

Can anaerobic digestion be a suitable end-of-life scenario for biodegradable plastics? A critical review of the current situation, hurdles, and challenges

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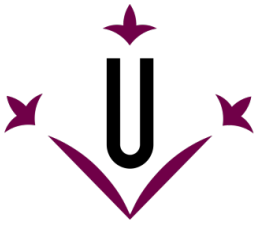
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Can anaerobic digestion be a suitable end-of-life scenario for biodegradable plastics? A critical review of the current situation, hurdles, and challenges

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Abstract:

Growing concern regarding non-biodegradable plastics and the impact of these materials on the environment has promoted interest in biodegradable plastics. The intensification of separate biowastes collection in most European countries has also contributed to the development of biodegradable plastics, and the subject of their end-of-life is becoming a key issue. To date, there has been relatively little research to evaluate the biodegradability of biodegradable plastics by anaerobic digestion (AD) compared to industrial and home composting. However, anaerobic digestion is a particularly promising strategy for treating biodegradable organic wastes in the context of circular waste management. This critical review aims to provide an in-depth update of anaerobic digestion of biodegradable plastics by providing a summary of the literature regarding process performances, parameters affecting biodegradability, the microorganisms involved, and some of the strategies (e.g., pretreatment, additives, and inoculum acclimation) used to enhance the degradation rate of biodegradable plastics. In addition, a critical section is dedicated to suggestions and recommendations for the development of biodegradable plastics sector and their treatment in anaerobic digestion.

Graphical abstract



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Abbreviations:

AD: Anaerobic Digestion

CSTR: Continuous Stirred-Tank Reactor

HRT: Hydraulic Retention Time

OFMSW: Organic Fraction of Municipal Solid Waste

PLA: poly(lactic acid)

PCL: poly(caprolactone)

PBAT: poly(butylene adipate-co-terephthalate)

PHAs: poly(hydroxyalkanoates)

PHBV: poly(3-hydroxybutyrate-co-3-hydroxyvalerate)

PBS: poly(butylene succinate)

TPS: thermoplastic starch

WF: wood flour

TS: Total solids

VS: Volatile solids

Highlights:

● Biodegradable plastics will play a key role in the future management of biowastes;

● Anaerobic digestion of biodegradable plastics performs better under thermophilic than under mesophilic conditions;

● Pretreatment can significantly enhance the biodegradation rate of biodegradable plastics by anaerobic digestion;

●The agronomic potential and safety of digestates treating biodegradable plastics streams have not yet been reported;

●There is currently a lack of norms and certifications for biodegradability in anaerobic digestion systems.

Keywords: anaerobic digestion, biodegradability, bioplastic, digestate, labeling, European norm, pretreatment

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1) Introduction

More than half a century ago, synthetic polymers started to be used in a broad range of applications, and plastics have become indispensable to modern-day life (Shah et al., 2008). Every year, 368 million tons of plastics are produced globally, and biodegradable plastics account for nearly 1% of the total production of plastics (Abraham et al., 2021; European Bioplastics, 2019; Plastics Europe, 2020). Astonishing amounts of plastics are released into the terrestrial and marine ecosystems as industrial waste products (Shah et al., 2008; Shimao, 2001). Indeed, due to insufficient recycling and poor end-of-life management, used plastics have become a major problem, resulting in a clear risk to the marine environment as well as to the safety of animals and humans (Emadian et al., 2017; Hegde et al., 2018; Karan et al., 2019). In 2014, over 250,000 tons of plastics were estimated to be floating in the sea (Eriksen et al., 2014). Geyer et al. (2017) estimated that almost 60% of all plastics ever produced, corresponding to 5 billion tons, have been discarded into the environment (natural or landfills). It is predicted that by 2030, the amount of plastic entering the world aquatic ecosystem could reach 90 Mt/year under the scenario where the current plastic production trend continues without improvements in the waste management system (Borrelle et al., 2020).

The vast majority of plastics are produced from non-renewable carbon sources (*e.g.*, petroleum derivatives), representing approximately 99% of the 2018 total plastic production (European Bioplastics, 2019). The growing awareness of policy makers and consumers regarding the finite availability of fossil fuels, combined with a desire to reduce the environmental impact, has led to a rethinking of plastic production, utilization, and its integration in our daily lives (Filho et al., 2021; Iwata, 2015). For these purposes, rational use of plastics is encouraged in Europe, and several single-use items made of non-biodegradable plastic are now banned in European countries. In parallel, several bioplastic initiatives have also been developed to address this problem, and they have been marketed as environmentally friendly solutions. The term bioplastics comprises (a) bio-based and/or (b) biodegradable plastics, although a bioplastic can also meet both of these criteria (Bátori et al., 2018; European Bioplastics, 2019; Pathak and Navneet, 2017; Ruggero et al., 2019). The bioplastics market is forecast to grow rapidly in the near future, with an estimated 36% increase by 2022 (European Bioplastics, 2019).

Among bioplastics, the biodegradable plastic sector is very promising and their development must occur in parallel with an in-depth consideration of end-of-life treatment systems and a global integration with organic waste management systems especially with the expansion of selective biowaste collection (Abraham et al., 2021; Bátori et al., 2018). Biodegradable plastics have the advantage of potentially being treated at their end-of-life by biological processes (composting or anaerobic digestion) (Balaguer et al., 2015; Bátori et al., 2018). Composting of biodegradable plastic has been well documented and is established at the International level, including analysis, assays, specifications standards, as well as labels certifying the biodegradability of a product under different composting conditions (home and industrial) (Iwata, 2015; Ruggero et al., 2019). To date, research carried out under anaerobic conditions has lagged relative to investigations of aerobic conditions, and it has been developed mostly at the lab scale, with scant data regarding continuous pilot- and industrial-scale processing (Ruggero et al., 2019).

Anaerobic digestion (AD) is a biological process whereby organic matter is transformed in the absence of oxygen into biogas (a mixture of carbon dioxide and methane) that can be further valorized by combined heat and power (CHP) production or by injection into the natural gas grid after an upgrading process (Miltner et al., 2017). The use of anaerobic digestion (AD) for biogas production remains widespread as a useful bioenergy production route due to the robustness of the process and its ability to process several organic wastes such as agricultural wastes, sludges, agro-food wastes, and organic municipal solid wastes (Meyer-Kohlstock et al., 2016). Due to favorable conditions, the number of AD plants has grown significantly in Europe, with 6 227 biogas plants in 2009 and 17 783 biogas plants at the end of 2017 (Scarlat et al., 2018; Wu et al., 2021). Furthermore, from an energy and environmental point of view (Bátori et al., 2018; Hermann et al., 2011; Piemonte, 2011; Rossi et al., 2015). For this purpose, AD of biodegradable plastics has attracted attention in the scientific community in the past few decades (Bátori et al., 2018; Greene, 2018; Narancic et al., 2018; Ruggero et al., 2019). Most of the biodegradable polymers that are biodegradable under industrial composting conditions are also biodegradable under AD conditions, albeit at a reduced rate (Abraham et al., 2021; Bátori et al., 2018; Bernat et al., 2021; Narancic et al., 2018). These polymers require more time to reach their final biodegradation level than the hydraulic retention time (HRT) usually applied in biogas plants (Narancic et al., 2018). Nonetheless, various

strategies can be used to enhance the biodegradation kinetics of these plastics. Three main strategies can be implemented before the AD process: pretreatment technologies, the incorporation of additives into the polymers (*e.g.*, fibers, enzymes, or calcium carbonate), and inoculum acclimation (Calabro et al., 2019; Ryan et al., 2017a, 2017b). Another important aspect when considering anaerobic digestion of biodegradable plastics is valorization of the digestate in agronomy as soil amendments/fertilizers. In this case, it is important to consider the safety of the digestates and the potential toxicity of residual plastics and microplastics on terrestrial and aquatic organisms (Fojt et al., 2020; Li et al., 2020; Shruti and Kutralam-Muniasamy, 2019; Zimmermann et al., 2020).

To help mitigate the environmental concerns associated with conventional plastics, a more effective synergy between the production of biodegradable plastics, their use, and their management in the organic recycling industry, particularly by anaerobic digestion, should be addressed in the future (Abraham et al., 2021; Bátori et al., 2018; Calabro et al., 2019). A number of interesting bibliographic reviews on the anaerobic digestion of biodegradable plastics have been published recently. The main topics of these papers are summarized in **Table 1**, with most of them covering specific aspects of the subject (Abraham et al., 2021; Bátori et al., 2018; Folino et al., 2020; Lagnet et al., 2020; Quecholac-Piña et al., 2020). In this review, a more global state-of-the-art is proposed, covering the various aspects of the end-of-life of biodegradable plastic wastes in anaerobic digesters (**Figure 1**). For this purpose, we first provide a brief description of the anaerobic digestion process, bioplastics and biodegradable plastics. Plastics biodegradability under mesophilic and thermophilic conditions, alone or in co-digestion, in batch, or in continuous assays is further described based on the data in the literature. Several pathways to enhance the biodegradability of plastics under anaerobic conditions are then presented, such as pretreatments, additive incorporation, and inoculum acclimation. In parallel, a specific section is dedicated to the developments required to make the introduction of biodegradable plastic wastes in anaerobic digesters possible: determination of the impact of plastic addition on the quality/safety of the digestate, and the establishment of international standards and labels to evaluate the biodegradability of a specific plastic by AD. Finally, a section is dedicated to suggestions and recommendations regarding the biodegradability of plastics and their integration into the waste management chain.

Table 1. The main focus area of recently published review papers on the anaerobic digestion of biodegradable plastics.

Figure 1. General overview of the main issues addressed in this bibliographic review.

2) Bioplastics and biodegradable plastics

The European Bioplastics Association defines bioplastics as *"plastics that are bio-based, biodegradable or both"* (European Bioplastics, 2019). The first concept, *i.e.*, "bio-based", relates to the origin of the materials used in the plastic production. A plastic can be classified as bio-based if it is wholly or partly derived from renewable carbon sources. In order to meet the requirements of the "OK bio-based" (TÜV Austria®) and "Din Geprüft bio-based" (DIN CERTCO) labels, at least 20 % of the carbon should be derived from renewable sources according to the EN 16785-1 or the ASTM D6866 standard test method (TÜV Austria, 2021; TÜV Rheinland, 2021). Bioplastics can be directly extracted from biomass (starch, cellulose, etc.), they can be chemically synthetized from bio-derived monomers (*e.g.*, PLA, bio-PE, bio-PET, bio-PP), or can be produced by microorganisms (*e.g.*, PHAs) (Avérous and Pollet, 2012; Song et al., 2011).

The other aspect of this definition is related to the end-of-life of the plastic and its biodegradable nature. A biodegradable plastic can be defined according to ISO 472 (2013) as a plastic that undergoes a significant degree of mineralization (conversion into water, CH_4, and/or CO_2) under specific environmental conditions due to the action of naturally occurring microorganisms in a given period.The biodegradation has to be measured by standard test methods appropriate to the plastic and to the application (ISO 472, 2013). Nonetheless, oxo-degradable plastics (*i.e.*, conventional plastics that contain specific additives designed to promote the oxidation of the material at its end-of-life) cannot be considered to be biodegradable plastics (Deconinck and De Wilde, 2013; Ettlinger et al., 2016). The pro-oxidant additives allow faster cleavage of the macromolecules induced by environmental factors (*e.g.,* UV light, heat, oxygen) (Dilkes-Hoffman et al., 2019; Sivan, 2011). However, the small size/molecular weight plastics that are produced are not necessarily subsequently mineralized (Deconinck and De Wilde, 2013; Ettlinger et al., 2016). The biodegradation of a plastic is tightly linked to the environmental

conditions in which it occurs. Currently, industrial composting is the favored organic valorization route for biodegradable plastics; thus, the polymers commonly referred to as biodegradable are certified under industrial composting conditions, *i.e.*, according to the EN 13432 standard.

Based on the bioplastic definition, there are three families of bioplastics: **1) *Bio-based and non-biodegradable plastics***, also called "drop-in", which share the same properties as their petrochemical counterparts (*e.g.*, bio-PE and PE, bio-PET and PET, bio-PP and PP), except that they are partially produced from renewable carbon sources and, therefore, help to reduce the dependency on fossil resources (Álvarez-Chávez et al., 2012; Bátori et al., 2018). They represented 44.5% of the global bioplastic production capacity in 2018 (***Figure 2***). **2) *Petrochemical and biodegradable plastics,*** such as PBAT, PBS, and PCL. They accounted for 19.1% of the global bioplastic production capacity in 2018 (***Figure 2***). **3) *Bio-based and biodegradable plastics,*** such as starch blends, PLA, and PHAs (accounting for 36.4% of the global bioplastic production capacity in 2018; ***Figure 2***). These have the benefit, compared to the two bioplastic families mentioned above, of reducing the fossil fuel dependency while also being suitable for an additional waste management option. Thus, the word "bioplastic" is ambiguous and can be confusing about its ability to biodegrade.

Commercially available plastics are often blends of different polymers in order to meet the intended technical requirements (Long and Chen, 2009). Polymers or co-polymers can also be compounded with organic or inorganic fillers to enhance certain properties, thereby contributing to a reduction in the cost of the plastic (Meereboer et al., 2020; Shahlari and Lee, 2012; Song et al., 2011). A wide range of additives are included in the plastics for various purposes. In 2014, these comprised, in decreasing quantities, plasticizers, flame-retardants, coupling agents and impact modifiers, antimicrobials, UV stabilizers, and antioxidants (Global Plastic Additives Market Size & Share - Industry Report, 2016). A plasticizer is a molecule, generally of low molecular mass, that is interposed between the polymer chains, thereby reducing the inter- and intra-molecular bonds and thus increasing the free volume between these chains. The addition of a plasticizer decreases the polymer's viscosity by promoting sliding of the chains relative to each other. At the same time, the crystallinity of the material decreases and its flexibility increases (Rahman and Brazel, 2004; Xiao et al., 2009). In the case of composites combining a polymer with an organic filler (*e.g.*, a vegetable filler added to mechanically reinforce the polymer), the addition of a coupling agent to the mixture is frequently used with the intention of improving the interface between the polymer and the filler (Muthuraj et al., 2015). These are either bifunctional products capable of reacting with the functional groups of the two phases and thereby resulting in the creation of covalent bonds between them, or amphiphilic compounds that have an affinity for each of the two phases (Mittal and Chaudhry, 2015).

