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# Anaerobic Co-Digestion of Bioplastics and Food Waste under Mesophilic and Thermophilic Conditions: Synergistic Effect and Biodegradation

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**Abstract:** To mitigate the various problems caused by using conventional plastics, bioplastic (BP) has emerged as a substitute for plastics. BP wastes after use are commonly treated using composting, causing many environmental problems. Anaerobic digestion (AD) has become prominent as an alternative method of producing renewable energy. The aim of this study was to estimate the methane production yield (MPY) of BPs (polylactic acid (PLA) and polyhydroxyalkanoate (PHA)) with mechanical pretreatment (particle size < 0.5 cm) and investigate the effect of co-digestion of BPs and food waste (FW). Batch experiments were conducted under mesophilic and thermophilic conditions at various mixing ratios (FW/PLA or PHA = 95:5 and 90:10 on a weight basis). During 20 d of digestion at temperatures of 37 and 55 °C, MPYs of PHA were 153.8–172.0 mL $CH_4$/g chemical oxygen demand (COD), but that of PLA was significantly low (<25.6 mL $CH_4$/g COD). Higher MPYs were attained at 55 °C than at 37 °C. The synergistic effects of FW addition on BP AD were observed at both temperatures, especially at 55 °C. By comparing theoretical (based on mono-digestion results) and actual (based on co-digestion results) MPYs, the synergistic effect of FW addition on MPY of co-digestion reached 8.5–26.6% and 12.7–25.5% for PLA- and PHA-fed tests, respectively. The biodegradation rates (on a volatile solids (VS) basis) of PLA and PHA were 6.0–13.7% and 49.1–52.3% and increased by 1.8–4.3 and 1.2–1.5 times in the PLA- and PHA-fed co-digestion tests, respectively. Co-digestion of FW might be a feasible treatment option for BPs combined with simple mechanical pretreatment.

**Keywords:** bioplastics; food waste; co-digestion; synergistic effect; temperature; biodegradation



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## 1. Introduction

Plastic is a useful material in today's society and has been widely used in various industrial fields, including packing and building [1]. Owing to its beneficial properties, in particular high physical and chemical resistance, light weight, and low cost for manufacturing, annual production has consistently increased to 368 million tons in 2019 [2]. Conventional plastics made from petroleum products are barely biodegradable and easily accumulate in nature, affecting the quality of human life and ecosystems [3,4]. When incineration and landfill are utilized for treating plastic wastes, a significant amount of greenhouse gases (GHGs) and toxic gases are emitted, which means these treatment methods are also not environmentally friendly [5,6].

Due to these problems associated with plastic management and environmental concern, the interest in bioplastic (BP) as a replacement for conventional plastics has been rising considerably. In general, BPs are produced from bio-based materials and should decompose by more than 90% during aerobic composting within six months, thereby helping mitigate the environmental concerns of plastics [7,8]. Among various types of biodegradable BPs, polylactic acid (PLA) and polyhydroxyalkanoate (PHA) may be considered representative substances. PLA made from lactic acid monomers produced from microbial fermentation

is the second most produced BP, accounting for 13.9% of global total BP production, and is expected to be the main contributor to the BP industries [5,9]. Compared to PLA, PHA synthesized by numerous bacteria within their cell cytoplasm is more easily degraded by microorganisms; under anaerobic conditions at 37 °C, polyhydroxybutyrate (PHB), a sort of PHA, decomposed by approximately 90% in 10 days, whereas PLA decomposed by only 7% even in 90 days [7,10,11].

As BP production and consumption have increased, the methods for managing and treating BP wastes have attracted great attention globally. Although conventional plastic treatment methods are still mainly utilized, many researchers have attempted to apply biological treatment methods for BPs. Composting, a representative aerobic treatment of organic wastes, has been tested, but it appears difficult to fully degrade BP waste, mainly PLA, in industrial composting facilities [9,12]. In another case, it was observed that various types of composting plastics were barely degraded for 31 weeks under home composting conditions [13]. Considering the purpose of the compost, it is questionable whether BPs are proper feedstocks since BPs are mainly composed of carbon (C) but not N and P [14]. The increase in the C source by adding BPs may promote GHG emissions associated with direct emissions of $CO_2$ and energy consumption rather than enhancing the quality or quantity of byproducts. On the other hand, AD may be more suitable than composting, offering an energy-producing BP treatment strategy; C contained in BPs can be converted into $CH_4$. AD is a serial multi-stage biological process: hydrolysis, acidogenesis, acetogenesis, and methanogenesis [15,16]. Among the various steps, hydrolysis is often the rate-determining step of the BP the AD process, and hence, various strategies have been developed to enhance biodegradation [17,18].

