

Article

Biodegradation of Bioplastic Using Anaerobic Digestion at Retention Time as per Industrial Biogas Plant and International Norms

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Abstract: Bioplastics are gaining interest as an alternative to fossil-based plastics. In addition, biodegradable bioplastics may yield biogas after their use, giving an additional benefit. However, the biodegradability time in international norms (35 days) far exceeds processing times in anaerobic digestion facilities (21 days). As the bioplastic packaging does not indicate the actual biodegradability, it is important to understand the time required to biodegrade bioplastic if it ends up in the anaerobic digestion facility along with other organic waste. For this work, cellulose bioplastic film and polylactic acid (PLA) coffee capsules were digested anaerobically at 55 °C for 21 days and 35 days, which are the retention times for industrial digestors and as set by international norms, respectively. Different sizes of bioplastics were examined for this work. Bioplastic film produced more biogas than bioplastic coffee capsules. The biodegradability of bioplastic was calculated based on theoretical biogas production. With an increase in retention time, biogas production, as well as biodegradability of bioplastic, increased. The biodegradability was less than 50% at the end of 35 days for both bioplastics, suggesting that complete degradation was not achieved, and thus, the bioplastic would not be suitable for use in biogas digesters currently in use.

Keywords: bioplastic; biodegradability; thermophilic; bio-based; anaerobic digestion

1. Introduction

The rise in use of plastic has increased plastic production from 1.5 million metric tons in 1950 to 359 million metric tons in 2018 [1]. Its low-cost, lightweight, durable, and chemical-resistant properties have increased its demand [2–4]. Plastic packaging contributed to 57% of the total plastic waste produced in 2018 [5] and covers a significant fraction of municipal waste [6]. These wastes end up in recycling, thermal plants, or dumping sites [7]. Of plastic wastes produced in 2018, 20% were recycled, and energy was recovered from another 20%, but on the other hand, 29% of plastic waste was landfilled or dumped, while 31% of the waste was disposed improperly or leaked [5]. Owing to the fact that plastic is non-biodegradable and uncontrolled incineration produces toxic gases, these options are not environmentally friendly [4,8]. The problems related to plastic management and environmental concern have shifted the interest to bioplastics [9–11]. Bioplastics are derived from bio-based materials, are biodegradable, or both [12]. Bioplastics reduce carbon footprint, provide better waste management options, and reduce dependency on fossil fuel, which can help in limiting greenhouse gas emission [13,14]. While bioplastics have been gaining enormous interest in replacing their conventional plastic counterparts, it is also necessary to understand the end-of-life use of bioplastics and whether they are compatible with existing processing plants [15].

Most biodegradable plastics are either recycled, composted, or landfilled. Landfilling of bioplastics, along with other organic waste, produces methane, which is released into the environment [16], while composting produces carbon dioxide; both contribute to greenhouse gas emissions [17]. On the other hand, the anaerobic digestion process involves breaking down the organic matter in the absence of oxygen into biogas. Anaerobic digestion, as compared to aerobic digestion, produces energy in the form of biogas and has shorter retention time [18]. Polylactic acid (PLA) and cellulose-based bioplastics are considered biodegradable and are mostly used in food packaging and 3D printing [12,19]. A study done by Piemonte [9] on the final disposition of bioplastic using composting, anaerobic digestion, incineration, or recycling shows that anaerobic digestion of PLA bioplastic led to greenhouse gas (GHG) savings higher than those of open-loop recycling, incineration, and composting processes. Anaerobic digestion can be a promising process for the management of bioplastic, as it can help in energy recovery in the form of biogas and produces less greenhouse gas emission in a shorter retention time. In a home composting facility, complete degradation of PLA does not occur and is a relatively slow process [19], where degradation time is usually more than six months. The previous study on PLA degradation under the composting condition of 90 days using the GB/T19277–2003/ISO 14855-1:2005 standard showed 80% degradation at 80 days' incubation time [20]. Injection-molded flower pots made from 100% PLA incubated at 58 °C for 60 days did not show any change in the physical appearance and had only 13% ± 3% degradation [21]. PLA bottles showed 84.2% and 77.8% mineralization and cellulose showed 86% mineralization after 58 days in two different simulated composting systems [22]. A study by Itävaara et al. comparing aerobic and anaerobic biodegradation of polylactide found 10% degradation in 210 days at 37 °C under the aerobic conditions as compared to 60% degradation in 100 days at 37 °C under anaerobic conditions, and concluded that the lactic acid is more favorable for anaerobic microorganisms [23]. The biodegradation depends on factors like bioplastic chemical composition, the processing technique (aerobic/anaerobic), and the operating conditions (temperature/time) [24]. As compared to aerobic degradation of bioplastic, anaerobic degradation of different types of bioplastics has not been studied extensively [25].