The main biodegradable polymers, whether bio-based or based on fossil resources (**Table 2**), are listed below.

- **Thermoplastic starch (TPS),** Starch is one of the most abundant and cheapest biodegradable polymers as it represents the main energy reserve of plants (Kaseem et al., 2012). In plants, it is mainly stored in tubers and seeds. The main sources of starch are corn, wheat, potato, cassava, pea, and rice. Starch is also the main source of carbohydrates in the human diet. Chemically, it is a mixture of two homopolymers of α-D-glucose units, linked together by α-1,4 glycosidic bonds, *i.e.*, amylose (a linear or a very slightly branched macromolecule) and amylopectin (a branched macromolecule of glucose units based on α-1,6 bonds) (Kaseem et al., 2012). In plastics, starch is not used in its native form but in its plasticized form, which is obtained by the use of plasticizers. Thermoplastic starch (TPS) is a native starch that has been subjected to a thermomechanical treatment that allows thermoplastic transformation (Avérous and Pollet, 2012; Song et al., 2011). In most cases, water is used to make the starch a thermoplastic because water plays a dual role as a destructuring agent as well as an efficient plasticizer, due to its particularly low steric hindrance. However, water has a major drawback, namely its volatility. Over time, water migrates and evaporates from the material, promoting the phenomenon of starch retrogradation and thus alteration of its properties. Many other molecules of low molecular mass can, therefore, also be used, either as plasticizers or as destructuring agents. The main entities are glycerol (Chabrat, 2012); sorbitol (Li and Huneault, 2011); ethylene glycol and propylene glycol (Roz et al., 2006); amides including formamide, acetamide, and urea (Ma and Yu, 2004); and citric acid (Chabrat et al., 2012). Thermoplastic starch is, in most cases, blended with other polymers, *e.g.*, polyesters such as PLA, PBAT, PCL, PHAs, etc. (Bátori et al., 2018; Bulatović et al., 2021).

- **Poly(lactic acid) (PLA):** PLA is a linear aliphatic polyester made from renewable resources. It is synthetized by direct polycondensation of lactic acid or by ring-opening polymerization of lactide (Long and Chen, 2009). Lactic acid is commonly produced by fermentation of various biomasses (*e.g.*, corn, wheat, sugar cane, and sugar beet) (Song et al., 2011). As the carbon of lactic acid bearing the hydroxyl group is asymmetric, lactic acid can exist as two enantiomers (L or D). PLA can, therefore, have two different chemical structures depending on whether the chain of monomers is isotactic (L-PLA) or syndiotactic (D,L-PLA) (Pang et al., 2010). The stereoisomeric L/D ratio of the lactate monomers influences the properties of the PLA produced (*e.g.*, crystallinity, thermal and mechanical properties) (Pang et al., 2010; Zhang and Sun, 2005). Thus, the L-PLA obtained by polymerization of a single isomer has the capacity to crystallize while the D,L-PLA obtained from a mixture of the two isomers is amorphous (Iannace et al., 2014).
- **Poly(butylene adipate/terephthalate) (PBAT):** PBAT is an aromatic co-polyester made by co-polymerization (condensation reaction) of 1,4-butanediol with adipic and terephtalic acids (Avérous and Pollet, 2012; Long and Chen, 2009). PBAT is produced from non-renewable carbon resources. Its applications are diverse (Avérous and Pollet, 2012). With a low density and good resistance to humidity, stretching, and impact, it is suitable for applications such as rigid packaging for food or beauty products. Aliphatic aromatic co-polyesters, which contain small amounts of aromatic entities, are able to decompose. The breakdown of PBAT depends on the quantity of aromatic ester functions (Witt et al., 1996).
- **Poly(butylene succinate) (PBS):** PBS is a petroleum-based aliphatic polyester synthesized by polycondensation of 1,4-butanediol and succinic acid (Avérous and Pollet, 2012; Bátori et al., 2018). It is a soft and flexible semi-crystalline thermoplastic. Both building blocks (*i.e.*, 1,4-butanediol and succinic acid) can be produced from renewable raw materials such as starch (mainly from corn), glucose, and sucrose by fermentation, or from petrol. However, for the main commercial grades, the 1,4-butanediol block is of fossil origin, making these grades partially bio-based (approximately 50%). PBS is a very promising material because its mechanical properties are comparable to those of high-density polyethylene and isotactic polypropylene. Compared to PLA, it is much more flexible, and its melting point is much lower (approximately 115 °C). It can be used as a matrix polymer or in combination with other biodegradable polymers such as PLA (Su et al., 2019).
- **Poly(Ɛ-caprolactone) (PCL):** PCL is another fossil-based aliphatic polyester that is usually produced by ring-opening polymerization of Ɛ-caprolactone (Avérous and Pollet, 2012; Mohamed and Yusoh, 2015). Two types of PCL can be distinguished according to their molar mass. While the first group (molar masses of a few thousands of g/mol) is in a liquid form (used as precursors for polyurethanes, thinners for paints, or plasticizers for vinyl resins), the second type of PCL comprises molecules with higher molar masses (greater than 20,000 g/mol) (Jiang and Zhang, 2013). These latter PCL grades are semi-crystalline, and they exhibit interesting mechanical characteristics. They are generally used to modify the properties of other biodegradable plastics, including TPS (Arakawa and DeForest, 2017).
- **Polyhydroxyalkanoates (PHAs):** PHAs are a family of aliphatic polyesters produced from renewable carbon sources. They are synthesized (*i.e.*, naturally accumulated) by some microorganisms as energy and carbon reserves (Bugnicourt et al., 2014). Bioengineering methods that take advantage of this ability are used to produce commercial grades of PHAs (Bugnicourt et al., 2014; Song et al., 2011). PHAs are generally classified based on the number of carbon atoms in their monomer units: short-chain-length (scl) PHAs (or scl-PHAs) with 3-5 carbon atoms per monomer and medium-chain-length (mcl) PHAs (or mcl-PHAs) with 5-14 carbon atoms per monomer (Kynadi and Suchithra, 2014). Poly(3-hydroxybutyrate) (PHB) is the most widespread member of the PHAs family, followed by poly(3-hydroxyvalerate) (PHV) and their copolymer poly(3-hydroxybutyrate-cohydroxyvalerate) (PHBV) (Bugnicourt et al., 2014). PHB is a partially crystalline and biocompatible homopolymer, and it has mechanical properties close to those of isotactic PP, except for its elongation at break, which is lower (Thiré et al., 2006). Depending on their nutrition, bacteria can also produce other PHAs, e.g., PHBV (Albuquerque et al., 2011). PHBV is the second most common commercial PHA. The substituent group is randomly either a methylbutyrate or an ethylvalerate in the PHBV copolymer. The amount of valerate in the copolymer influences the final mechanical properties of PHBV. In particular, an increase in toughness and a decrease in modulus are observed with an increase in valerate content (Savenkova et

al., 2000). Indeed, lengthening of the aliphatic chain reduces the glass transition temperature and the degree of crystallinity (Chan et al., 2019). Depending on the composition, the PHBV material changes from a brittle and hard polymer to a flexible and elastic gum (Savenkova et al., 2000).

Figure 2. Global production capacities of bioplastics in 2019 (***Source:*** European Bioplastics, 2019).

3) Anaerobic digestion process

Anaerobic digestion (AD) is a biological process by which, in an oxygen-free environment, organic matter is converted into biogas (mainly composed of carbon dioxide and methane) and digestate (the non-degraded fraction, which is rich in nutrients). AD has turned out to be a promising method for the valorization of organic materials such as agricultural wastes (manure, crop residues, and winery wastes) (Monlau et al., 2013b), food wastes (Morales-Polo et al., 2018), and sewage sludges (Hanum et al., 2019). The biogas produced can be converted into heat and electricity through a cogeneration heat and power (CHP) system, or after biogas upgrading, biomethane can be introduced into the national gas grid or used as transport biofuel (Miltner et al., 2017; Sahota et al., 2018). The electricity produced can be sold providing economic benefits to the AD plant operator. The price of the electricity is dependent of the national policy and can vary among the various European countries (Monlau et al., 2012; Sambusiti et al., 2013). In parallel, digestate corresponding to the anaerobically non-degraded fraction composed of recalcitrant fibers (*e.g.*, lignin), and which is rich in mineralized nutrients, is also generated. To date, digestates have mostly been used at the farm scale as fertilizers or soil amenders (Monlau et al., 2015; Nkoa, 2014; Sheets et al., 2015). At the industrial scale, digestates are generally mechanically separated (belt press, sieve drum, screw press, sieve centrifuge, rotary screen, or decanter centrifuge) into liquid and solid fractions that are stored separately for ease of handling and transport (Monlau et al., 2015).

Anaerobic digesters are mainly operated at two temperature ranges, namely mesophilic (35-38 °C) or thermophilic (55-58 °C). There are three main full-scale reactor configurations. These are designed to treat feedstocks with different total solids (TS) contents. Upflow anaerobic sludge blanket (UASB) or anaerobic fluidized bed technologies are used to treat liquid feedstocks (< 3% TS), especially from urban wastewater and agro-food sectors. Continuous stirred-tank reactors (CSTR) are used to process feedstocks with a TS content between 8 and 15%. Solid-state AD (SS-AD) are designed for feedstocks with TS contents higher than 15% and are classified as dry batch AD and dry plug-flow AD. For the treatment of organic wastes, dry technologies currently represent approximately 75% of all AD plants in Europe (Mattheeuws, 2015). Unlike CSTR and SS-AD reactors, UASB reactors cannot be considered to be a consistent option for the treatment of solid organic wastes and biodegradable plastics as they are designed for liquid effluents.

To evaluate the biodegradability of organic products, two tests can generally be applied to assess anaerobic performances: Biochemical Methane Potential (BMP) test and continuous (or semi-continuous) pilot-scale experiments. The BMP test is a procedure developed to determine the methane production of a given organic substrate during its anaerobic digestion at a lab scale. It is a reliable method for obtaining the extent and rate of organic matter conversion into methane. Pilot-scale experiments use more realistic conditions than the BMP test: the feeding is continuous or semi-continuous, and several parameters (biological, operational, performances) are monitored during the assay. Pilot experiments provide precious insights regarding the process performance and stability over a long period of time. An important parameter that can influence AD performances is the C/N ratio of the feedstock. Hawkes (1980) suggested a carbon to nitrogen ratio for anaerobic digestion ranging from 20:1 to 30:1 for preventing both nutrient limitation and ammonia toxicity. Protein-rich wastes such as food wastes or municipal sludges have C/N ratios ranging from 6:1 to 16:1. By contrast, most biodegradable plastics contain carbon but no nitrogen. Thus, co-digestion of biodegradable plastics with proteinaceous substrates can increase the C/N ratio to the suggested values and result in a more stable process (Benn and Zitomer, 2018; Rabii et al., 2019; Rajagopal et al., 2013).

Finally, the extent of biodegradation of a substrate can be estimated by comparison of the experimental methane production to the theoretical methane production (Eq.1). The theoretical methane production can be calculated using the Buswell formula (Eq. 2). The estimation of the biodegradation by such method can rarely reach 100%; indeed a fraction of the substrate's carbon is not converted into biogas and is assimilated by the microbial biomass during the AD process (Shah et al., 2008).

$$\text{Eq.1: Biodegradation (\%)} = \frac{\text{Experimental methane production}}{\text{Theoretical methane production}}$$

Eq 2: (Boyle, 1976; Buswell and Mueller, 1952)

$$\text{Theoretical methane production (L } CH_4 \text{ g}^{-1} \text{ CxHyOzNnSs)}^* = \frac{22.4 \times \left(\frac{x}{2} + \frac{y}{8} - \frac{z}{4} - \frac{3n}{8} - \frac{s}{4}\right)}{12x + y + 16z + 14n + 32s}$$

*at standard temperature and pressure (0°C, 1 atm)

4) Anaerobic digestion of biodegradable plastics (Tables 3 & 4)

4.1 Batch anaerobic digestion by BMP

Most of the studies to date have investigated the biodegradability of biodegradable plastics in BMP. Tables 3 and 4 present the main results obtained under both mesophilic (35-38 °C) and thermophilic (55-58 °C) conditions. A detailed survey is provided below, first for mesophilic and then thermophilic conditions. It is important to keep in mind that the experimental conditions differ between studies; that the exact formulation of the tested plastics is not precisely known, and that the properties of different plastics made from the same polymer can differ significantly. Therefore, this implies that there is a degree of variability in the results obtained in the various studies.

a) Mesophilic conditions

First of all, the anaerobic digestion performances of biodegradable plastics under mesophilic conditions are presented in **Table 3**. Poly(3-hydroxybutyrate) is the most widespread member of the polyhydroxyalkanoates family. Complete or near-complete biodegradation of PHB samples in a short time period have been reported for mesophilic digesters. For example, Abou-Zeid et al. (2001) observed that a 19 mm film made of PHB Biopol® BX G08 (ICI, United Kingdom) was fully converted into methane after only 9 days of incubation in various microbial inocula. PHB is a very promising polymer given its ability to be biodegraded in non-harsh environments such as mesophilic anaerobic digestion, home composting, soil, etc. (Narancic et al., 2018). The short time needed to fully biodegrade PHB makes it compatible with the conventional hydraulic retention time used in industrial anaerobic digestion plants (Narancic et al., 2018). However, the methane conversion differed significantly depending on the grade of PHB used. Benn and Zitomer, (2018) found near-complete biodegradation of two PHB grades, Mirel M2100 (Metabolix) and methane-derived PHB from Mango Materials, while only 50 to 59% of ENMAT Y3000 (TianAn) and Mirel F1006 (Metabolix) were degraded at the same time. Poly(3-hydroxybutyrate-co-hydroxyvalerate) (PHBV) is a co-polymer of poly(3-hydroxyvalerate) (PHV) and poly(3-hydroxybutyrate), and it is also one of the main members of the PHA family. Similar to PHB, PHBV exhibited a very good level of biodegradation in a short time (Budwill et al., 1992; Day et al., 1994; Reischwitz et al., 1997; Ryan et al., 2017b, 2017a, Shin et al., 1997). For example, Reischwitz et al. (1997) reported a biodegradation level of 95% in 30 days for PHBV powder (420 µm) with 8.4% hydroxyvalerate (HV). A lower conversion into methane was reported by Abou-Zeid et al. (2004, 2001) and Day et al. (1994), between 29 and 55% in 40 and 42 days, respectively. Nunziato et al. (2018) reported a low degree of biodegradation for polyhydroxyoctanoate (PHO), which is a medium-chain PHA, with only 12% of the material being converted into methane after 56 days in a mesophilic digester. On the other hand, Federle et al. (2002) reported near-complete biodegradation (88%) for PHBO, a co-polymer of PHB and PHO (10% PHO), after 60 days of anaerobic digestion.