Pretreatment technologies, which can be categorized as thermal, chemical, physical, and a combination of these, have been applied prior to AD [5,19]. In the thermal pretreatment test, the maximum biodegradation rate of 75% and $CH_4$ production yield (MPY) of $0.405 \pm 0.1$ $m^3$ $CH_4$/kg volatile solids (VS) were obtained from BPs at 150 °C, whereas these values were significantly low at 70 °C [20]. It was also reported that the thermo-chemical pretreatments (temperature of 35–90 °C and pH 10–12) enhanced the $CH_4$ potential of PHB and PLA by up to 100% compared to the control [14]. However, the high temperature and pH value should require an additional process of controlling the temperature and pH value of feedstock so as not to inhibit the activity of microorganisms involved in the AD process. Mechanical pretreatment can be used alone. For instance, anaerobic biodegradation rates of BPs were improved by 405% at the particle size of 100–250 μm than those obtained with a particle size of 500–1000 μm [21]. $CH_4$ potential of BPs was also increased along with the shortened lag period as the particle size decreased [20]. Although $CH_4$ production from BPs alone can be attained by using the AD process combined with pretreatment technologies, it might be difficult to operate a digester stably for a long period. The optimal C/N ratio for AD ranges between 20:1 and 30:1, but there is a probability of increasing the ratio of digestate due to BP characteristics, as carbon-rich materials [22]. Hence, BPs are commonly digested with other organic waste such as food waste (FW), vegetable waste, municipal sewage sludge, and livestock manure [11,23–25]. Among the organic wastes, FW has a high energy potential and a relatively short decay rate, which makes it an outstanding feedstock for AD [26]. Thus, the co-digestion of BPs and FW has been actively investigated and one that has already been successfully applied on site [27]. MPYs obtained from the co-digestion of BPs and domestic FW ranged widely, from 0.05 to 0.41 $m^3$ $CH_4$/kg VS, depending on the type of BPs [25]. However, no study has clearly shown the effects of FW addition on the $CH_4$ production and biodegradation of BPs, and the synergistic effects of co-digestion of BPs and FW have not been discussed in depth. To our knowledge, operating temperature was barely considered in the previous studies investigating co-digestion with BPs even though it is an important parameter which significantly influences the microbial community and metabolisms [28,29]. For this reason, the synergistic effects of co-digestion may show different impacts depending on the operating temperature.

Therefore, this study aimed to investigate the effects of FW addition on mesophilic and thermophilic AD of PLA and PHA, of which particle size was reduced to <0.5 mm, without other pretreatment methods. First, the maximum $CH_4$ potential of PLA, PHA, and FW was attained by performing biochemical methane potential (BMP) assays at 37 and 55 °C, and the results were compared. To evaluate the synergistic effect of co-digestion with FW, co-digestion BMP assays were performed at various mixing ratios of BPs and FW (FW/PLA or PHA = 95:5 and 90:10 on a weight basis), and the synergistic increment was calculated using the results obtained from mono- and co-digestion experiments. After digestion, the expected biodegradation rates of PLA and PHA were calculated by monitoring the changes in theVS concentrations of digestates in all bottles.

## 2. Materials and Methods

### *2.1. Substrate and Inoculum Preparation*

FW used for BMP assays was collected from the research institute cafeteria, where more than 950 meals are served per day, and consisted of vegetables (40 wt.%), fruits (15 wt.%), rice (25 wt.%), and fish/meat (20 wt.%). After adding 300 g of tap water, 700 g of FW was ground by a blender and then sieved through a 2 mm sieve to discard inorganic matter such as fish and meat bone and seeds. Raw PLA and PHA resin particles were purchased from Lotte Chemical, Seosan-si, Korea, and Hebei Luozheng Technology Co., Ltd., Hebei, China, respectively. Both BPs were ground by a variable speed rotor mill (FRITSCH, Idar-Oberstein, Germany) equipped with a 0.5 mm sieve ring after freezing with liquified nitrogen ($N_2$) to suppress their property changes caused by the frictional heating generated during crushing (Figure 1). Characteristics of prepared FW, PLA, and PHA are shown in Table 1. The seed sludge was taken from an anaerobic digester in a local wastewater treatment plant (Goyang-si, Korea) and precultured for two months at 37 and 55 °C to obtain the active mesophilic and thermophilic anaerobic microorganisms. The pH, alkalinity, and VS concentration of the sludge were 7.8, 4.9 g $CaCO_3$/L, and 38 g/L, respectively.

