japan) and, CTO20AC oven (Shimadzu, Kyoto, Japan). The static phase was a 10.0 μm, 50 × 7.5 mm, bead-size guard column (Agilent Technologies, Santa Clara, CA, USA), followed by a 5 μm bead size, 300 × 7.5 mm, PLgel Mixed-D column (Agilent Technologies, Santa Clara, CA, USA). Analysis was performed at a constant temperature of 40 °C using HPLC grade THF as an eluent. Calibration was performed using commercial narrow molecular weight distribution polystyrene standards with molecular weights ranging from 580 to 299,400 g·mol⁻¹ (Polymer Laboratories, Long Beach, CA, USA).

3. Results and Discussion

3.1. BMP Experiment

The amount of biogas produced during a BMP laboratory experiment was determined to decompose PLLAs with different origins and molecular weights (Table 2). The highest biogas production was measured using FW and the lowest molecular weight, PLLA₂ as substrates. Taking into account the entire volume of biogas measured during the 125-day incubation, FW produced 503 mL, while FW and PLLA₁, FW and PLLA₂, FW and PLLA₃, and PLLA: produced 570 mL/g VS, 577 mL/g VS, 620 mL/g VS, 577 mL/g VS, and 103 mL/g VS, respectively (Figure 2). The methane content of the produced biogas fluctuated between 65.0% and 65.7%. The maximum methane content, 65.7%, was observed in biogas produced using FW and PLLA₂ as substrates. This can most probably be attributed to the different physicochemical characteristics of the PLLAs used in this study. According to Hamad et al. (2015) [16], PLA blends are resistant to attack by microorganisms in mesophilic conditions. They must be exposed to hydrolysis at temperatures above 58 °C in order to lose some of their molecular weight and begin to degrade. Therefore, the fastest biodegradation of PLLA₂ most probably originates from its reduced molecular weight. Upon completion of the mesophilic anaerobic digestion stage, the biogas potentials determined for the PLLAs used in the current study were comparable to those described in the literature for other biomasses frequently utilized as feed in anaerobic digestion. For instance, Gunaseelan (1997) [17] discovered that the hand-sorted organic fraction of municipal solid wastes (HS-OFMSW) had a biomethane potential of 190 LCH₄/kg VS, while the biomethane potentials of OFMSW, food wastes, and cattle manure were 180, 223, and 188 LCH₄/kg VS, respectively [18].
Figure 2. Cumulative biogas production in the anaerobic co-digestion of FW and PLLAs of different molecular weights.

### 3.2. Pilot Experiments

To simulate full-scale anaerobic degradation, biomethane extraction from PLLA was assessed on the 180 L pilot scale. All operational parameters and outcomes of the pilot-scale study are described in Table 3. The effects of the composition of the mixtures under evaluation on the co-digestion process and the most effective mixing ratio for batch reactor operation were investigated in two experiments using the same reactor. Figure 3 presents total biogas production (L/kg VS) for mixtures of 20% v/v FW, 50% LPM, and 30% inoculum (mixture A) and a mixture of 20% v/v FW, 50% v/v LPM, 30% v/v inoculum, and 1 kg PLLA (mixture B). The addition of PLLA increased the production of biogas and had no apparent negative impact on the reactor's operation. More specifically, co-digestion with PLLA increased biogas generation by 1.1 times, a 6.5% improvement in biogas compared to the control (Figure 3). In detail, the AD process in mesophilic conditions generated 1.710 L (185.5 L/kg VS) and 1.889 L (197.6 L/kg VS) of biogas, with average biomethane contents of 64% v/v and 65% for mixtures A and B, respectively. The mixture of FW-LPM and PLLA1 generated 1.228 L (128.5 L/kg VS) of cumulative biomethane. As summarized in Table 3, the anaerobic co-digestion of FW and LPM investigated in the research unit significantly enhanced the biomethane generation in the presence of PLLA at a concentration of as little as approximately 0.5%. The life cycle of PLLA—bioplastics may become more sustainable due to the rise in biomethane (from 1.094 to 1.228 L) (Table 3). This result is comparable to the best that has previously been reported in the literature. Importantly, as the degradation results from the laboratory trial and the pilot-scale experiment were comparable, we can safely assume that the full-scale experiments could be effectively simulated. The valuable byproduct of bioplastic decomposition, biomethane, might be used to offset the higher expense of collecting and transporting bioplastics separately. According to Zhang et al. (2018) [6], film and pellet samples had noticeably different rates of biogas production in mesophilic conditions, which were 97 and 17 L/kg VS, respectively. Coffee cups produced 127 L/kg VS of biogas in mesophilic conditions, whereas film and particle samples degraded only about half as quickly [19]. Compared to pellet samples, Wang et al. (2018) [20] showed a superior biogas yield (30% more), which was consistent with the results for PLA samples reported by Zhang et al. (2018) [6]. According to Nachod et al. (2021) [21], the cumulative CH4 production for the PLA-FW treatments was 730% higher than the FW treatment. Moreover, Shrestha et al. (2020) [22] and...
Luo et al. (2019) [23] mentioned that the increased production of biogas from the addition of bioplastic boosted anaerobic biodegradability. Kang et al. (2022) [24] showed that the synergetic effect of FW addition on methane production yield of co-digestion with PLA reached 8.5–26.6%.

Figure 3. Total Biogas production during the experimental period for different substrate, mixture A (20% v/v FW, 50% LPM, and 30% inoculum) and mixture B (20% v/v FW, 50% v/v LPM, 30% v/v inoculum, and 1 kg PLLA).