PLA has been one of the most investigated biodegradable plastics to date. According to Vargas et al. (2009) and Vasmara and Marchetti, (2016), rigid pieces of PLA (3 mm and < 1 cm^2) did not biodegrade in 60 and 90 days, respectively. Similar observations have been reported for smaller pieces of PLA. For example, 0.15 mm particles and 20 x 40 mm film did not exhibit any significant biodegradation in 40 and 100 days, respectively (Benn and Zitomer, 2018; Shin et al., 1997). However, other authors have reported methane production during the digestion of PLA at 35 ± 2 °C (Day et al., 1994; Greene, 2018; Itävaara et al., 2002; Massardier-Nageotte et al., 2006; Yagi et al., 2014). For Day et al. (1994), Greene, (2018) and Massardier-Nageotte et al. (2006), minor biodegradation levels were observed, with between 10 and 23% of the PLA being converted into methane in 20 to 40 days. Yagi et al., (2014) investigated anaerobic digestion of PLA ground to 125-250 µm over a long period of time (277 days). At the end of the test, the PLA was biodegraded between 29 and 49% (depending on the run) but the methane production did not reach a plateau as methane production was still increasing. They explained the low biodegradation rate of PLA by the fact that the bacteria present in the mesophilic digesters did not have the ability to biodegrade higher molecular weight PLA. The microorganisms were only able to use PLA after a reduction of its molecular weight caused by a random hydrolytic chain scission of the ester linkages (Ali Akbari

Ghavimi et al., 2015). Bernat et al. (2021) also performed mesophilic anaerobic digestion of PLA over a long period of time (280 days). After 40 days of lag phase, there were two phases of constant biogas production. Firstly, 1.4 L kg^{-1} VS d^{-1} of biogas was produced between the 40^{th} and 90^{th} day, and then, between the 90^{th} and 280^{th} day, the biogas production increased to 2.6 L kg^{-1} VS day^{-1}. Finally, after 280 days, the biogas production reached 66% of the theoretical value, although the plateau phase was nonetheless not reached.

Thermoplastic starch exhibited a very high level of biodegradation in a short time in mesophilic digesters, according to Narancic et al. (2018). Indeed, TPS from BIOTEC (TPS Bioplast®) was fully biodegraded in 56 days at 35 °C. However, TPS is rarely used on its own, instead, it is usually blended with other biodegradable polymers. PCL is often used in blends with TPS (Ali Akbari Ghavimi et al., 2015). Nunziato et al., (2018) showed that the addition of TPS increased the biodegradation rate of PCL in mesophilic and thermophilic anaerobic digesters. The biodegradation of PCL was reported to be very slow and in all cases, with the degradation level between 0 and 22% at the end of the tests (Abou-Zeid et al., 2004, 2001; Day et al., 1994; Federle et al., 2002; Massardier-Nageotte et al., 2006; Narancic et al., 2018; Nunziato et al., 2018; Puechner et al., 1995). Yagi et al. (2014) performed mesophilic anaerobic digestion tests on PCL powder (125-250 µm) over a very long period of time (277 days). They found a very low biodegradation rate. Only 3 to 22% of the PCL was converted to methane, and they assumed that the same biodegradation mechanisms as with PLA were involved. The microorganisms were only able to degrade lower molecular weight PCL resulting from the random hydrolytic chain scission of the ester linkages.

Other plastics that were found to be biodegradable under industrial composting conditions such as poly(butylene succinate) and poly(butylene adipate-co-terephthalate) did not undergo significant biodegradation in mesophilic anaerobic digesters (Cho et al., 2011; Massardier-Nageotte et al., 2006; Narancic et al., 2018; Shin et al., 1997; Svoboda et al., 2018). PBS was not degraded at all, even with the very long digestion time (277 days) used by Yagi et al. (2014). According to Shin et al. (1997), a co-polymer of PBS and polyethylene (PBES) showed no sign of biodegradation after 100 days in anaerobic media. The biodegradability of a number of other minor biodegradable plastic was also measured. Calabro et al. (2019), Puechner et al. (1995) ,and Shin et al. (1997) digested plastics derived from cellulose. Calabro et al. (2019) and Shin et al. (1997) found a good degree of methane conversion of the polymers (310 L CH_4 kg^{-1} VS and 85% in 44 and 20 days, respectively) while Puechner et al. (1995) observed a relatively low degree of biodegradation (22% in 60 days) for cellulose acetate (Bioceta®, Mazzucchelli, Italy). Mesophilic digestion of butanediol/adipic acid and butanediol/adipic acid/terephthalic acid resulted in a low level of methane conversion in 42 days of between 1.1 and 10% (Abou-Zeid et al., 2004).

To date, most of the data available in the literature has been in regard to the biodegradability of the main biodegradable polymers individually. However, different polymers are often blended in order to meet the intended technical requirements (Long and Chen, 2009) and there is a paucity of data available in the literature regarding the biodegradability of commercial plastic blends (*e.g.*, Mater-Bi®, Ecovio®) in anaerobic digestion. For example, in the case of Mater-Bi® (Novamont, Italy), which is the most studied biodegradable blend in AD, the composition of the blend has not been clearly established. Mater-Bi® is a family of compounds based on TPS and other polymers such as cellulose acetate, poly(vinyl alcohol), poly(Ɛ-caprolactone), and poly(butylene adipate-co-terephthalate) (Aldas et al., 2020). Biodegradation of Mater-Bi® was reported to be relatively limited in mesophilic AD, with between 0 and 32% of the material being converted into methane over a period of 15-80 days (Calabro et al., 2019; Day et al., 1994; Massardier-Nageotte et al., 2006; Puechner et al., 1995; Scandola et al., 1998; Vasmara and Marchetti, 2016). Cazaudehore et al. (2021) explored AD of three commercial coffee capsules composed of biodegradable blends (Vegemat® from Vegeplast, Ecovio® from BASF, and Mater-Bi® from Novamont). After 100 days, the plateau phase of biodegradation was not reached for any of those blends; they only underwent 12 to 20% biodegradation. Recently, Dolci et al. (2022) have investigated the methane potential of four Mater-Bi® bags and biodegradability ranged from 71% to 93% in less than 56 days.

Finally, the degradation of non-commercial plastic blends, produced at a laboratory-scale, has, however, been investigated in several scientific publications (Guo et al., 2011; Lee et al., 2016; Narancic et al., 2018; Russo et al., 2009). Guo et al. (2011) examined anaerobic digestion of different PVOH/starch blends produced from various starch origins (from wheat, maize, and potato). The different blends were converted into methane between 58 and 62% in a relatively short period of time (5-6 days). Similarly, Russo et al. (2009) investigated the degradation of TPS/PVOH blends with different polymer contents (90/10, 75/25, 50/50, and 0/100). The PVOH content significantly affected the rate of the starch biodegradation. The methane production was higher for the blends with higher TPS contents. Narancic et al. (2018) reported a synergistic effect of blending two biodegradable polymers, with blends having higher methane production levels in some cases than individual polymers. This

better biodegradation of the blends could be explained by the lower crystallinity of the blend compared to the individual polymers (Narancic et al., 2018). In addition, the blend could have a better fragmentation capacity like the plastic-fiber composites, which would increase the surface area exposed to hydrolysis and thus the biodegradation rate (Ryan et al., 2017b).

b) Thermophilic conditions

The degradation of biodegradable plastics under thermophilic conditions has been studied much less than under mesophilic conditions, with fewer data available in the scientific literature. However, the operating conditions and performances of these tests are listed in **Table 4**.

Only three studies to date have described the digestion of PHB in thermophilic digesters (around 55 °C). Itävaara et al. (2002) and Yagi et al. (2013) found a very high level of biodegradation (between 73 and 88%) in a short time (between 18 and 20 days). Narancic et al. (2018) also noted near-complete mineralization of PHB but, strangely, reported a very long digestion period (127 days). No data regarding PHBV degradation by thermophilic digestion could be found in the scientific literature. As with mesophilic conditions, PHO was weakly degraded (6%) under thermophilic conditions in 50 days (Nunziato et al., 2018).

The methane conversion of PLA under thermophilic conditions was more effective than under mesophilic conditions. Hegde et al. (2018), Narancic et al. (2018), Šmejkalová et al. (2016) and Yagi et al. (2013, 2009) reported a high level of biodegradation of 82 to 90% in a mean digestion time of 90 days. Other authors have found a lower level of biodegradation, between 40 and 60%, with a similar timeframe for the digestion (Itävaara et al., 2002; Tseng et al., 2019; Vargas et al., 2009; Vasmara and Marchetti, 2016). However, it should be noted that the biodegradation levels reported by Itävaara et al. (2002) and Vargas et al. (2009) (60% and 40%, respectively) were not the final biodegradation levels, as the BMP tests were stopped before they reached the plateau of methane production. Šmejkalová et al. (2016) showed that a decrease in the molecular weight of PLA had a positive effect on the biodegradation kinetics. Shreshta et al. (2020) assessed thermophilic degradation of rigid pieces of PLA (1 x 1, 2 x 2, and 3 x 3 cm) and observed negligible methane production of approximately 10 to 30 L CH_4 kg^{-1} VS.

Thermoplastic starch was found to be readily biodegradable by thermophilic anaerobic digestion, with (Camacho-Muñoz et al. (2020) and Nunziato et al. (2018) observing a high level of biodegradation (between 77 and 92%) in a short period of time (30 days) for a 70% starch and 30% glycerol polymer. Narancic et al. (2018) recorded a biodegradation level of 81 % after 127 days in a solid-state anaerobic digestion system at 52 °C.

Near-complete mineralization of PCL (87-92%) was reported in thermophilic reactors in a time period of 45 to 127 days (Narancic et al., 2018; Yagi et al., 2013, 2009). Šmejkalová et al. (2016) tested the biodegradation of PCLs with different molecular weights (from 13 kg mol^{-1} to 58.1 kg mol^{-1}) in the form of a film or powder, and an increase in the molecular weight was associated with a reduction of the biodegradation kinetics. It took 140 days to reach the ultimate methane production of the higher molecular weight PCLs, while 70 days were required for the lower molecular weight PCLs. Only a small difference in the biodegradation level (54-60%) was noted for the different PCLs. Interestingly, Nunziato et al. (2018) found a low level of conversion into methane (11.3%) of PCL films cut into 1-cm square pieces after 30 days.

Most of the publications to date regarding the degradation of biodegradable plastics under thermophilic conditions have focused on PLA, PHB, or PCL. Only one scientific publication has explored the thermophilic degradation of PBAT (Svoboda et al., 2018). Svoboda et al. (2018) reported a very low level of PBAT biodegradation (8.3%) in 126 days at 55 °C. However, they observed a significant reduction in the molecular weight of PBAT from 93 000 to 9430 g mol^{-1} (Svoboda et al., 2018). Similarly, only three publications have presented data regarding the degradation of PBS, which was weakly mineralized, between 1 and 22%, over a period of 90-120 days (Dvorackova et al., 2015; Narancic et al., 2018; Yagi et al., 2013).