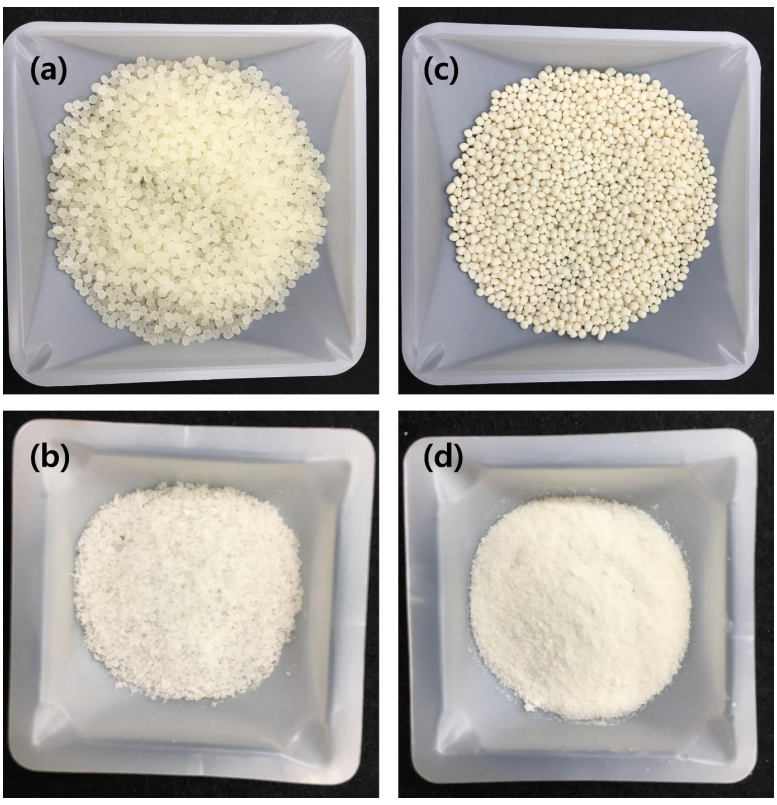


**Figure 1.** The images of raw and ground bioplastics (particle size $\leq$ 0.5 mm) ((**a**) raw PLA resin particle, (**b**) ground PLA, (**c**) raw PHA resin particle, (**d**) ground PHA).

**Table 1.** Characteristics of food waste and bioplastics (PLA and PHA).

| | Units | TS [a] | VS | COD |
|---|---|---|---|---|
| Food waste | g/kg | 85.7 | 81.9 | 114.5 |
| PLA | kg/kg | 0.99 | 0.98 | 1.3 |
| PHA | kg/kg | 0.99 | 0.98 | 1.5 |

[a] Total solids.

### *2.2. Biochemical Methane Potential (BMP) Assay*

BMP assays were conducted using an automated BMP testing system (AMPTS, BPC instruments Co., Sweden). Glass bottles (total volume 500 mL) with a working volume of 250 mL were utilized. The seed sludge was added to reach a VS concentration of 4.0 g/L, and a substrate concentration of mono- and co-substrates (FW, PLA, and PHA) was added at 2.0 g chemical oxygen demand (COD)/L, reaching the inoculum/substrate ratio of 2 g VS/g COD. Afterward, $NH_4Cl$, $KH_2PO_4$, and $FeCl_2{\cdot}4H_2O$ were supplemented to yield a COD/N/P/Fe ratio of 100:5:1:0.33. In addition, trace nutrients were added as follows (in mg/L): $NaHCO_3$ 1000; $MgCl_2{\cdot}6H_2O$ 100; $CaCl_2{\cdot}2H_2O$ 75; $Na_2MoO_4{\cdot}4H_2O$ 0.01; $H_3BO_3$ 0.05; $MnCl_2{\cdot}4H_2O$ 0.5; $ZnCl_2$ 0.05; $CuCl_2$ 0.03; $NiCl_2{\cdot}6H_2O$ 0.05; $CoCl_2{\cdot}2H_2O$ 0.5; $Na_2SeO_3$ 0.05. The initial pH was adjusted between 7.8 and 8.0 with a 3 N KOH solution, and then the prepared bottles were sealed with a plastic capper. The headspace in the bottle was purged with $N_2$ gas (99.99%) for 10 min to provide anaerobic conditions and placed in an incubator (IST series, Lab Companion). Temperature and agitation speed were controlled at 37 ± 1 or 55 ± 1 °C and 100 rpm, respectively. A blank was also prepared to subtract the produced $CH_4$ from the substrates. The biogas produced from the bottles was moved to other serum bottles for capturing $CO_2$ with 3 N KOH. The volume of pretreated biogas was measured and recorded during digestion until the amount of produced biogas was negligible. BMP assays were performed in duplicate, and the results were averaged. To analyze the cumulative $CH_4$ production curve, two types of modified Gompertz models were applied to determine $CH_4$ production potential and lag period [30].