VS concentrations of digestates were evaluated in order to assess the biodegradation rates of PLLA. Figure 4 shows the variation of VS during both experimental periods (without and with the addition of PLLA) in the digester’s input and output. When the reactor was running with mixture A, the average concentration of VS$_{in}$ was found to be 51.2 ± 3.2 g/L, while during the reactor operation with mixture B, it was 53.1 ± 2.4 g/L. As a result, the VS concentration was nearly the same. For the scenario with mixtures A and B, the removal efficiency in VS concentration was 62.5% from 51.2 to 19.2 g/L and 63.8% from 53.1 to 24.0 g/L. (Figure 4). Therefore, VS removal was almost the same. Kang et al. (2022) [24] showed that the biodegradation rates (on a VS basis) increased by 1.8–4.3 times in the PLA-FW co-digestion tests. In a recent study, co-digestion was found to improve the reduction of VS. According to a study by Benn and Zitomer (2018) [25], the VS reduction in bioplastics was approximately 75% during mono-digestion but increased to 81% when bioplastics were digested along with other organic waste.
Figure 4. VS removal and concentrations in the influent (in) and effluent (out) stages for mixtures A and B. Error bars indicate standard deviation.

PLLA incorporation in AD resulted in a practically unchanged total COD reduction (Figure 5). In the digester operated with mixtures A and B, the average removal of TCOD was 38.8% and 36.2%, respectively. Compared to the initial mixture, the digestate’s characterization revealed the removal of volatile solids and a total chemical oxygen demand of 63.8% and 36.2%, respectively. These outcomes resulted from hydrolyzing organic matter and converting soluble organic matter to biomethane. Furthermore, this degree of removal matches previously reported values in the 30–40% range found as COD reductions during anaerobic degradation of FW [26].

Figure 5. TCOD removal and concentrations in the influent (in) and effluent (out) stages for mixtures A and B. Error bars indicate standard deviation.

According to Bátori et al. (2018) [27], not all biopolymers can be used in anaerobic digestion. In the present study, the PLLA produced from FW was combined with food residues as a substrate. Not only did this synergy enhance biogas production, but it also
proved that PLLA does not interfere with the reactor’s operation and can be degraded under mesophilic conditions. This outcome demonstrates that renewable energy generation could be achieved using existing infrastructure and minimal process modification. Biogas production was not shown to differ significantly among the materials tested. However, the PLLA with the lowest molecular weight appeared to biodegrade more efficiently.

Another problem that needs to be addressed in the future is the presence of bioplastic residues in the digestate. Understanding the fate of bioplastics in the soil is crucial because digestate leftovers from bioplastics can escape into the ground. Starch-based and PLA-based bioplastics were reported to degrade in soil between 1.5 and 4.5 years [28,29]. These timeframes are significantly less than the times needed for the decomposition of petroleum-derived plastics in soil (1000–5000 years), even though they are longer than those reported for the degradation of bioplastics during anaerobic digestion and composting [3]. According to a recent study [4], the amount of bioplastic broken down during waste management determines how much leaks into the environment. The remaining untreated solubilized PLLA would be landfilled. Kolstad et al. [30] reported that microorganisms living in anaerobic conditions are unable to degrade the high molecular weight of PLA. Therefore, it is assumed that the residual untreated PLLA from the anaerobic digestion process will not continue to emit biogas when landfilled and will offset emissions via carbon sequestration.

Research shows that AD can be a sustainable method of managing bioplastics during biomethane production. Increasing the amount of biomethane produced from bioplastics contributes to economic sustainability since it increases the amount of renewable energy produced overall. Composting after anaerobic digestion should also be evaluated, as early research indicates that residual biopolymers could be transferred to the final product. The policy should be revised with caution considering the fates of these materials in the environment depending on the results of further investigations on the degradation rates under environmentally relevant conditions. The insights gained will contribute to implementing optimized measures and waste management policies. Moreover, the potential application of AD processes for processing bioplastic substrates could be seen as a value-added product to assist in the transition from petroleum-based plastics to bioplastics such as PLLA from FW.

4. Conclusions

Because the use of bio-based products, including PLLA, has increased during the last few decades, biopolymer waste has also increased in municipal waste streams. Therefore, studies evaluating the fate, effect, and means to degrade bioplastics become increasingly important. Since anaerobic digestion is a method of choice for managing and validating organic waste, this method was used to evaluate the fate of PLLA produced from food waste (in the framework of the B2B project) under lab- and pilot-scale mesophilic conditions. This work highlights the viability of this approach and demonstrates that PLLA becomes a more effective substrate when its molecular weight decreases. BMP tests devoted to assessing the impact of the characteristics of PLLA on the yields and kinetics of biogas production were performed, demonstrating that the methanization rate of all bioplastics tested can be substantially enhanced with their addition to the feed. In agreement with laboratory experiments, pilot-scale experiments resulted in maximum biogas and biomethane production when PLLA was incorporated into the feed. Biomethane production was found to be 8% higher; notably, PLLA did not affect the operation of the digester. At the same time, VS and TCOD removal remained practically the same. More importantly, the fact that PLLA produced from food is compatible with the existing anaerobic digestion units regardless of its molecular weight is promising for further valorization of PLLA in the context of the circular economy. In order to ensure that this material has no unfavorable effects on the environment, it is important to precisely evaluate the presence of residual PLLA in both the liquid and solid effluent stages. Considering these, current experiments aim at fine-tuning the characteristics of PLLA produced from food waste and investigating their full effect on biogas generation and digestate fate.
Author Contributions: Methodology, A.M. and K.V.; Investigation, A.M.; Resources, K.V.; Writing—original draft, A.M.; writing—review and editing, A.M.; Supervision, C.T. and T.M. All authors have read and agreed to the published version of the manuscript.

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References

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