As with mesophilic conditions, Mater-Bi® (Novamont) is one of the few commercial grades that has been investigated in terms of biodegradation in thermophilic anaerobic digesters. Thermophilic digestion of small pieces (< 1 mm) of plastic bags made of Mater-Bi® resulted in a 55% conversion into methane over a period of 90 days (Vasmara and Marchetti, 2016). Similarly, Calabro et al. (2019) digested whole plastic bags made of Mater-Bi®, and reported a methane potential of 186 L CH_4 kg^{-1}VS in 30 days. Similarly, Cazaudehore et al. (2021) performed thermophilic anaerobic digestion on three types of commercial coffee capsules composed of biodegradable plastic blends (*i.e.*, Mater-Bi®, Ecovio®, and Vegemat®). After 100 days of digestion, a plateau phase was reached, and the biodegradation rate varied between 49% and 69% of the blends. In parallel,

anaerobic digestion under thermophilic conditions of non-commercial blends produced on a laboratory scale has been investigated (Narancic et al., 2018). A synergistic effect of blending biodegradable polymers was observed in the sense that the methane production of the plastic blends was reported to be equivalent or higher than the methane production of the individual polymers. For example, PCL−TPS (70/30) and PHB−PCL (60/40) blends produced 37% and 18% more methane, respectively, than expected from the performance of the individual polymers during AD.

c) Summary of the mesophilic and thermophilic AD performances

The biodegradability performances observed in the literature of the main biodegradable polymers available on the market (*i.e.,* PHB, PHBV, PLA, PCL, PBS, PBAT, and TPS) are summarized in **Figure 3.** The data are represented for both mesophilic and thermophilic conditions, with the number of experiments and studies reported. Not all biodegradable plastics have received the same degree of attention, as the majority of studies have focused on mesophilic degradation of PHB, PHBV, PCL, and PLA, and on thermophilic degradation of PLA. Overall, anaerobic digestion of biodegradable plastics under thermophilic conditions is more efficient than under mesophilic conditions (Nunziato et al., 2018; Vargas et al., 2009; Vasmara and Marchetti, 2016). For example, the degradation plateau of PCL and PLA is generally reached in less than 100 days under thermophilic condition, while this same plateau does not occur in less than 300 days under mesophilic conditions (Bernat et al., 2021; Yagi et al., 2014, 2013). According to these observations, biodegradable polymers can be classified into three categories based on their degradation performances and duration time in mesophilic and thermophilic AD (**Figure 3 and Figure 4**).

a) Polymers that are readily biodegradable. These reach complete or near-complete biodegradation in a relatively short period of time (10-50 days) under both mesophilic and thermophilic anaerobic digestion conditions. This is the case for PHB and TPS. It can be assumed that PHBV also belongs in this category, despite the absence of data for thermophilic conditions, in light of its rapid degradation (31-95% in approximately 35 days) under mesophilic conditions. These polymers could potentially be processed in biogas plants without increasing the hydraulic retention time used at the industrial scale (generally approximately 30-50 days for system treating biowastes).
b) Polymers that are biodegraded very slowly under mesophilic conditions and much faster under thermophilic conditions. This is the case for PLA and PCL. It was previously demonstrated that PLA and PCL exhibited low levels of biodegradation under mesophilic conditions (over a period exceeding 280 days) (Bernat et al., 2021; Yagi et al., 2014), which is much higher than the typical retention time observed in industrial biogas plants (around 20-50 days for biowastes, 50-120 days for agricultural wastes). By contrast, under thermophilic conditions, the hydrolysis is enhanced, resulting in much higher degradation rates. This has been documented very well by Bernat et al. (2021), who reported a biogas production rate under mesophilic conditions of between 1.4 and 2.6 L kg^{-1}VS d^{-1} and under thermophilic conditions of around 33 L kg^{-1} VS d^{-1}. Under mesophilic conditions, hydrolysis is the limiting step, and pretreatment strategies are a promising way to enhance degradation under mesophilic conditions (Brémond et al., 2018; Carrere et al., 2016).
c) The polymers that are biodegradable under industrial compositing conditions according to the EN 13432 standard and poorly or not at all biodegraded by either mesophilic or thermophilic AD. This is the case for PBAT and PBS. The operational conditions and particularly the microorganism diversity between anaerobic and aerobic processes can probably explain such observations.

Figure 3: Biodegradation (%) reached at the end of the BMP tests under mesophilic and thermophilic condition. The number of experiments (noted exp.), and the number of studies per biodegradable polymers are also presented. White circles correspond to each experiment reported. The Figure is based on data provided in Table 3 and Table 4.

Figure 4. Mean biodegradation (%) vs mean time of incubation (days) for the various data on biodegradable plastics found in literature for both mesophilic and thermophilic conditions. The Figure is based on data provided in Table 3 and Table 4.

4.2 Pilot-scale and full-scale experiments

Although BMP is a reliable, quick, and powerful tool to evaluate the biodegradability of biodegradable plastics, it cannot be used to simulate the future performances of industrial biogas plants. Indeed, biodegradation tests in BMP do not sufficiently reflect the industrial process and this can lead to overestimation of the biodegradation level. For this, pilot experiments with feeding under continuous or semi-continuous conditions are necessary. Pilot experiments provide valuable insights regarding process performances and stability over long periods of time. To date, few studies have investigated the anaerobic digestion of biodegradable plastics on a pilot scale (Benn and Zitomer, 2018; Venkiteshwaran et al., 2019; Zhang et al., 2018, Dolci et al., 2022). Benn and Zitomer, (2018) performed a continuous anaerobic co-digestion experiment on untreated and pretreated PHB (ENMAT™ Y3000 and Mirel™ F1006). Pretreatment consisted of hydrolysis at pH 1 and 55 °C for 24 or 48 h. Eight continuously stirred tank reactors (CSTR) with 2.5 L working volumes were operated at 35.7 °C. Firstly, these reactors were fed for 115 days with synthetic municipal primary sludge (a mixture of dog food, basal nutrients, and alkaline media) at an organic loading rate of 3.6 g COD d^{-1} L^{-1}. The reactors were then fed, in duplicate, with a mixture of synthetic municipal primary sludge and PHB samples (two PHB grades, pretreated and untreated), with plastics corresponding to 20% of the initial organic loading rate. When plastics were co-digested, the methane production increased by 17% compared to synthetic sludge alone. Degradation of the untreated PHB was estimated to be between 78 and 92%. Pretreatment resulted in a 5% increase in the conversion of PHB into methane. Synergistic effects of co-digestion of biodegradable plastics with food wastes or pig slurry have also been reported elsewhere (Hegde et al., 2018; Vasmara and Marchetti, 2016). The addition of biodegradable plastics, which are substrates with a high carbon content, can help adjust the carbon-nitrogen ratio to the recommended values and prevent both nutrient limitation and ammonia toxicity (Benn and Zitomer, 2018; Esposito et al., 2012; Hawkes, 1980b). Similarly, Zhang et al., 2018, looked at anaerobic biodegradation of nine different biodegradable plastics (according to EN 13432 standard) in batch-scale and semi-continuous pilot co-digestions. Eight of the plastics (three cellulose-based films, one cellulose diacetate film, two starch-based films, and one PLA film) were digested in the form of 1 x 1 cm square films, while a PLA blend was digested in pellet form. The feedstock in the semi-continuous trial was synthetic food waste, cardboard packaging, and bioplastics at volatile solids (VS) ratios of 80:18:2. The digesters exhibited stable performances and there was no evidence throughout the 144-day experimental period of potential inhibition of the feeding with biodegradable plastics. Cellulose-based films exhibited a high weight loss of 57.4 to 93.4% during the test, whereas starch-based films only lost a small part of their weight ranging from 2.1 to 7.9%. A PLA film lost 57.5% of its mass, while PLA pellets only lost 3.1% of their mass, thus demonstrating the impact of the initial morphology of the polymer. In parallel,, Wang et al. (2012) investigated co-digestion of a mixture of kitchen garbage and PLA in two different reactor configurations. A two-phase anaerobic system consisting of a hyperthermophilic (80 °C) reactor and a thermophilic (55 °C) reactor was compared with a single-phase thermophilic reactor (55 °C). Two types of plastic bags (100% PLA and 70% PLA) were used as pieces that were 2 x 2 x 0.1 mm in size. The methane conversion ratios of the two-phase systems were 82.9% and 80.8%, respectively, which was higher than the 70.1% conversion ratio of the single-phase system (Wang et al., 2011). Dolci et al., (2022) have also recently investigated the semi-continuous co-digestion tests of two MaterBi® biodegradable bags together with food wastes (11/91 % in term of DCO). Low degradability was observed as regards bioplastic bags residues, the overall mass of undigested pieces resulted equal to 93% (bag 1) and 69% (bag 2) of the inserted weight suggesting that an aerobic composting phase is necessary to achieve satisfactory biodegradation yield.

While the compostability of biodegradable plastics at an industrial scale has been well documented, only one test has been carried out at the European level on real biogas plants (Kern et al., 2018, European Bioplastics, 2015). Recently, Kern et al. (2018) performed real tests on various biogas plants on Mater-Bi® resin (Novamont). The biodegradation was performed in four German AD biogas plants: two discontinuous batch processes operated at thermophilic or mesophilic temperature and two horizontal plug-flow technologies operated at thermophilic temperature. All of the technologies tested resulted in a reduction of the biodegradable plastics content in terms of dry matter by 40% to 58%, except for the mesophilic process, for which no significant degradation was observed. For all the AD plants, the process was coupled with a composting process followed by maturation. After maturation of the compost, full degradation of the plastics in terms of dry matter was measured.

4.3 Microbial communities involved in biodegradable plastics AD:

The microorganisms involved in the AD process have undergone intensive study in recent years (Azizi et al., 2016; Castellano-Hinojosa et al., 2018; De Vrieze et al., 2018; Guo et al., 2015; Levén et al., 2007; Li et al., 2015; Liu et al., 2018; Sundberg et al., 2013). However, little is known about the specific microorganisms involved in AD of biodegradable plastics. In the reviews by Shah et al. (2014, 2008), Emadian et al. (2017), and Pathak and Navneet (2017), several microorganisms were reported to degrade biodegradable plastics, although most of them were not from an anoxic environnement. Yagi and co-workers have produced several publications regarding the detection of microorganisms that participate in anaerobic digestion of biodegradable plastics. For this, they performed denaturing gradient gel electrophoresis of the 16S rDNA amplicons (RT-PCR-DGGE). In their first two papers, they did not successfully identify the microorganisms responsible for thermophilic digestion of PLA (Yagi et al., 2011, 2010). However, they highlighted that a number of the microorganisms participating in anaerobic digestion of cellulose and PLA at 55 °C were different. Subsequently, Yagi and co-workers carried out the same experiment under mesophilic and thermophilic conditions for PLA, PCL, and PHB (Yagi et al., 2014, 2013). This time, they successfully identified several microorganisms involved in the anaerobic digestion process of these polymers, but their precise involvement in the process remains unknown. Under mesophilic conditions, *Arcobacter thereius* participated in the degradation of PHB and PCL, while *Xanthomonadaceae bacterium* and *Mesorhizobium* sp. participated in the digestion of PLA. Under thermophilic conditions, they successfully identified *Bacillus infernus* and *Propionibacterium* as microorganisms participating in the anaerobic degradation of the three biodegradable plastics. They then found that *Peptococcaceae bacterium Ri 50*, *Bacteroides plebeus,* and *Catenobacterium mitsuokai* were specific to anaerobic digestion of PHB. Similarly, *Ureibaccillus* was specific to PLA digestion. Venkiteshwaran et al. (2019) examined the microbial community shift during anaerobic co-digestion of PHB and synthetic primary sludge (dog food and basal nutrients) by Illumina sequencing. No previously known PHB degraders were observed in the co-digesters. OTUs associated with *Deferribacter*, *Geobacter*, *Kosmotoga*, and *Ruminococcus* were found to correlate positively with increased methane production resulting from PHB co-digestion. Peng et al. (2022) examined the microbial populations during the digestion and the co-digestion of PLA/PBAT blend with food waste under both mesophilic and thermophilic condition. Under mesophilic condition, no significant differences were noted between microbial communities from reactors fed with or without biodegradable plastics. However, under thermophilic conditions, the three genera: Clostridium (sensu stricto), Streptococcus and Caldicoprobacter were highlighted for being responsible for the difference between communities from the reactor fed with a blend of PLA/PBAT and other reactors (i.e., blank reactor, reactor fed with food waste and the reactor performing co-digestion of food waste and PLA/PBAT) (Peng et al., 2022). Tseng et al. (2019) examined the microbial populations involved in anaerobic digestion of PLA under thermophilic conditions by PCR-DGGE. They pointed out the importance of the genus *Tepidimicrobium* as a key genus in the anaerobic degradation of PLA. Subsequently, (Tseng et al., 2020) isolated and characterized a strain of *T. xylanilyticum* from this digester. They also found that the physicochemical depolymerization of PLA to lactate was inhibited by the accumulation of lactate. *T. xylanilyticum* promoted PLA degradation by scavenging the lactate produced, thereby providing methanogens with CO_2, H_2, and acetate (Tseng et al., 2020). More recently, Cazaudehore et al. (2021) identified a member of the *Tepidimicrobium* genus as key microorganisms involved in thermophilic anaerobic digestion of three coffee capsules composed of biodegradable plastics (*i.e.*, Mater-Bi®, Ecovio®, and Vegemat®). These coffee capsules are made of plastic blends and their exact composition is not known, so the genus *Tepidimicrobium* may be involved in anaerobic digestion of polymers other than PLA. More studies on *Tepidimicrobium* are needed to gain a better understanding of their involvement in the biodegradation of other biodegradable polymers. Unfortunately, Cazaudehore et al. (2021) were unable to identify the key microorganisms involved in mesophilic anaerobic digestion of these biodegradable blends, probably due to the low efficiency of the mesophilic process on such biodegradable plastics blends.

4.4 Factors influencing the anaerobic biodegradability of biodegradable plastics

The biodegradation of polymers is generally summarized as occurring in three main stages, during which a combination of biological and chemical mechanisms are involved. In the first step, called deterioration or biodeterioration, the physical and the chemical properties of the plastic are altered. Then, during the fragmentation or disintegration step, the polymer is cleaved into a simpler form. Finally, the fragmented plastic is assimilated and degraded by microorganisms (Abraham et al., 2021; Artham and Doble, 2008; Meereboer et al., 2020). Abiotic (*i.e.,* pH, temperature, and moisture content) and biotic (*i.e.*, the presence of specific

microorganisms or enzymes) parameters have a significant impact on biodegradation performances. In parallel, several factors related to the physical and the chemical properties of the polymer can influence the biodegradation rate (Ahmed et al., 2018; Artham and Doble, 2008). Parameters associated with the surface condition (*e.g*., surface area, morphology, and hydrophilic and hydrophobic properties), the first-order structures (*e.g*., chemical structures, molecular weight, and molecular weight distribution), and the higher-order structures of polymers (*e.g*., glass transition temperature, melting temperature, modulus of elasticity, crystallinity, and crystal structure) have a pronounced impact on the biodegradation process (Tokiwa et al., 2009). Similarly, the exposition to specific condition before the introduction in an anaerobic digestion reactor has been investigated and will be further discussed in the next section. Among the various biotic parameters, the specific microorganisms present in the inoculum can influence the rate of biodegradation. The impact of the inoculum origin, composition, and potential acclimation will be discussed more in-depth in Section 5.3. The impacts of some of factors influencing the anaerobic biodegradability have been investigated and reported in the literature.