### *2.3. Liquid and Solid Characteristics*

Concentrations of total solids (TS), VS, COD, and alkalinity (as g $CaCO_3$/L) were measured according to the standard methods [31]. pH value was measured using a pH meter (HM-3R, DDK-TOA). The ground BPs (PLA and PHA) were used for measuring COD of BPs, and the results were compared with the theoretical COD of BPs. Based on the BP mass and molecular structure, the calculated theoretical COD for PLA and PHA was 1.33 kg and 1.67 kg COD/kg PHA, respectively. As shown in Table 1, the differences between the values in this study and theoretical values seemed to be acceptable.

### *2.4. Calculations*

To investigate the synergistic effects of co-digestion on $CH_4$ production, theoretical MPY in co-digestion tests was calculated using the MPY obtained from mono-digestion of each substrate (Equation (1))

$$M_{co} = (M_{bp} \times W_{bp}) + (M_{fw} \times W_{fw}) \quad (1)$$

where $M_{co}$ is the theoretical MPY of co-digestion (mL/g COD), $M_{bp}$ is the MPY of bioplastic mono-digestion (mL/g COD), $W_{bp}$ is the amount of COD addition of each BP in a bottle (g COD), $M_{fw}$ is the MPY of food waste mono-digestion (mL/g COD), and $W_{fw}$ is the amount of COD addition of food waste in a bottle (g COD). The synergistic effect was calculated as follows (Equation (2)):

$$\text{Synergistic effect (\%)} = (M_{ex}/M_{co}) \times 100\ (\%) \quad (2)$$

where $M_{co}$ is the theoretical MPY calculated by using Equation (1) (mL/g COD), and $M_{ex}$ is the actual MPY from the co-digestion test (mL/g COD).

## 3. Results and Discussion

### *3.1. Mono-Digestion of Food Waste, PLA, and PHA*

BMP assays were carried out to evaluate the $CH_4$ potential of individual substrates with mesophilic and thermophilic AD. The pH values of digestates in the bottles were maintained above 7.5 during digestion, indicating there was no substrate and product inhibition in methanogenesis. Figure 2 shows the cumulative $CH_4$ production of mono-digestion of FW, PLA, and PHA for 20 d of operation. In the FW- and PHA-fed tests, $CH_4$ production was detected under mesophilic and thermophilic conditions as soon as the experiment began, while it started to evolve after 2–4 d in the PLA-fed one. Interestingly, a significant amount of $CH_4$, 38–42% of total $CH_4$ production, was produced within 1 d only in the FW-fed test. The expected reason is that easily biodegradable materials such as carbohydrates (rice) were contained in FW, and cooking and grinding might simulate hydrolysis [5,32]. In the case of PLA- and PHA-fed tests, lag periods were within the reported results obtained from untreated PLA considering their particle size (<0.5 mm). The lag period for $CH_4$ production was shortened as the particle size of BPs decreased, and the lag period of <0.1 d was obtained from PLA with a particle size of 0.5 mm without another pretreatment method [20,21]. However, there were some studies reporting longer periods (>10 d). This may be because commercial BP products were used as the main substrate, not raw BPs [14,33]. In general, various agents are used to make commercial BP products to strengthen mechanical stability and improve formability, which may disturb the biodegradation of BPs and the biological reactions associated with the AD process. $CH_4$ production of PLA and PHA occurred at a stable, constant rate under both temperature conditions. On the other hand, there was no large difference in cumulative $CH_4$ production curves for each substrate between mesophilic and thermophilic conditions.

The model fitting results ($r^2 \geq 0.97$) for mono-digestion are arranged in Table 2. Considering the purpose of BMP assays, $CH_4$ production potential values are mainly discussed in this section. If the cumulative $CH_4$ production curve had two lag periods, the parameters for the second one are summarized in the table to show the maximum $CH_4$ production potential of substrates. During digestion, the $CH_4$ production potential of FW, PLA, and PHA at 37 °C were 126, 5.8, and 76.9 mL, while their values increased by 10.6%, 54.6%, and 10.6%, respectively, at 55 °C. The operating temperature is an important factor in the AD process, affecting thermodynamic and enzymatic reactions. For example, propionate degradation, which is one of the endergonic reactions, is promoted at a higher temperature, resulting in the enhancement of $CH_4$ production [33]. Bernat et al. (2021) also showed similar results in which biogas production obtained from PLA under thermophilic conditions was almost two times higher compared to mesophilic conditions [29]. In terms of MPY, shown in Table 3, 252.0–281.9 mL/g COD, equivalent to 72–81% of theoretical MPY (350 mL $CH_4$/g COD), was obtained from the FW at 37 and 55 °C, which is within common range of MPY for FW [34]. The MPYs of PLA at 37 and 55 °C were only 11.6 and 25.6 mL/g COD, respectively. $CH_4$ production from PLA without pretreatment seemed difficult, in particular for a short operating time. Its $CH_4$ production through either mesophilic or thermophilic AD was quite low, obtaining only <5% of theoretical MPY during 30–60 d of digestion [20]. With sufficient time for digestion, the MPYs of PLA could be increased. Compared to PLA, significantly higher MPYs were attained from PHA (153.8 at 35 °C and 172.0 mL/g COD at 55 °C), which seemed slightly low compared with the previous literature results but acceptable considering operating time. For example, MPYs obtained from various types of PHB for 60 d of operation were 199–316 mL $CH_4$/g COD under mesophilic conditions, while $CH_4$ production from PLA was negligible [14].