Despite the advantage of anaerobic digestion, the question that arises is whether the biodegradability potential of bioplastics is fully used [25]. The extent of biodegradability of bioplastics must be mentioned so that the existing digestion facilities can make a decision on whether to process it [26]. Previous studies have been carried out on anaerobic digestion of bioplastics. The anaerobic degradation of polylactide was conducted by Itävaara et al. using the ASTM D 5210 aquatic anaerobic test at 37 °C and anaerobic solid state digestion test ASTM D5511 at 52 °C [23]. The mesophilic condition resulted in 60% mineralization at 100 days, while thermophilic condition resulted in 60% mineralization in 40 days, suggesting that biodegradation is faster at higher temperatures. The anaerobic biodegradation was faster than aerobic degradation under the thermophilic condition. In a study by Shin et al., an ASTM D 5210-91 test was carried out at mesophilic condition for biodegradation of PLA [27], and no biodegradation occurred in 100 days. Biodegradation did not occur in 170 days in a high solid anaerobic digestion test of PLA conducted by Kolstad et al. [28]. The MODA-B apparatus was used by Yagi et al. [18] to study PLA powder and cellulose degradation at 35 and 55 °C. A total of 80% of degradation occurred in cellulose in mesophilic and thermophilic conditions in 15 and 13 days, respectively. Degradation was below 15% for PLA at 35 °C and 77 days. At 55 °C, 60% of PLA degraded in 30 days and 90% degraded in 90 days. ISO 14853 was used for anaerobic degradation study of PLA and cellulose for 28 days [29]. PLA did not degrade in 28 days, while 62.2% degradation of cellulose was reported. Gómez and Michel Jr [30] state that after 50 days of anaerobic digestion, only 20–25% of bio-based materials were converted into biogas. Bátori et al. [25] state that some biopolymers can be degraded at hydraulic retention times, usually applied at biogas plants, while other biopolymers hardly degrade or only under thermophilic conditions. Results on anaerobic biodegradation of bioplastics differ a lot, and many studies were performed at conditions that do not match industrial biogas plants, and, therefore, the effectiveness of anaerobic biodegradation of bioplastics remains unanswered.

This study aims to compare the biodegradation potential and biogas production of bioplastics at time spans equal to those of industrial biogas plants and the international norm NEN-EN 13432:2000 [31] in order to investigate to which extent the norm on bioplastics represents industry practices. According to EN 13432:2000, for anaerobic biodegradation of bioplastic used for packaging, the test period has a maximum of two months and the percentage of biodegradation has to be 50% or more of the theoretical value of the bioplastic, and for disintegration from anaerobic bio-gasification, the test duration must have a maximum of five weeks with 90% of the original dry matter of the material passing through a <2 mm sieve [31]. Industrial anaerobic digestion facilities operate, however, with a hydraulic retention time of 21 days. Therefore, this study focuses on biodegradation and biogas production of bioplastics used as coffee packaging and coffee capsules by an anaerobic digestion process for 21 days, the retention time adopted by industrial biogas plants, and for 35 days, which is indicated by the international norm. Bioplastics were digested at thermophilic conditions, since several bioplastics do not degrade at mesophilic conditions [25]. Thermophilic digestion of different sizes shows different biodegradation rates [32]. So, the effect of particle size and type of plastic (rigid versus foil) on biodegradation and biogas production was investigated.