Among them, an increase in the temperature of the anaerobic digestion process promotes the conversion of biodegradable polymers into methane (Nunziato et al., 2018; Vargas et al., 2009; Vasmara and Marchetti, 2016; Yagi et al., 2013, 2012). This is demonstrated by the fact that thermophilic AD provides better results than mesophilic AD. The mobility of the polymeric chains are increased with temperature, above the glass transition temperatures the disorganization of the chain facilitate the accessibility to chemical and biological degradation (Lucas et al., 2008; Maity et al., 2021). Moreover, faster biochemical reaction rate are found at higher temperatures (Gebreeyessus and Jenicek, 2016). Shi and Palfery, (2010) have highlighted the impact of the glass transition temperature on the anaerobic biodegradability of PLA. The glass transition temperature of PLA was reduced from 60.7 °C to 49.4 °C by the use of different plasticizers. Both the original and the modified PLA were then digested at 50 °C. Interestingly, the time to fully degrade the modified PLA was approximately 5 months, while 9 months were required for the untreated PLA. These authors have assumed that when the degradation temperature is higher than the glass transition temperature of the plastic, the amorphous part of the polymer becomes readily accessible to microorganisms, which then accelerates biodegradation of the plastic (Shi and Palfery, 2010). Ryan et al. (2016) and Yagi et al. (2009) investigated the impact of size reduction (increase in the surface area) of rigid plastics composed of PHBV and PCL. Both studies found that a smaller particle size correlated with faster degradation; however, the final methane potential was not increased. Crystallinity is another important physical parameter that can affect anaerobic digestion performances, and amorphous parts are more readily degraded than the crystalline fraction (Meereboer et al., 2020; Monlau et al., 2013a). In polymers, the crystalline fractions are more ordered, while the amorphous fractions are more flexible, thereby making them more vulnerable to microorganism-mediated hydrolysis (Abraham et al., 2020). The amorphous region is known to allow permeation of moisture and microorganisms. Its degradation, therefore, increases the surface area of the available crystalline region (Meereboer et al., 2020). Molecular weight is another important parameter, and it has been reported that the molecular weight was negatively correlated with the biodegradation rate (Abraham et al., 2021). Šmejkalová et al. (2016) tested the biodegradation of different molecular weight PLAs and PCLs by thermophilic anaerobic digestion. They observed that a reduction in the polymer molecular weight was associated with an enhancement of the biodegradation kinetics (Šmejkalová et al., 2016). An additional complicating factor is the complexity of the commercial biodegradable plastic blends, which can affect their physicochemical properties and thus their rates of biodegradation (Artham and Doble, 2008). Indeed, commercial plastics are not merely composed of a single chemically homogenous component. Rather, they can contain different polymers (blends) or low molecular weight additives (Artham and Doble, 2008; Meereboer et al., 2020).

5) Strategies to enhance the biodegradability of biodegradable plastics

As previously described, especially under mesophilic conditions, most biodegradable plastics except starch and PHAs exhibit a very low degree of biodegradability by anaerobic digestion (Bátori et al., 2018; Narancic et al., 2018). Furthermore, most biodegradable polymers have slower biodegradation kinetics than the conventional substrates (manures, biowastes, sludges, etc.) that are fed into biogas plants. This point is important for the development of efficient organic waste management systems, as it implies that the residence time of biodegradable plastics in AD is longer than the current residence times of industrial AD reactors (Narancic and O'Connor, 2019). In order to enhance the biodegradability kinetics and ideally the methane potential, three main

strategies can be implemented: pretreatment technologies, the incorporation of additives into the polymers (fibers, enzymes, calcium carbonate, etc.), and inoculum acclimation or bioaugmentation (Calabro et al., 2019; Ryan et al., 2017a, 2017b).

5.1 Pretreatment technologies

Pretreatments have been widely investigated in the past several decades to enhance the biodegradability of various substrates (*e.g.*, sludges, manures, animal by-products, municipal solid wastes, algae) by anaerobic digestion (Brémond et al., 2018; Carlsson et al., 2012; Carrere et al., 2016). Pretreatment is seen as a strategy to enhance AD in terms of increased methane yield, faster kinetics, and solids reduction (Carlsson et al., 2012; Monlau et al., 2012). Pretreatment can be applied at different levels of a biogas plant: at the entrance of the digester, between the digester and the post-digester, or in recirculation of the solid digestate, in which case it is called "post-treatment". Four main categories of pretreatment can be distinguished: mechanical, thermal, chemical, and biological (Carlsson et al., 2012; Carrere et al., 2016). Mechanical pretreatments generally consist of grinding, shredding, extrusion, or cavitation, and they are widely applied in current biogas plants (Carrere et al., 2016; Monlau et al., 2012). Regarding chemical pretreatments, for the time being, they are limited to lab-scale applications due to their high costs and their environmental consequences, even though some alkaline treatments (calcium hydroxide notably) have displayed promising results, especially with lignocellulosic substrates and animal by-products (Brémond et al., 2018). Thermal pretreatment can be applied at low temperatures (50 to 70 °C for a few hours or days) or high temperatures (170 to 190 °C for a few minutes with or without pressure). The most used commercial processes are: THP from Cambi®, with more than 30 operating facilities; and Biothelys® or Exelys® from Veolia, with approximately 10 facilities constructed to date (Carrere et al., 2016). Finally, biological pretreatments can be divided into three parts: enzymatic, anaerobic, and aerobic (Brémond et al., 2018). For enzymatic pretreatment, which appears to be one of the most promising biological options, the three key players in the enzyme market are Novozymes (Denmark), DSM (Netherlands), and Dupont (USA) (Brémond et al., 2018).

Few studies to date have investigated the use of pretreatments to enhance the biodegradability of biodegradable plastics by AD (Benn and Zitomer, 2018; Vargas et al., 2009; Yagi et al., 2009). The main results obtained from the data in the literature are presented in Table 5. First of all, some authors have investigated the impact of mechanical size reduction on the anaerobic biodegradability of various biodegradable plastics (Ryan et al., 2016; Yagi et al., 2012, 2009). Yagi et al. (2009) investigated the impact of particle size reduction on the anaerobic biodegradability of PCL. Interestingly, no differences in the biodegradation of PCL powder (after 60 days of incubation) for the various particle size distributions (0–250 µm, 250–500 µm, 0–125 µm, and 125–250 µm) were observed, and approximately 80-90% biodegradability was achieved for all conditions. However, particle size reduction has a positive effect on the kinetics of biodegradation, as small particle sizes result in faster degradation kinetics. Similarly, Ryan et al. (2016) investigated the impact of particle size (from 10 µm to 3 900 µm) on the anaerobic biodegradability of PHBV. For all of the granulometry tested, the methane production was approximately 530 ± 12 mL/g, corresponding to a biodegradability of 86 ± 2%. Nonetheless, with particle sizes below 840 µm, longer lag phases were observed. That observation could be explained by a faster hydrolysis of the small-sized plastic particles leading to a temporary inhibition of methanogenesis due to VFAs accumulation. On the other hand, Yagi et al. (2012) observed a slower rate of anaerobic digestion with crushed or small pieces of PLA film than with large pieces of film or with uncut film under thermophilic conditions. They assumed that the small pieces of PLA floated on top of the sludge, thereby reducing the total surface area in contact with the methanogenic sludge and thus negating the advantage of size reduction.

The data in the literature indicates that thermal and thermo-chemical pretreatments have also been investigated for the degradation of plastics such as PLA (Battista et al., 2021; Hobbs et al., 2019; Vargas et al., 2009) and PHB (Benn and Zitomer, 2018). For instance, Vargas et al. (2009) investigated the impact of steam pretreatment (3 h, 120 °C) on the anaerobic digestion of PLA under both mesophilic and thermophilic conditions. Under mesophilic conditions, the methane potential was enhanced from 2 to 90 NL CH_4 kg^{-1} VS, whereas under thermophilic conditions, the methane potential was enhanced from 187 to 225 NL CH_4 kg^{-1} VS. Hobbs et al. (2019) investigated the impact of alkaline pretreatment (21 °C, pH > 11, 15 days) to enhance the methane potential of crystalline and amorphous PLA in co-digestion with food wastes. Amorphous and crystalline PLA

reached near-complete solubilization at 97% and 99%, respectively, when alkaline pretreatment was applied (Hobbs et al., 2019). Treated PLA in co-digestion with food wastes did not exhibit a lag phase and it produced 1021 NmL of CH_4 at day 70 compared to 756 NmL of CH_4 for untreated PLA (Hobbs et al., 2019). However, a 15-day pretreatment period does not seem suitable for industrial purposes. Benn and Zitomer, (2018) have also investigated the impact of alkaline pretreatment (35-55 °C, pH = 10-12, 24-48 h) to enhance the methane potential of various PHAs and PLAs under mesophilic conditions. For all the polymers tested, alkaline pretreatment allowed enhancement of the methane potential, especially for PLA, for which the methane potential increased from 1 L CH4 kg^{-1} VS to 86 L CH4 kg^{-1} VS (Benn and Zitomer, 2018). Furthermore, Benn and Zitomer, (2018) reported a reduced lag time for pretreated PLA compared to untreated PLA. Calabro et al. (2019) have also investigated the impact of pretreatments (pre-digestion, alkaline, mechanical) on the anaerobic digestion of Mater-Bi®-compostable bags (Calabro et al., 2019). In one of their experiments, they reported a positive impact of alkaline pretreatment (NaOH 5% w/w, 24 h) on the methane potential, resulting in a 70% enhancement, but pre-digestion and mechanical pretreatment did not result in a discernible enhancement. Wang et al. (2011) also investigated a novel combination of anaerobic hyperthermophilic treatment in the presence of ammonia followed by thermophilic anaerobic digestion of two grades of PLA. Such a combination allowed final methane conversion ratios of 82% and 77% to be achieved after 22 days with two types of PLA. Nonetheless, some other studies have also highlighted the absence of an impact, or even a negative impact, when thermo-chemical pretreatments were used with biodegradable plastics (Battista et al., 2021; Endres and Siebert-Raths, 2011). For instance, Endres and Siebert-Raths, (2011) attempted pretreatment of PLA at 70 °C for 1 h with no pH control, but this resulted in less biomethane production than with untreated PLA. More recently, Battista et al. (2021) also investigated the impact of thermo-acid (48 h, pH = 2, HCl) and thermo-alkaline (48 h, pH = 12, NaOH) pretreatments on the AD performances of PLA. Interestingly, Battista et al. (2021) demonstrated that both of these pretreatments did not enhance the methane potential of the PLA.

No studies to date have reported regarding the use of biological pretreatments (*e.g.*, aerobic, fungal, enzymes) to enhance the anaerobic degradation of biodegradable plastics. Nonetheless, such pretreatment technologies have already been shown to improve the biodegradability of organic wastes (Brémond et al., 2018; Carrere et al., 2016; Mahdy et al., 2015; Yu et al., 2013). Recently, several enzymes from fungal strains have been found to increase the solubilization and degradation of various biodegradable plastics such as PLA, PHA, and PCL (Emadian et al., 2017; Oda et al., 2000; Panyadhanakul et al., 2019; Roohi et al., 2017; Tokiwa et al., 2009). Among the enzymes that have been found to enhance the degradation of biodegradable plastics are esterases, proteases, lipases, cutinases, and PHA depolymerases (Narancic and O'Connor, 2019; Numata et al., 2009; Oda et al., 2000; Tokiwa and Calabia, 2006). For instance, Oda et al. (2000) tested the polylactide-degrading activity of 56 commercially available proteases. Purafect 4000L, Protin A, and Savinase® 16.0L were the most efficient enzymes for PLA degradation, with polylactide-degrading activities of 15.8, 23.4, and 41.9 U/mg protein, respectively. The application of enzyme products in anaerobic digestion can be carried out in three configurations: (1) pretreatment with enzymes followed by a single-stage anaerobic digestion, (2) direct enzyme addition to a single-stage digester, and (3) direct enzyme addition at the first stage (hydrolysis and acidification) of a two-stage digestion system (Romano et al., 2009).

5.2 Addition of additives into plastic composition to enhance biodegradable plastics biodegradability

Another strategy to enhance the biodegradability kinetics and rate is direct addition of co-product fillers in polymers, such as fibers (Ryan et al., 2017a, 2017b), calcium carbonate (Hegde et al., 2018), lactic acid monomer (Lee et al., 2016), eggshells (González Petit et al., 2015), or enzymes (https://www.carbiolice.com/). It has been shown that the addition of fillers in plastics not only reduces the cost but also makes use of recycled waste streams and enhances properties such as fluid barrier properties and thermal stability (Ryan et al., 2017a, 2017b; Syafri et al., 2017). Ryan et al. (2017a, 2017b) recently investigated the addition of oak wood flour (WF) as a filler in raw PHBV and maleated PHBV. As shown in Table 5, Ryan et al. (2017b) investigated the biodegradation of PHBV with different ratios of WF (0%, 20%, and 40%). The lag time for CH_4 production decreased linearly with increasing WF fraction. The decrease in lag time can be attributed to increased surface accessibility for microbial enzymatic hydrolysis as a result of fibers being present at the composite surface. Ryan et al. (2017b) have reported the main mechanisms involved in anaerobic degradation of biodegradable plastics in the presence of fillers such as wood fibers. When exposed to moisture, the WF becomes increasingly saturated, leading to

cracking, which exposes additional surface area to enzyme hydrolysis (Ryan et al., 2017b). By contrast, the extent of the composites biodegradation (*i.e.*, the ultimate CH_4 production) decreased linearly with increasing WF content, due to incomplete biodegradation of the lignin-rich WF, which is not degraded during the AD process (Barakat et al., 2012; Ryan et al., 2017b). Ryan et al. (2017a) have also investigated the impact on the AD process of two fiber-matrix compatibilization treatments that are generally applied to enhance the moisture resistance of biocomposites: (1) hydrophobic silane treatment of the wood fiber and (2) grafting of hydrophilic maleic anhydride groups to the PHBV matrix (mPHBV). Neat PHBV and mPHBV produced 94 and 95% of their respective theoretical CH_4 yields. For PHBV mixed with 20% fibers, the maximum rate of anaerobic biodegradation was enhanced by 30% for the maleic anhydride treatment and 92% for the silane treatment relative to untreated samples. Similarly, Hegde et al. (2018) have investigated the addition of calcium carbonate (5% w/w) as a filler in a PLA/PBS blend. Interestingly, calcium carbonate addition enhanced the degradation rate (45-49% compared to 37% in the control) and the biomethane potential of the PLA/PBS mixture. Such an enhancement of the biodegradation rate can be explained by an increase in the permeability to water as a result of the incorporation of natural fillers. As the hydrolytic chain scission mechanism can be expected to increase with the water permeability of the materials, this could contribute to enhancement of the biodegradation of composite materials compared to the neat matrix (David et al., 2019).