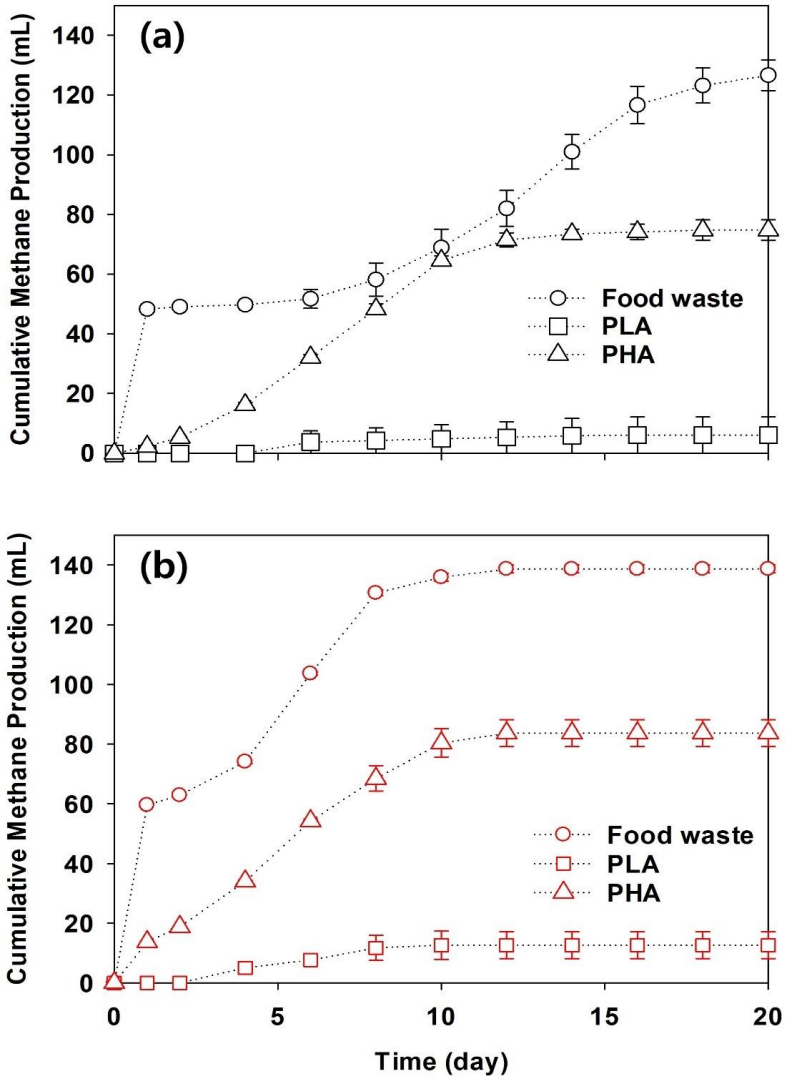


**Figure 2.** The cumulative methane production from food waste, PLA, and PHA during biochemical methane potential (BMP) assays under (**a**) mesophilic and (**b**) thermophilic conditions.

**Table 2.** Kinetic parameters in biochemical methane potential assays from fitting with the modified Gompertz equation.

| Mixing Condition | | P [a] (mL) | Rm [b] (mL/d) | λ [c] (d) | $r^2$ |
|---|---|---|---|---|---|
| Mesophilic AD (37 °C) | Food waste [d] | 126.0 | 6.9 | 6.59 | 0.99 |
| | PLA | 5.8 | 1.2 | 3.59 | 0.99 |
| | PHA | 76.9 | 9.5 | 2.42 | 0.99 |
| | F95L [d] | 89.4 | 5.2 | 6.11 | 0.99 |
| | F90L [d] | 78.6 | 4.1 | 3.46 | 0.99 |
| | F95H [d] | 123.1 | 6.4 | 1.34 | 0.99 |
| | F90H [d] | 123.2 | 6.4 | 2.02 | 0.99 |
| Thermophilic AD (55 °C) | Food waste | 141.0 | 17.2 | <0.1 | 0.97 |
| | PLA | 12.8 | 2.4 | 2.26 | 0.99 |
| | PHA | 86.0 | 10.4 | 0.42 | 0.99 |
| | F95L | 114.1 | 13.6 | <0.1 | 0.97 |
| | F90L | 91.4 | 15.2 | <0.1 | 0.97 |
| | F95H | 134.4 | 18.4 | <0.1 | 0.97 |
| | F90H | 130.4 | 23.0 | <0.1 | 0.98 |

[a] $CH_4$ production potential; [b] $CH_4$ production rate; [c] lag period; [d] second lag period.