It needs to be noted, however, that most of the previous studies operated at very long hydraulic retention times, over 40 days, much longer than hydraulic retention times usually applied at biogas plants [25], and the focus was on higher biogas yield. Anaerobic digestion facilities generally operate on the retention time of 15–20 days [33]. Furthermore, the anaerobic biodegradability test was studied using standards like ASTM D5210, ISO 14853, ASTM D5511, and MODA apparatus, while EN 13432 has not been used for anaerobic biodegradation tests. This test provides a measure for biodegradation as well as disintegration by passing it through 2 mm sieve. This process has not been reported in previous works. It is critical for the operators to understand if the bioplastics degrade in a similar way and do not create any operational challenges. Thus, the novelty of this work is the focus on understanding whether the idea of anaerobic digestion of bioplastics meets the industrial practices and set standards. If not, then anaerobic digestion could not be a viable option for energy recovery from bioplastics, unless one develops new facilities for bioplastics themselves to operate for longer residence times. The anaerobic digestion process in this study is strictly set to the retention time of the industrial digester and according to the standard. The work provides an insight into sample preparation methods in terms of sizes for higher energy recovery from bioplastics at the end of their use. Furthermore, this work gives a new approach in understanding the international norms and the industrial practice that could help in defining future research around biodegradability of different types of bioplastics, which could help in making policies on the end-of-life use of bioplastics.

2. Materials and Methods

2.1. Materials

Biomethane potential was measured using a gas displacement method. For the anaerobic digestion process, a 500 mL Schott reactor bottle was used. It was connected to a 1 L bottle containing a barrier solution that acted like a liquid gasometer (270 g/L NaCl solution corrected to pH 2 with 1 M HCl) that, in turn, was connected to a measuring cylinder. Thermophilic sludge obtained from Attero, a waste management company in the Netherlands, was used as inoculum. For the experiment, rigid bioplastic used as a coffee capsule and foil bioplastic used as coffee packaging were used. Film bioplastics were made of cellulose, and compostable film and coffee capsules were made of PLA. Bioplastic was then cut into pieces of 1 × 1 cm, 2 × 2 cm, and 3 × 3 cm for both the rigid bioplastic and the foil bioplastic; they were named 1F, 2F, 3F, 1R, 2R, and 3R from now, with the number representing the size of the bioplastic, F representing Foil, and R representing Rigid. Bioplastics were digested without coffee debris on them.

2.2. Methods

The dry matter and volatile solid were calculated using the U.S. Environment Protection Agency (EPA) Method 1684. Dry matter is the amount of residue left in the vessel after heating it to a temperature of 105 °C overnight. Volatile solid is the weight lost after the sample has been heated overnight to 550 °C. The sludge obtained from Attero had a dry matter of 20.3% and a volatile solid of 40%. It was allowed to stay for 3–4 days at room temperature under the fume hood to decrease the biogas production activity produced solely by sludge and remaining organics in the sludge. Sludge was then slowly heated in the oven to a temperature of 37 °C and increased to 55 °C. Film bioplastic had a dry matter of 95.05% and a volatile solid of 96.19%, while rigid bioplastic had a dry matter of 99.25% and a volatile solid (VS) of 82.57%. The inoculum to bioplastic ratio was maintained at 2:1 (dry matter). The pH of the mix was measured using an HACH HQ40D meter.

The reaction was carried out in a 500 mL bottle with 400 mL operating volume for 21 and 35 days. Digestion was conducted for two scenarios: (i) Blank (sludge only) and (ii) mixture of sludge and bioplastic. One of the mixtures included sludge and film bioplastic, and another included sludge and rigid bioplastic. Bioplastics along with 10 mL macronutrients (13.5 g/L KH₂PO₄, 13.2 g/L NH₄PO₄, 2.5 g/L MgCl₂·6H₂O, and 1.88 g/L CaCl₂·H₂O) and 1 mL micronutrients (160 mg/L MnCl₂·4H₂O, 50 mg/L FeCl₂·4H₂O, 12 mg/L ZnCl₂, 5 mg/L CoCl₂·6H₂O, 2.5 mg/L (NH₄)₆Mo₇O₂·4H₂O, 2.5 mg/L AlCl₃·6H₂O, 1.7 mg/L CuCl₂·2H₂O, and 1.25 mg/L H₂BO₃) were added in the reactor bottle. Oxygen-free deionized water was added to make up the total reactor volume of 400 mL. Sludge diluted to 2% VS was then added to the reactor, and the mixture was stirred. pH was measured immediately, and the bottle was sealed. Nitrogen was flushed to the reactor headspace for 5 minutes to purge out the oxygen in the reactor. The barrier solution bottle, which acted as a gasometer, was attached to the reactor bottle. The reactor bottle was put in the water bath that was maintained at 55 °C, and was allowed to stabilize for 15 minutes. During this stabilization process, the hose from the reactor bottle to the barrier solution was clamped, which was opened after 15 minutes. A small amount of liquid was displaced from the gasometer to the measuring cylinder, and the experiment was started. The amount of barrier solution that was displaced by the gas was noted daily, and it represented the amount of gas produced due to the digestion process.