5.3 Inoculum acclimation (biostimulation) or bioaugmentation

Although there have been no studies to date specifically in regard to these techniques for improving the biodegradability of biodegradable plastics, the inoculum origin and acclimation (a type of biostimulation), as well as bioaugmentation, have been shown to enhance the anaerobic biodegradability of organic wastes (De Vrieze et al., 2015; Nzila, 2017; Sambusiti et al., 2014; Yang et al., 2016). Sambusiti et al. (2014) have investigated the impact of four inocula (urban, agricultural, mixture of agricultural and urban, and granular) on the methane potential and methane production rate of ensiled sorghum. Interestingly, the nature of the inoculum did not affect the final methane potential, although it did have a significant influence on the methane production rate. In particular, the fastest biomethanization occurred using urban sludge (hydrolytic kinetic constant kh = 0.146 d^{-1}) while the slowest was with agricultural sludge (kh = 0.049 d^{-1}). Similar results were shown by Koch et al. (2017) during the anaerobic digestion of different substrate (*i.e.,* sewage sludge, dried whole crop maize, food waste) using three inocula (*i.e.,* from plant treating wastewater, agricultural waste or biowaste). According to these results, it would appear that the nature of the inocula as well as the nature of the biomass has a major impact on the results. It is, therefore, relevant to, in the future, investigate the impact of the inoculum origin on the anaerobic biodegradability performances of biodegradable plastics.

Another strategy to enhance the anaerobic biodegradability of biodegradable plastics is inoculum acclimation prior to the AD process. This has previously been applied successfully to anaerobic digestion of various organic wastes (Gonçalves et al., 2012, Yangin-Gomec et al., 2018). For instance, Gonçalves et al. (2012) demonstrated that the use of an acclimated microbial consortium is a promising way to accelerate the start-up of the digestion process and to enhance the overall anaerobic treatment of a real-life oily wastewater such as olive mill wastewater. These results suggest that changes in the structure of a microbial community can lead to changes in biogas production, and controlling the ultimate methanogenic archaeal community may promote successful methane production in anaerobic reactors (Yangin-Gomec et al., 2018). Aside from acclimation or in combination with biostimulation, bioaugmentation of inocula, consisting of the addition of efficient biomethane-generating single or mixed microorganisms (fungi, bacteria, or enzymes) in bioreactors, can enhance the AD process (Hu et al., 2016; Nzila, 2017). Bioaugmentation, by the addition of specific microbial strains, is a promising technique to accelerate the biodegradation of biodegradable plastics so that they degrade in time frames comparable with other organic materials (Castro-Aguirre et al., 2018). This strategy has been used for the past 15 years to increase biomethane production of organic wastes (Mshandete et al., 2005; Weiß et al., 2011; Zhong et al., 2011). Bioaugmentation has been successfully used in composting-mediated aerobic degradation of plastics (Castro-Aguirre et al., 2018; Nair et al., 2016). For instance, Castro-Aguirre et al. (2018) evaluated the effect of bioaugmentation (using *Geobacillus*) on the biodegradation of PLA and PLA bio-nanocomposites (BNCs) under simulated composting conditions. Bioaugmentation with *Geobacillus* increased the production of CO_2 and accelerated the biodegradation phase of PLA and BNCs when tested in compost and inoculated vermiculite with compost mixed culture, probably due to the secretion of esterases from such microorganisms. A better

understanding of the key bacteria involved in the biodegradation of plastics is needed to be able to use bioaugmentation strategies in anaerobic digesters.

6) Developments required to introduce biodegradable plastic wastes in anaerobic digesters

To make the treatment of biodegradable plastics in anaerobic digestion systems possible, the impact of anaerobic digestion on the quality and safety of the digestate has to be evaluated. Similarly, international standards need to be established in order to facilitate the collection and introduction of biodegradable plastic wastes in anaerobic digestion.

6.1) Impact of biodegradable plastics AD on the agronomic quality and safety of digestate

Although a number of emerging digestate valorization routes are being explored (Monlau et al., 2015; Sheets et al., 2015), the most common use of digestate is for agronomic purposes (Nkoa, 2014). Prior to being used, digestates need to be compatible with their application in agriculture (Nkoa, 2014). Digestates fulfilling this compatibility criterion can be directly applied to crop fields or they can undergo a solid-liquid separation (*e.g.*, screw press, filter press, centrifugation) resulting in a solid and a liquid digestate fraction (Akhiar et al., 2017; Guilayn et al., 2020; Möller and Müller, 2012). The solid fraction can be used directly as a soil amender or it can be aerobically composted to attain the attributes required for being applied as a soil amendment (Tambone et al., 2010). In contrast, the liquid fraction can be used as liquid fertilizer for plant growth (Möller and Müller, 2012; Tampio et al., 2016).

Biodegradable plastics are expected to become a significant fraction of biogas plant feedstock, especially in municipal organic wastes, where some items will be packaged or manufactured with biodegradable plastics (Möller and Müller, 2012; Narancic et al., 2018; Zhang et al., 2018). To date, the potential impact of biodegradable plastics on the digestate quality is expected to be low, although this has not been specifically investigated (Narancic et al., 2018; Zhang et al., 2018). The main contribution of biodegradable plastics to feedstock composition will depend on the composition of the biodegradable plastics: enrichment in C, O, and H, with basically no other elements, including nitrogen or sulfur (Zhang et al., 2018). Typically, the quality parameters of digestates are those that affect their agronomic value, mostly as plant nutrients or soil amendments. As soil amendments, digestates contribute to soil organic matter turnover, thus improving the soil's biological, chemical, and physical characteristics (Tambone et al., 2010). In terms of plant nutrients, the properties associated with the agronomic quality of the digestate are the dry organic matter content; total N; total NH_4^+-N; total P; water-soluble P, K, Ca, Mg, and S content; C/N ratio; and pH. The attributes of digestates from AD with biodegradable plastics-containing feedstock have not been assessed. However, it is reasonable to assume that the impact of biodegradable plastics on the quality of agronomic digestates (*i.e.*, fertilizers and amendments) is minor. To our knowledge, there is still no information in the literature regarding the effects of biodegradable plastics on the quality and the properties of digestates.

Aside from agronomic evaluation, the safety of digestates should also be monitored. Toxic chemicals and physical contaminants depend on the predominant composition of the feedstock, and the presence of such entities in the original biodegradable plastic should be carefully monitored (Coelho et al., 2018; Zhang et al., 2018). No specific recommendations have been made to date regarding the presence of heavy metals in digestates from AD plants treating biodegradable plastics, but the same threshold level as that in the European Norms for organic wastes (Saveyn et al., 2014) seems appropriate. In the case of industrial compost, the European Norm "EN 13432" specifies that threshold levels should not be exceeded for heavy metals or for the fluorine content in biodegradable plastics treated by composting. In parallel with heavy metals, the content of organic contaminants (polycyclic aromatic hydrocarbons, polychlorinated biphenyls) also needs to be monitored (Briassoulis et al., 2010). It is then of prime importance to evaluate the potential benefits, or otherwise the phytotoxicity, on plants through growth tests. Such tests can be performed in climatic chambers, greenhouses, or ideally in real field-scale assays (Adamcová et al., 2019; Gell et al., 2011; Nkoa, 2014). As for the composting process, the EN13432 Norm specifies carrying out assays with two plant species (one monocotyledon and one dicotyledon) at two compost doses (25% or 50% w/w), according to the protocol defined in the OECD 208 guidelines. It is likely that there will be further developments in ecotoxicity testing in the near future, especially given the increasing amount of biodegradable plastics on the market and the consequent increase in the amount of biodegradable plastics in organic wastes (Ruggero et al., 2019). For ecotoxicity testing, it could be quite pertinent to verify the impact of the digestate on edaphic (bacteria, protozoa, and earthworms) and aquatic ecosystem organisms (*e.g.* algae, daphnia) (Pivato et al., 2016; Sforzini et al., 2016; Tigini et al., 2016).

In case the biodegradable plastics or their blends undergo substantial biodegradation during the AD, the carbon is converted to CH_4 and CO_2, and only non-biodegraded components will remain in the digestate. The latter will

preferentially stay in the solid fraction (*e.g.*, fragments of films and packaging), while some of the residue will remain in both the liquid and solid fractions of the digestate (molecules derived from biodegradable plastics after the AD process, mainly in the form of micro- and nanoplastics). Generally, plastic fragments smaller than 5 mm are considered to be microplastics (Shruti and Kutralam-Muniasamy, 2019). Definition of the term "nanoplastics" is still a matter of debate, and different studies have set the upper size limit at either 1 000 nm or 100 nm (Gigault et al., 2018). High concentrations of microplastics can affect the AD performance and impact the digestate dewaterability and other properties (Li et al., 2020). Furthermore, plastic fragments that remain visible to the naked eye after the AD compromise the acceptability of the digestate as a commercial product (Zhang et al., 2018) as they may not meet the farmers' expectations. Therefore, biodegradable plastics that undergo significant biodegradation under AD are expected to have a minor impact on the digestate quality, and this is especially the case under thermophilic AD conditions, whereby biodegradation is promoted by the temperature. Nevertheless, as the time required to treat biodegradable plastics by AD is longer than the current residence times for conventional organic waste treatment, especially in the case of mesophilic AD, it is paramount that the impact of residuals plastics on the agronomic quality and safety of the digestate is monitored (Accinelli et al., 2020; Adamcová et al., 2019; Narancic and O'Connor, 2019). For this purpose, the occurrence and the impact on terrestrial and aquatic ecosystem of micro- and nanoplastics that may be generated during AD of biodegradable plastics should be taken into account, especially if the digestate is used for agronomic applications (Shruti and Kutralam-Muniasamy, 2019; Zimmermann et al., 2020). Microplastics and nanoplastics are small enough to be ingested by a wide variety of organisms, and thus they also represent a pathway for the introduction of various environmental contaminants (*e.g.*, heavy metals, organic contaminants) into the food chain (Ng et al., 2018; Shruti and Kutralam-Muniasamy, 2019). A number of preliminary studies have demonstrated that biodegradable microplastics have similar ingestion mechanisms, exhibit similar characteristics, and cause analogous effects as those of conventional microplastics (Shruti and Kutralam-Muniasamy, 2019; Straub et al., 2017) even though, due to their biodegradable nature, their persistence in soil and aquatic environments is shorter than that of conventional microplastics.

6.2) International norms and labels to evaluate the biodegradability by AD

To enhance the acceptability of biodegradable plastics among the general public and to facilitate results comparison, it is of prime importance that standards are established to assess the biodegradability of biodegradable plastics by anaerobic digestion. Generally, the norms are classified as specifications or analyses and tests. Specification norms set the requirements, characteristics, and performance thresholds to be achieved during biodegradation trials, while analyses and test norms harmonize the methods for the quantification of these characteristics and performances. To date, the major focus of society and policy makers has been in regard to composting as an end-of-life scenario for biodegradable plastics (Briassoulis et al., 2010; Narancic and O'Connor, 2019). Currently, the composting process has two specification norms (EN 13432 Packaging - Requirements relating to packaging recoverable by composting and biodegradation - Test program and criteria for evaluation of acceptance packaging, EN 14995 Plastics - Evaluation of compostability - Test scheme and specifications) that specify the requirements, characteristics, and performance thresholds by addressing four characteristics: 1) initial polymer characterization 2) biodegradability, 3) disintegration during biological treatment, and 4) effect on the quality of the resulting compost. Both EN 13432 and EN 14995 also include a number of specifications for coupling AD with composting. Nonetheless, to date, no specification norms have been devised at the international level for the stand-alone AD process. Only analyses and test norms (ISO and ASTM) have been developed to date to evaluate the biodegradability of biodegradable plastics in the AD process.

Table 6 shows the main analyses and test norms for the evaluation of the biodegradability of biodegradable plastics in AD process, along with their main operational parameters. Presently, at the international level, two norms (NF EN ISO 14853 and ASTM D5210) are referenced for the biodegradability in a liquid medium (1-3% TS) under mesophilic conditions. Similarly, one norm (ISO 13975) is designed for wet processes (< 15% TS) and is applicable in mesophilic and thermophilic environments. Finally, two norms (ASTM D551 and ISO 15985) are made for simulating dry processes (> 15% TS) under thermophilic conditions. Covering all of the typologies of anaerobic digesters is difficult and there is presently an absence of norms to assess the biodegradability of plastics under dry mesophilic AD conditions. Up to date the norms ISO 13975 (simulating CSTR under mesophilic or thermophilic conditions), ASTM D5511 and NF EN ISO 15985 (simulating the dry process under thermophilic conditions) appear to be the most useful and representative norms to simulate and assess the biodegradability

of plastics in anaerobic digestion at industrial-scale processes. All of these norms are based on the principle of the biochemical methane potential (Holliger et al., 2016; Raposo et al., 2011), with a number of variations for the operational conditions and for the nature of the inoculum (Table 6). For all norms, the biodegradability rate is calculated based on the carbon emitted in gaseous form in the biogas (CH_4, CO_2) compared with the amount of total organic carbon in the tested material. Only the ISO 14853 and ISO 13975 norms take into account the inorganic carbon dissolved in the medium, which increases the accuracy. Failure to account for dissolved inorganic carbon can result in underestimation of the biodegradability.