**Table 3.** $CH_4$ production yield obtained from the mono- and co-digestion experiments and the synergistic effect of co-digestion on $CH_4$ production.

| Experimental Conditions | | Theoretical $CH_4$ Production Yield (mL/g COD) | Actual $CH_4$ Production Yield from Co-Digestion (mL/g COD) | Synergistic Effect (%) |
|---|---|---|---|---|
| Mesophilic AD (37 °C) | Food waste | - | 252.0 | - |
| | PLA | - | 11.6 | - |
| | PHA | - | 153.8 | - |
| | F95L | 164.9 | 178.9 | 8.5 |
| | F90L | 124.2 | 157.2 | 26.6 |
| | F95H | 213.1 | 246.2 | 15.5 |
| | F90H | 196.3 | 246.4 | 25.5 |
| Thermophilic AD (55 °C) | Food waste | - | 281.9 | - |
| | PLA | - | 25.6 | - |
| | PHA | - | 172.0 | - |
| | F95L | 189.1 | 228.3 | 20.7 |
| | F90L | 145.6 | 182.7 | 25.5 |
| | F95H | 238.4 | 268.7 | 12.7 |
| | F90H | 219.6 | 260.7 | 18.7 |

### *3.2. Co-Digestion and Its Synergistic Effect*

Figure 3 shows the cumulative $CH_4$ production in the co-digestion of FW and BPs (PLA or PHA) under mesophilic and thermophilic conditions. The $CH_4$ production curves were similar to those of FW mono-digestion, probably due to the high FW portion (90–95% on a weight basis). Although overall curve tendencies with two lag periods for $CH_4$ production seemed not to differ significantly between 37 and 55 °C, the slope of the curves at 55 °C was steeper compared to 37 °C. Based on the model fitting results shown in Table 2, the $CH_4$ production potential of the mixtures of FW and PLA at 37 °C were 89.4 and 78.4 mL in F95L and F90L, while the values at 55 °C increased by 21.6% and 13.9%, respectively. Compared to PLA, the $CH_4$ production potential of F95H and F90H was higher under the two temperature conditions, while the differences in $CH_4$ production potential between 37 and 55 °C were smaller: the values at 55 °C were only 5.5–8.4% higher than those at 37 °C (Table 2).