A total of 21 reactors were set up for this process: Control (C), foil 1 × 1 cm (1F), foil 2 × 2 cm (2F), foil 3 × 3 cm (3F), rigid 1 × 1 cm (1R), rigid 2 × 2 cm (2R), and rigid 3 × 3 cm (3R), all in triple replication. After 21 and 35 days, the gas collected in the barrier solution bottle was forced into a gasbag, which was then connected to the Binder COMBIMASS® gas analyzer. The reactor liquid was passed through a 2 mm sieve to collect plastic of size greater than 2 mm. The weight of the plastic fraction >2 mm was determined. The collected plastic was washed with distilled water, and the OLYMPUS SZ X7 stereo microscope was used to see the image of the bioplastics.

3. Results

3.1. Biogas Production

Bioplastic film produced cumulative biogas of 197 (1F), 146 (2F), and 138 mL (3F) per gram volatile solid added for the first 21 days and 283 (1F), 264 (2F), and 252 (3F) mL per gram volatile solid added at the end of 35 days, as shown in Figure 1. The 1 × 1 cm bioplastic film showed maximum biogas production among other sizes during all periods of incubation. The 2 × 2 cm and 3 × 3 cm bioplastic films showed a similar pattern of biogas production. Sizes of plastic showed a small difference in the biogas production, with smaller pieces of plastic producing more biogas than larger pieces.

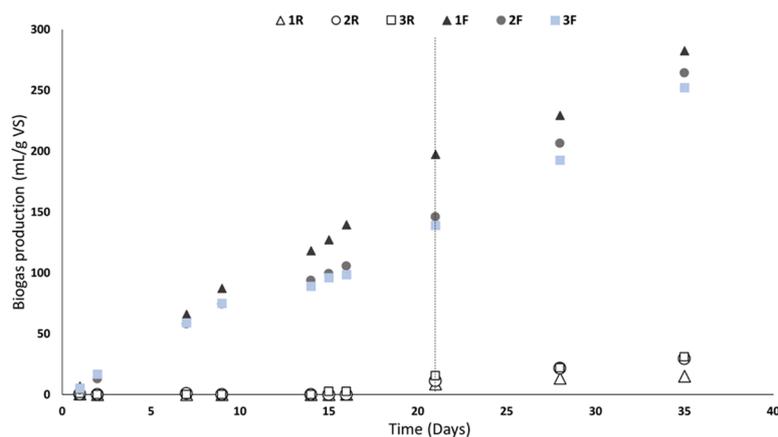


Figure 1. Biogas production over time with the dotted line indicating a 21 day operational period, equal to operating times of industrial biogas plants. The numbers indicate the sizes of the bioplastics (1 × 1 cm, 2 × 2 cm, and 3 × 3 cm), while F stands for foil and R stands for rigid.

Methane and carbon dioxide concentrations in the headspace increased with retention time, with methane concentrations of 41–43% in the biogas after 35 days, as shown in Figure 2. The conventional biogas plant has a methane concentration of 50–85% [34]. Interestingly, the methane production was not leveling off at 35 days, meaning that biodegradation and, consequently, biogas production had not finished. Increasing retention time could increase biogas production. Rigid bioplastic capsules produced very little biogas for all of the sizes for 35 days, and due to the low production, biogas composition could not be measured. The initial and final pH levels were around pH 7 in accordance with the optimum methane-forming conditions, as indicated in the literature [35]. Our results for biogas production from cellulosic film bioplastic were lower than reported in the literature for cellulose paper, which yielded cumulative methane gas production of 220.8 mL after 17 days [36]. Previous research indicates the biomethane production of PLA to be 282 mL per gram VS at thermophilic conditions [37], while, in this research, the production was very low. Industrial digesters use 21 days of retention time for organic wastes, and if a rigid bioplastic cup is allowed to stay in the digester for this duration in mono-digestion conditions, a negligible contribution in biogas production will be observed. Co-digestion of bioplastic could be another option to enhance biogas production [37,38].