Finally, at present, there are no specification norms (as is the case for composting, soil, and marine environments) nor analysis and testing norms to assess the disintegration of plastics by AD and the ecotoxicity of the resulting digestate. The recent development of the anaerobic digestion sector, as well as the diversity of the AD processes encountered, may explain this delay in European standardization, even though there have been a number of new initiatives in recent years such as the Open-Bio project (Open-Bio, 2016). The Open-Bio project investigated how markets can be opened for bio-based products through standardization, labelling, and procurement, and there was a specific focus on the biogas sector. Finally, it is important to design and implement test standards representative of industrial reality but that also take into account economic constraints in order to make them accessible to as many stakeholders as possible. The deployment of a biodegradable plastics sector will also require better organization and interactions between the various stakeholders, including public organizations, industrial entities, end-users, and policy makers.

Table 6. International standards available and operational parameters to assess the biodegradation of plastics under anaerobic conditions (adapted from Lagnet et al., 2020)

7) Perspectives (*Figure 6 associated*)

This review has highlighted the potential of biodegradable plastics in the near future and their end-of-life in anaerobic digestion, especially for the urban, agricultural, and agro-industrial sectors. However, and contrary to the composting process, the treatment of biodegradable plastics in the AD sector is still in its infancy. This review has highlighted that many technical, environmental and societal barriers will have to be addressed in coming years, covering the various steps of the AD chain shown in ***Figure 5***. Indeed, future works and efforts should address the following points:

1) **Composition of the biodegradable plastics**: It is clear that the nature of the polymer (PLA, PHA, PBS, TPS, etc.) significantly influences its biodegradation by AD. There has, however, been a paucity of investigations of the biodegradability of commercial biodegradable plastic blends (except Mater-Bi®) as well as a number of emerging polymers such as mcl-PHAs and crystallized poly(lactic acid) by anaerobic digestion. Further research is needed in order to obtain reference values regarding the anaerobic biodegradability of the most common and emerging biodegradable polymers that can be marketed. In parallel, additives are generally added to biodegradable polymers (between 0.05 to 70% (w/w)) to manipulate selected physical properties, such as toughness, flexibility, or barrier properties, thereby improving their functional capabilities (Brebu, 2020; Hahladakis et al., 2018). The most commonly used additives are: plasticizers, flame retardants, antioxidants, acid scavengers, light and heat stabilizers, lubricants, pigments, antistatic agents, slip compounds, and thermal stabilizers, as well as fillers and reinforcements (Hahladakis et al., 2018). For the near future, it will thus be interesting to assess the influence of such additives on the performances and stability of the AD processes, and their influence on the microbial communities. Recently, a number of biobased alternatives to additives have been developed, such as polyphenols (Diouf-Lewis et al., 2018). Nonetheless, the incorporation of such additives, even if they are environmentally friendly, should take into account their impact on the AD process, as such compounds have previously been reported to be inhibitory of anaerobic micro-organisms (Monlau et al., 2014).
2) **Morphology and properties of the biodegradable plastics**: The shape and size of biodegradable plastic wastes after their use, and especially their thickness and morphology (bags, films, bottles, food packaging, etc.) should be considered to evaluate the biodegradability in AD process. These parameters will influence the biodegradation rate, incorporation in the digester, and the application of eventual pretreatments upstream of the AD process. The typology and the morphology of plastic supports can also affect the process later on by creating obstructions during the introduction in the biogas plants.

Consequently, pretreatments can be required in some processes, especially for liquid CSTR system. Furthermore, the rate of biodegradation will also be influenced by the physicochemical properties of the polymers (*e.g.*, the chemical composition, crystallinity, porosity, accessible surface area, molecular weight). A better understanding of the correlation between the physicochemical properties of plastics and their biodegradation rate will be useful to determine key parameters and thus provide guidance to plastic manufacturers with their grading in order to develop plastics that are resistant during their use while still having biodegradability rates suitable to conventional HRT of industrial biogas plants.

3) **Pretreatments**: As previously mentioned, biodegradable plastics can be subjected to pretreatment strategies (mechanical, thermal, and thermo-chemical) to enhance their biodegradability. In the future, results should be consolidated at a pilot scale to validate the promising results obtained in lab-scale and further drawn technico-economic analysis. In parallel, other pretreatment strategies including physical (Garuti et al., 2018) and biological processes (*i.e.*, enzymes, bacteria, and fungi) should be investigated (Brémond et al., 2018). The incorporation of fillers or enzymes is another interesting way for enhancing biodegradability. The incorporation of enzymes is still in its infancy and should be implemented at an industrial scale, although fibres and mineral additives have been shown to enhance the biodegradation of plastics and are hence a way to reduce the price of plastics (Herzberger et al., 2018; Ryan et al., 2017a).
4) **AD process stability and performances:** Currently, the biodegradability of biodegradable plastics by AD is determined by protocols defined at the European and the international level. The methodology is based on BMP (biochemical methane potential) tests (Holliger et al., 2016; Raposo et al., 2011). Nonetheless, aside from their accuracy, BMP tests are very time-consuming (several days). In recent decades, alternative methods for determining quickly the anaerobic biodegradability of organic wastes have been developed and can be probably transfer to assess the biodegradability of biodegradable plastics (Jingura and Kamusoko, 2017; Lesteur et al., 2011). Among these methods, the use of near-infrared spectroscopy (Mortreuil et al., 2018; Triolo et al., 2014); chemical composition model (Gunaseelan, 2007; F Monlau et al., 2012), quick respirometric tests (Lesteur et al., 2011), and the Envital kit® (Bellaton et al., 2016) have proven useful. Although all of these technologies are promising tools for fast biodegradability analysis, as they were developed for organic wastes, they need to be tailored to biodegradable plastics. In regard to the biodegradability of plastics by AD, it will also be important to consider the various typologies (CSTR, dry batch, dry continuous, etc.) of biogas plant available. Indeed, the biodegradability and the performances will differ according to the technologies and the operational conditions.

   Aside from the technologies used, the operational parameters (*e.g.*, HRT, organic loading rate, temperature) will influence the biodegradation performances of biodegradable plastics. Assays in continuous mode are required, as batch tests that have been mostly used, which may fail to truly predict the performance of full-scale anaerobic reactors (Sambusiti et al., 2013). The co-digestion of plastics with biowastes should also be better investigated in the future, as biodegradable plastics can counterbalance the low C/N ratio of some organic wastes such as food wastes and thereby further enhance the AD stability and performances. Finally, there is a paucity of information available in the literature regarding the influence of biodegradation of plastics on AD microbial communities.
5) **Post-treatment and agronomic quality of digestate:** Last but not least, digestate valorization should be carefully considered in the overall biodegradable plastics-AD chain. Generally, digestates are used for agronomic applications and are separated into solid and liquid phases through various processes (screw press, press filter, centrifuge, vibrating screen, etc.) (Akhiar et al., 2017; Guilayn et al., 2020). The liquid phase is generally used as a fertilizer whereas the solid phase is applied as a soil amender. In parallel, plant phytotoxicity tests of digestates involving biodegradable plastics should be managed in greenhouse and field-scale operations along with the influence of residual plastics and plastic additives on soil microbial communities. Aside from agronomic and safety parameters (heavy metals, organic contaminants), it will be interesting to assess the influence of different digestate phase separation systems on the distribution of residual plastics. Indeed, incomplete degradation during waste management processes and leakage of bioplastics into the environment are becoming major concerns that need to be further investigated (Cucina et al., 2021). It is of prime importance to better understood the becoming of the residual biodegradable microplastics present in digestates in a composting process or during soil application (Cucina et al., 2021; Peng et al., 2022). In the future, the research priority

should be to gain a better understanding of the fate and the potential impact of microplastics and nanoplastics, as this is still largely unknown (Qin et al., 2021). As concerns ecotoxicity parameters, besides residuals biodegradable microplastics in the digestate, a particular attention should also be paid on the additive that can be liberated during the fragmentation of biodegradable plastics. Presently, common types of plastic additives are classified into stabilizers (to prolong lifespan of plastic products), plasticizers (to modify mechanical properties), antioxidants (to delay oxidation of plastics), pro-oxidants (to obtain faster degradation), surfactants (to promote surface properties), and other additives (to improve functionality) based on their different purposes (Qin et al., 2021). Finally, emerging valorization routes of digestates other than for agronomic purposes should also be considered if the digestate is not suitable for agronomic application (Monlau et al., 2015; Sheets et al., 2015).

6) **Normalization and standards:** The improvement of a normative and certification system on anaerobic digestion is a main challenge in the coming years in order to improve the perception of the general public and facilitate the comparison of results. The complexity will be to take into account the existing biogas plant diversities (dry batch, dry continuous, wet) and their operating conditions, which differ according to process temperatures and feeding (HRT around 30-50 days for biowastes, 50-120 days for agricultural wastes). If analytical and testing standards exist for the evaluation of biodegradability in both mesophilic and thermophilic conditions, there is clearly a lack of specifications norms (as is the case for composting, soil, and marine environments). Specification norms set the requirements, characteristics, and performance thresholds to be achieved during biodegradation trials.

Aside from the technical feasibility of the end-of-life of biodegradable plastics in the AD process, it is also important to consider the overall sustainability from an economic, environmental, and social perspective. In regard to the environmental impact, there have been few studies in the past several decades (Alarico et al., 2017; Piemonte, 2011) regarding comparison of the life cycle assessments (LCA) of various biodegradable plastics end-of-life (composting and anaerobic digestion) versus conventional plastic disposal (recycling, incineration, landfill, etc.). Alarico et al. (Alarico et al., 2017) investigated the LCA of the end-of-life of biodegradable plastics packaging in various scenarios, including incineration, landfill, composting, and anaerobic digestion. Interestingly, they found that AD had the lowest potential global warming impact among the various scenarios, mainly due to the recovery of a large amount of thermal energy and electricity in the process (Alarico et al., 2017). Piemonte (Piemonte, 2011) investigated the LCA for both PLA and Mater-Bi®, taking into consideration composting, incineration, anaerobic digestion, and mechanical recycling processes as the final scenarios. They found that incineration, composting, and to a lesser extent anaerobic digestion processes clearly underperformed, from an environmental point of view, relative to mechanical recycling. Nonetheless, if the biodegradable plastics are contaminated with biowastes or with other organic wastes, mechanical recycling becomes complex and, therefore, anaerobic digestion can become an attractive option.

Figure 5: The main R&D perspectives that should be addressed in the future to enhance the recovery of biodegradable plastics by anaerobic digestion.

8) Conclusions

This review highlights the potential of end-of-life treatment of biodegradable plastics by anaerobic digestion. The main conclusions are as follows:

- Not all polymers and reactor typologies have received the same degree of attention; anaerobic digestion under thermophilic conditions has been investigated less than mesophilic conditions. There have been few investigations of the anaerobic biodegradability of commercial blends (except Mater-Bi®) and emerging polymers (such as mcl-PHAs and crystallized poly(lactic acid)). Furthermore, most of the data regarding biodegradability performances come from studies based on BMP test experiments, which may not provide a true picture of the performance of full-scale anaerobic reactors. To date, there is little information available regarding continuous anaerobic digestion experiments at pilot or industrial scales.
- Thermoplastic starch and PHAs can reach near-complete mineralization in a relatively short period under both mesophilic and thermophilic conditions. These materials could potentially be treated in

biogas plants without alteration of the hydrolytic retention time used. Other polymers such as poly(lactic acid) and poly(Ɛ-caprolactone) have been shown to have very low biodegradation rates under mesophilic conditions and not in adequation with biogas plants HRT. Under thermophilic conditions, their biodegradation rate is greatly increased. Lastly, some polymers that appear to be biodegradable under industrial composting conditions were very poorly biodegraded even with very long digestion times, such as poly(butylene succinate) and poly(butylene adipate/terephthalate).

- Little is known about the microorganisms involved in the anaerobic digestion of biodegradable plastics. Recently, some publications have reported that *Tepidimicrobium* sp. plays a key role in the anaerobic biodegradation of PLA and commercial blends. A better understanding of the microbial taxa involved in the process could open the way to the use of bioaugmentation to improve the performance of the process. In parallel, a number of pretreatment strategies (*e.g.*, mechanical, thermal, and thermo-chemical) and the addition of various additives in the plastic formulation (*e.g.*, fibers, minerals, enzymes) have been shown to enhance the biodegradation rate of the biodegradable plastics in anaerobic digestion, albeit with varying degrees of success. Finally, there is a grey area surrounding the quality and potential toxicity of the digestate from anaerobic digestion of biodegradable plastics.
- Standardization, labelling, and further research on the sustainability (*i.e.*, economic, environmental, and societal) is required in the near future in order to improve the public perception and the social acceptance of these new products.

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DECLARATION OF COMPETING INTEREST

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

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G. Cazaudehore: Conceptualization, Methodology, Writing the original draft. **R. Guyoneaud** and **F. Monlau**: Conceptualization, Methodology, Writing - Review & Editing, Supervision, Project administration, and Funding acquisition. **P. Evon:** Writing - Review & Editing, **L Martin-Closas**: Writing - Review & Editing, **A. M. Pelacho**: Writing - Review & Editing; **C. Raynaud**: Writing - Review & Editing.

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Figures and Tables captions

Figure 1: General overview of the main issues addressed in this bibliographic review.

Figure 2: Global production capacities of bioplastics in 2019 (***Source:*** European Bioplastics 2019).

Figure 3: Boxplot representing the degradation level of the various biodegradable polymers, the number of experiments (designated exp.), and the number of studies. White circles correspond to each study reported.

Figure 4. Mean biodegradation (%) vs mean time of incubation (days) for the various data on biodegradable plastics found in literature for both mesophilic and thermophilic conditions

Figure 5: The main R&D perspectives that should be addressed in the future to enhance the recovery of biodegradable plastics through the anaerobic digestion process.