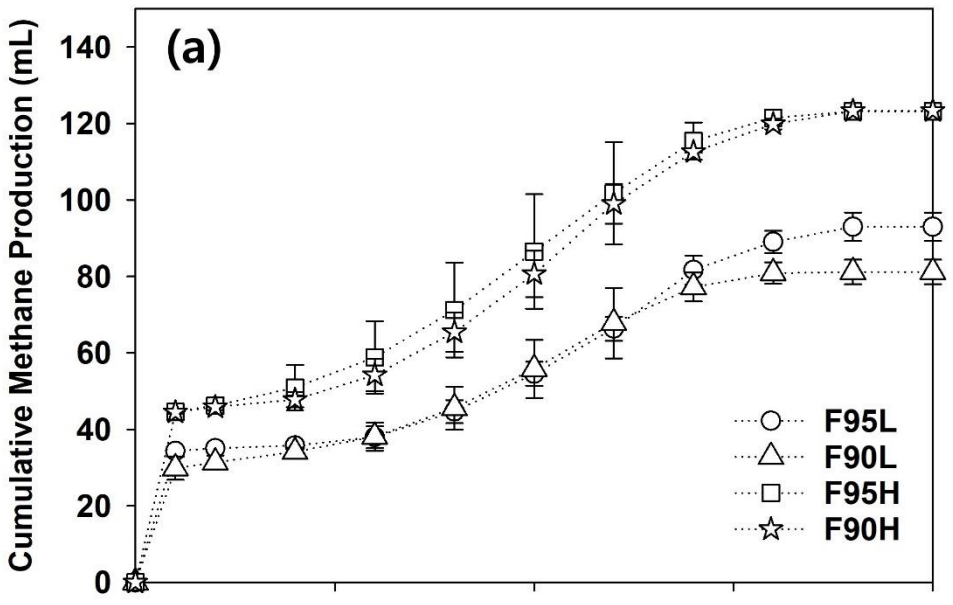


**Figure 3.** *Cont.*

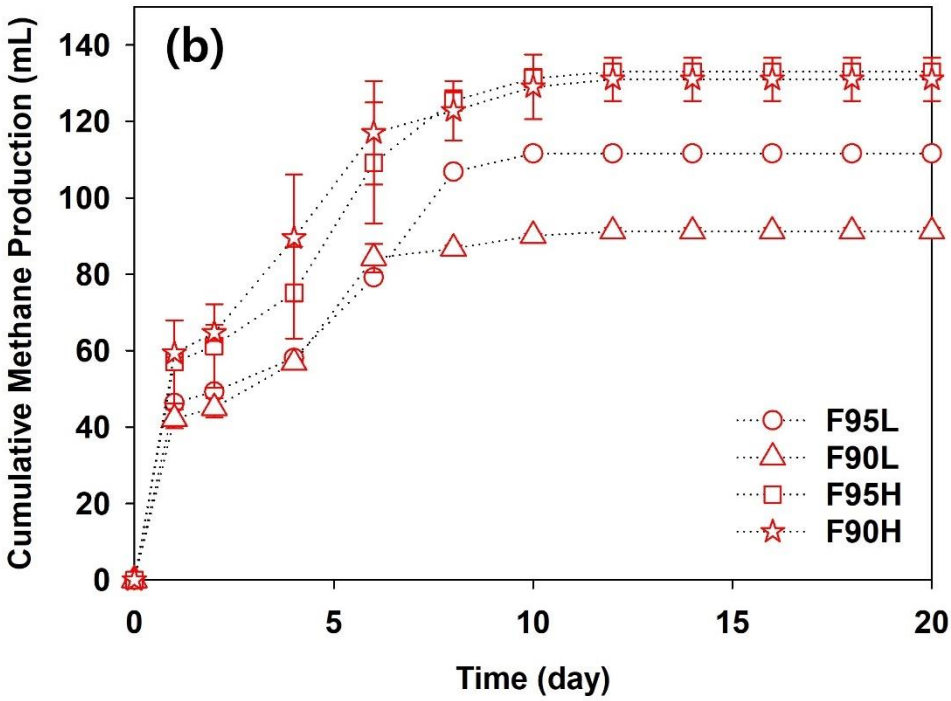


**Figure 3.** The cumulative methane production in the anaerobic co-digestion of food waste and bioplastics (PLA or PHA) under (**a**) mesophilic and (**b**) thermophilic conditions at the various substrate mixing ratios (FW/PLA or PHA = 95:5 and 90:10 on a weight basis).

In order to estimate the synergistic effect of co-digestion, theoretical MPYs were calculated using MPYs obtained from mono-digestion at 37 and 55 °C according to Equations (1) and (2), and the results are summarized in Table 3. Compared to the co-digestion of PHA and FW, higher synergistic effects were attained in the co-digestion of PLA and FW, except for F95L. The values obtained for F95L and F90L under thermophilic conditions increased by 20–25% compared to theoretical MPT. As the addition portion of BPs increased, the effects were enhanced at 37 and 55 °C. Especially, the synergistic effect for F95L was only 8.5% and increased by approximately three times when the portion of PLA was increased from 5% to 10%. On the other hand, co-digestion with FW seemed to have more impact on the synergistic effect at 37 compared to 55 °C since its enhancement was observed in F90L, F95H, and F90H, but the gaps were negligible. The expected reason for the additional $CH_4$ production by synergistic effect is nutrient balance. To maintain the optical conditions for the AD process, it is important to provide a suitable environment where anaerobic microorganisms involved with AD can grow well [28]. For their growth, C, N, P, and other nutrients are required, but it may be impossible to supply only BPs as a substrate since common BPs mainly consist of C, as mentioned above [35]. The supply of easily biodegradable substrate to non-biodegradable can facilitate the rapid development of the active microbial community, leading to a shortened lag period and fast $CH_4$ production [36].

### *3.3. Biodegradation of Bioplastics*

In order to estimate the biodegradation rates of PLA and PHA, VS concentrations of digestates present in the bottles were measured at the end of the operation. Initial and final VS concentrations are shown in Tables 4 and 5, respectively. For 20 d of digestion, VS concentrations of inoculum were reduced by 22.4 and 26.8% at 37 and 55 °C, respectively. With these results, the expected VS reduction efficiency of FW ranged from 79.0 to 88.2% at the temperatures of 37 and 55 °C (ex. $(1 - 0.30/1.43) \times 100 = 79$ at 37 °C). The VS reductions of PLA and PHA at 37 °C were 6.0 and 49.1%, respectively, and the values obtained at 55 °C increased to 13.7 and 52.3%, respectively. Like the biodegradation rates of PLA in

this study, untreated PLA was slightly degraded under mesophilic conditions, obtaining <3% VS reduction for 25–28 d of digestion [20,37]. According to the previous studies, higher biodegradation efficiency (54%) is attained in mono-digestion, which is probably due to the long operating time and/or small particle size [19,38]. In the co-digestion of BPs and FW, VS reduction efficiencies of PLA and PHA were significantly enhanced at both temperatures. In the case of PLA, the VS reductions at 37 and 55 °C were 2.8–4.3 and 1.8–2.2 times, respectively, higher than the values from mono-digestion, while the values for PHA were only 1.2–1.5 times higher at both temperatures. The enhancement of VS reduction by co-digestion was observed in a previous study. According to the report by Benn and Zitomer, (2018), the VS reduction of PHB was approximately 75% during mono-digestion, but that increased to 81% when PHB was digested with other organic waste [14].