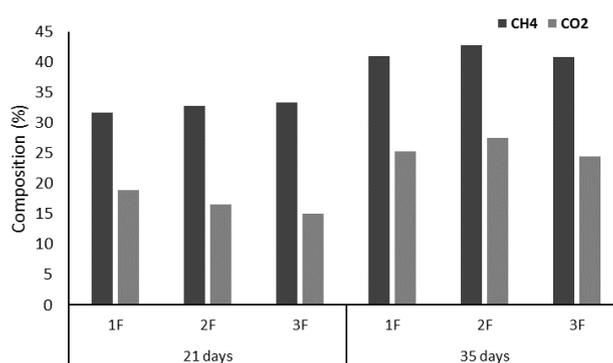


Figure 2. Biogas composition after 21 days of operation (typical retention times of industrial anaerobic digestion facilities) and 35 days of operation (retention times as set by international norm NEN-EN 13432:2000).

3.2. Anaerobic Biodegradation

Cups/rigid bioplastic had lower biodegradation than foils. After five weeks of digestion, 35–40% of foil bioplastic remained in the reactor bottle, while for cups/rigid bioplastic, this was 48–70%. The biodegradation test on the basis of theoretical biogas production (1 g VS yields, in theory, 1.5 L

biogas at vapor pressure corrected at 55 °C) [31] resulted in 12.7%, 9.4%, and 9.0% biodegradation, while rigid bioplastic showed biodegradation of less than 1% for a retention time of 21 days. For a retention time of 35 days, the biodegradability for 1F, 2F, and 3F was 18.3%, 17.1%, and 16.3%, respectively, while rigid bioplastic showed degradation of less than 3%. At the higher retention time, the biodegradability for different sizes of bioplastic was almost comparable. Both types of bioplastic used in this experiment showed less than 50% degradation and did not follow the norm for anaerobic biodegradability. The biodegradability rate in the experiment was lower than as reported by Yagi et al. for biodegradation of cellulose and PLA [32]. Low biodegradability could be due to the lack of stirring in the process, and greater thickness of the bioplastic capsule could be another reason affecting biodegradability.

For both foil and rigid bioplastic, the bioplastic after anaerobic digestion for 35 days was retained in the 2 mm sieve, suggesting that it did not disintegrate, as shown in Figure 3. The bioplastic film after the digestion still seems to be intact, although the layers of the foil (bioplastic and aluminum foil) did separate and had a dark green/brownish appearance. The rigid cups were also intact and had slight brown spots. With the stereo microscope, it was visible that for film bioplastic, the layers had separated but were still intact, while for the rigid bioplastics, some cracks on the surface were visible, and some fibers stood out from the edge (Figure 3). The lower biodegradability of the bioplastics could lead to clogging and other operational challenges in an industrial biogas plant.



Figure 3. Physical appearance of bioplastic at different stages.

4. Conclusion

Bioplastics as a packaging material are gaining popularity along with the claims that these bioplastics are biodegradable. These claims are based on norms, such as the international norm NEN-EN 13432:2000. However, industrial biogas plants operate at much shorter hydraulic retention times than mentioned in the norms. In this study, we investigated bioplastic degradation and biogas production after 21 days—industrial biogas plant operation times—and 35 days—maximum operation time for anaerobic digestion of bioplastics of norm NEN-EN 13432:2000. There was a large discrepancy between 3 and 5 weeks of anaerobic digestion, with biogas production going up with 43% (1F) up to 81% (2F) and 82% (3F) in that period (Figure 1). In addition, biogas production of bioplastic foils did not level off at 35 days (Figure 1), meaning that the biodegradation potential of the bioplastics was not fully used to produce biogas. In addition, nearly all bioplastics were retained at a 2 mm sieve. At industrial biogas plants, this would mean that the large bioplastic particles are sieved out of the digestate and either landfilled or incinerated. In addition, separation of bioplastic particles is an added step in the process, along with the risk of causing clogging and other operational challenges. Landfilling or incineration after anaerobic digestion are not worth time and money, but the biodegradation potential of plastics is also not used, and more GHGs are emitted [10,17]. Ironically, bioplastics were invented, amongst others, to replace fossil fuels and mitigate GHG emissions.

The biodegradation results show that at industrial biogas plants, the full potential of biodegradable plastics may not be reached. The microscopic results further showed that bioplastic is not disintegrated, but remains in its original physical form, and, consequently, that the claims of biodegradability on the packaging may not be (fully) achieved. For effective biodegradation of bioplastics at industrial biogas plants, future studies should focus on the discrepancy between international norms and industrial practice. This can be achieved by extending the operational times of biogas plants, as previous studies show higher degradation percentages of bioplastics at longer retention times, or by aligning the international norms on bioplastics to industrial biogas plants.

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