**Table 4.** Initial VS concentration of the digestates in each bottle.

| | Inoculum (g VS/L) | Food Waste (g VS/L) | PHA (g VS/L) | PLA (g VS/L) | Total (g VS/L) |
|---|---|---|---|---|---|
| Blank | 4.00 | - | - | - | 4.00 |
| Food waste | 4.00 | 1.43 | - | - | 5.43 |
| PLA | 4.00 | - | - | 1.51 | 5.51 |
| PHA | 4.00 | - | 1.31 | - | 5.31 |
| FW95L | 4.00 | 0.90 | - | 0.54 | 5.44 |
| FW90L | 4.00 | 0.63 | - | 0.76 | 5.39 |
| FW95H | 4.00 | 0.85 | 0.51 | - | 5.36 |
| FW90H | 4.00 | 0.58 | 0.70 | - | 5.28 |

**Table 5.** Expected VS concentration of the digestates in each bottle after digestion.

| Experimental Conditions | | Inoculum (g VS/L) | Food Waste (g VS/L) | PLA [d] (g VS/L) | PHA [d] (g VS/L) | Total [a] (g VS/L) |
|---|---|---|---|---|---|---|
| Mesophilic AD (37 °C) | Blank | 3.10 | - | - | - | 3.10 |
| | Food waste | 3.10 | 0.30 [b] | - | - | 3.41 |
| | PLA | 3.10 | - | 1.42 | - | 4.52 |
| | PHA | 3.10 | - | - | 0.67 | 3.77 |
| | F95L | 3.10 | 0.19 [c] | 0.41 | - | 3.71 |
| | F90L | 3.10 | 0.13 [c] | 0.52 | - | 3.75 |
| | F95H | 3.10 | 0.18 [c] | - | 0.20 | 3.48 |
| | F90H | 3.10 | 0.12 [c] | - | 0.26 | 3.48 |
| Thermophilic AD (55 °C) | Blank | 2.93 | - | - | - | 2.93 |
| | Food waste | 2.93 | 0.17 [b] | - | - | 3.10 |
| | PLA | 2.93 | - | 1.30 | - | 4.23 |
| | PHA | 2.93 | - | - | 0.62 | 3.55 |
| | F95L | 2.93 | 0.11 [c] | 0.30 | - | 3.33 |
| | F90L | 2.93 | 0.07 [c] | 0.46 | - | 3.46 |
| | F95H | 2.93 | 0.10 [c] | - | 0.10 | 3.13 |
| | F90H | 2.93 | 0.07 [c] | - | 0.16 | 3.15 |

[a] Total: VS concentration of the digestate in each bottle after mesophilic or thermophilic anaerobic digestion; [b] Food waste: estimated VS concentration for food waste (food waste = total − inoculum); [c] Food waste: estimated VS concentration for food waste (food waste = initial VS concentration of food waste × (1 − degradation rate of food waste)); [d] PLA and PHA: estimated VS concentration for PLA or PHA (PLA or PHA = total − inoculum − food waste).

## 4. Conclusions

The batch experiments were carried out to assess the effect of co-digestion of FW and ground BPs (particle size < 0.5 mm) on the $CH_4$ production and biodegradation in mesophilic and thermophilic AD. $CH_4$ production was observed within 4 d at 37 and 55 °C in all mono-digestion tests. The MPYs of FW and PHA reached 72–81% and 42–49% of theoretical MPY, respectively, while that of PLA was <8%, which meant it seemed difficult to produce $CH_4$ efficiently from PLA alone within 20 d. FW addition could significantly

enhance the performances of BP AD, especially PLA. MPYs obtained in the PHA-fed tests at 37 and 55 °C were 15.5–25.5% and 12.7–18.7% higher than the theoretical values, and the synergistic effects in PLA-fed tests by co-digestion with FW were above 20% except for one case. On the other hand, the synergistic effect was slightly enhanced at the higher temperature. Without FW addition, PHA was degraded by approximately 50% during mesophilic and thermophilic AD, whereas PLA was barely degraded, obtaining the maximum biodegradation rate of 14% at 55 °C. The degradation rates of BPs were also significantly increased to 22.8–44.1% for PLA and 61.1–80.6% for PHA, respectively, by co-digestion with FW.

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