Table 1. The main focus area of recently published review papers on anaerobic digestion of biodegradable plastics.

Table 2. The main biodegradable polymers commercially available, and their most important physicochemical properties.

Table 3. Mesophilic biochemical methane potential and operational parameters described in the literature.

Table 4. Thermophilic biochemical methane potential and operational parameters described in the literature.

Table 5. Strategies tested in the literature for enhancement of the methane potential and the kinetics of biodegradable plastics.

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Can anaerobic digestion be a suitable end-of-life scenario for biodegradable plastics? A critical review of the current situation, hurdles, and challenges

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Figure 1. General overview of the main issues addressed in this bibliographic review

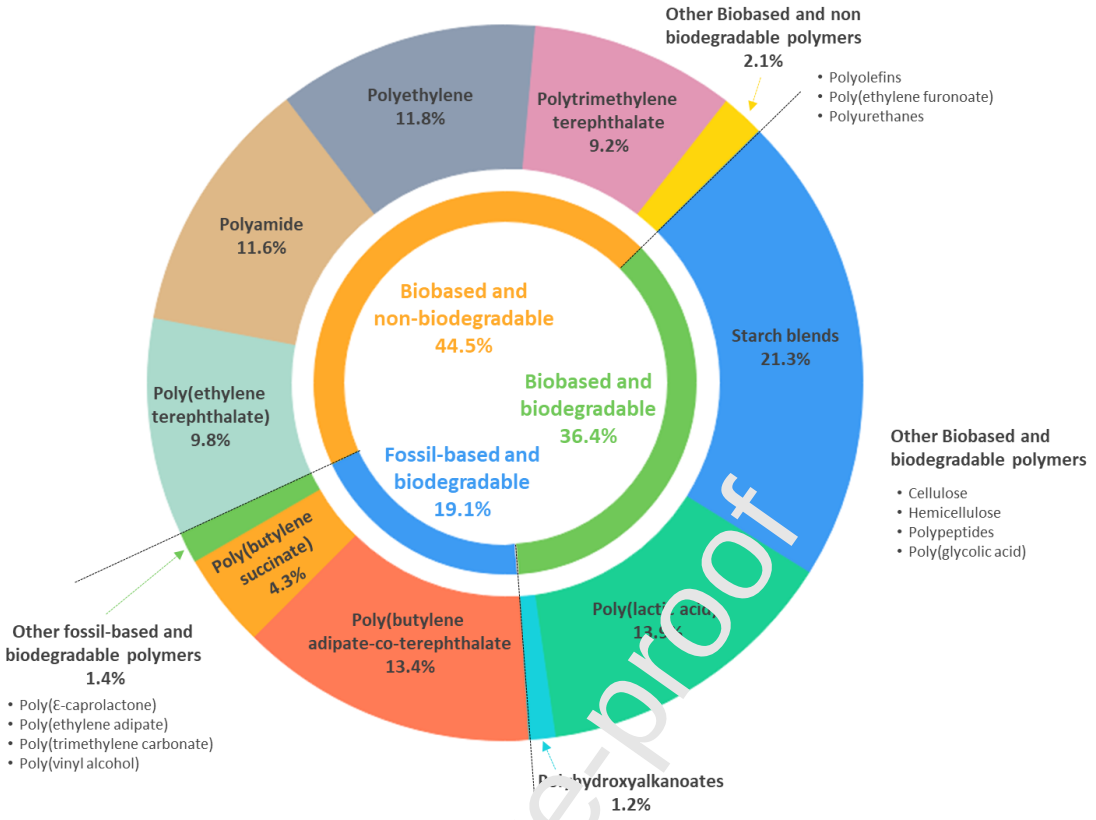


Figure 2. Global production capacities of bioplastics in 2019 (Source: European Bioplastics 2019, Nova Institute).

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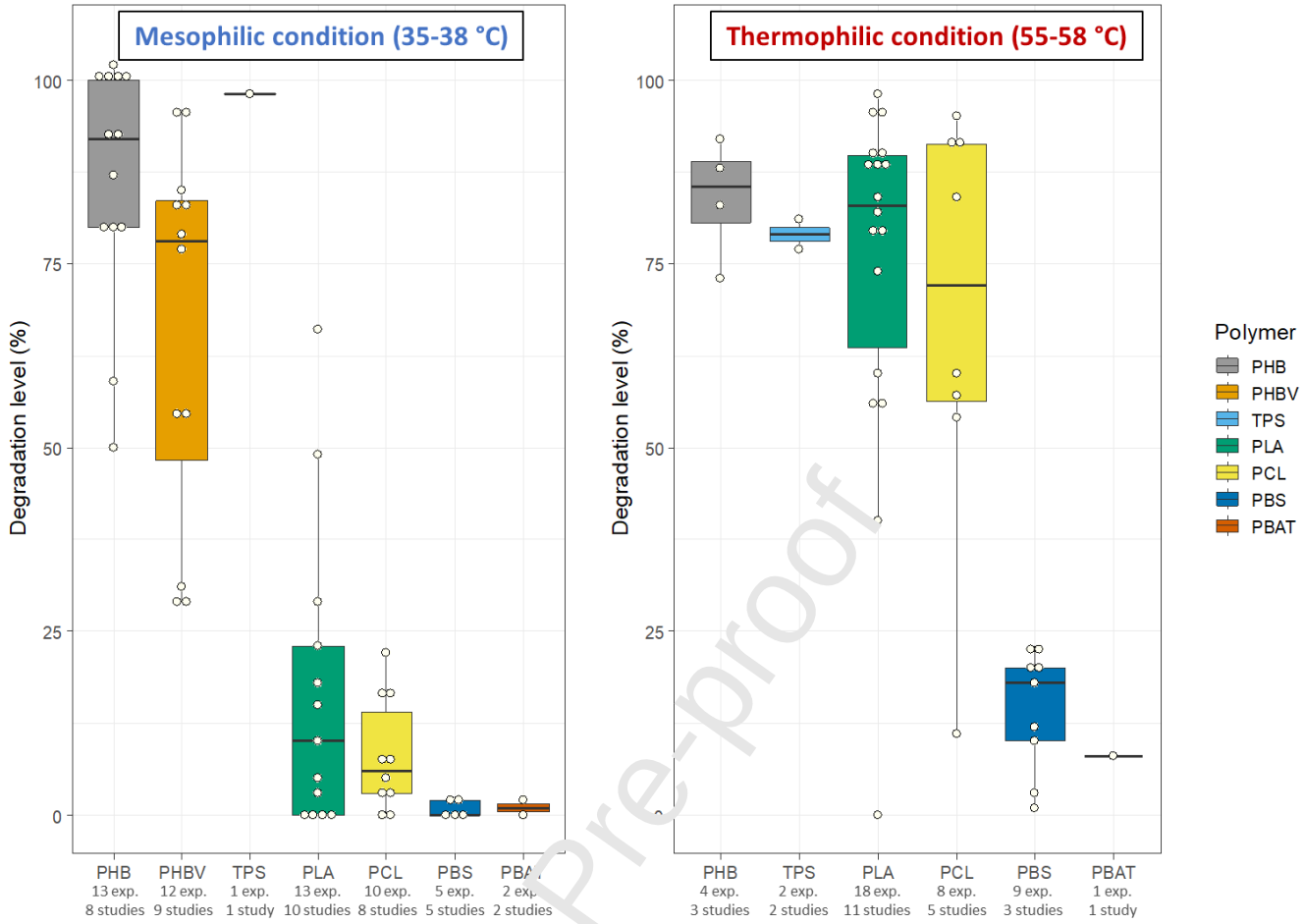


Figure 3: Biodegradation (%) reached at the end of the BMP tests under mesophilic and thermophilic condition. The number of experiments (noted exp.), and the number of studies per biodegradable polymers are also presented. White circles correspond to each experiment reported. The Figure is based on data provided in Table 3 and Table 4.

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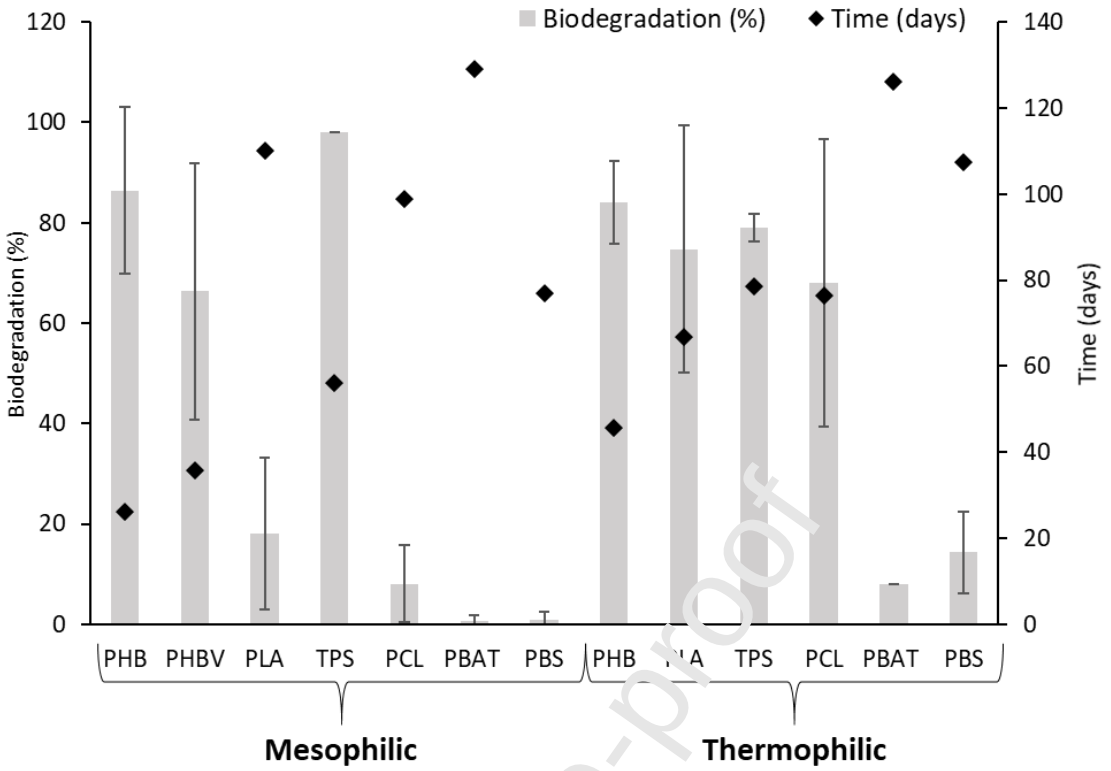


Figure 4. Mean biodegradation (%) vs mean time of incubation (days) for the various data on biodegradable plastics found in literature for both mesophilic and thermophilic conditions. The Figure is based on data provided in Table 3 and Table 4.

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Figure 5. The main R&D perspectives that should be addressed in the future to enhance the recovery of biodegradable plastics by anaerobic digestion.

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Table 1. The main focus area of recently published review papers on the anaerobic digestion of biodegradable plastics

Main Authors and publication year	Title	Journal	Main topics
Abraham et al., 2021	Anaerobic co-digestion of bioplastics as a sustainable mode of waste management with improved energy production – a review	Bioresource Technology	A review on anaerobic digestion of biodegradable plastics in comparison with composting process. Data are compiled in exhaustive tables. Explanation of the main biodegradation mechanisms. Use of pretreatments to improve the biodegradability Challenges and research needs
Lagnet et al., 2020	Norms review of the bioplastics biodegradability-Synthesis	ADEME report (French environmental and energy management agency)	A review on standardization and normalization of biodegradation test in different environments. A critical synthesis on the existing norms and potential improvements required
Quecholac-Piña et al., 2020	Degradation of plastics under anaerobic conditions: A short review	Polymers	Short review on plastics degradation under anaerobic digestion including also landfill system. Explanation of the main degradation mechanisms and techniques used to assess the biodegradability
Folino et al., 2020	Biodegradation of Wasted Bioplastics in Natural and Industrial Environments: A Review	Sustainability	A review on the biodegradability of biodegradable plastics in different environments including composting, anaerobic digestion but also aquatic and soil environment. Comparison of biodegradability in these different environments. A specific paragraph is dedicated to the standardization and normalization of biodegradable plastics
Bátori et al., 2018	Anaerobic degradation of bioplastics: A review	Waste Management	A review on anaerobic digestion of biodegradable plastics. A specific paragraph is dedicated for each typology of biodegradable plastics. Anaerobic digestion of biodegradable plastics in biogas plants: challenges, standards, and suggestions
Ruggero et al., 2019	Methodologies to assess biodegradation of bioplastics during aerobic composting and anaerobic digestion: A review	Waste Management & Research	Standards to assess biodegradation of biodegradable plastics under aerobic composting and anaerobic digestion. Description and comparison of the main Norms existing and methodologies for evaluating the biodegradability

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Table 2. The main biodegradable plastics commercially available, and their most important physicochemical properties

Polymer type	**TPS**	**PLA**	**PBAT**	**PBS**	**PCL**	**PHB**	**PHBV**
Main producers	- Vegeplast (France) - Novamont (Italy) - Biotec® (UK)	- NatureWorks (USA) - Total Corbion PLA (NL) - Futerro (Belgium) - BASF (Germany)	- BASF (Germany) - Jinhui Zhaolong (China) - Biofed (Germany) - Eastman (USA)	- PTT MCC Biochem Company Ltd (Thailand) -Succinity (Germany) - MCPP (Japan) - Showa Denko (Japan)	- Perstorp (UK) - BASF (Germany)	- TianAn (China) - Yield10 Bioscience (USA) - Bio-Fed® (Germany) - Biomer® (Germany)	- TianAn, (China) - Yield10 Bioscience (USA) - Biomer (Germany)
Melting point (°C)	n.m.	155[b]	110-120[c]	115[d]	58-60[e]	170-185[f]	170-176[g]
Glass transition (°C)	n.m.	60[b]	-30[h]	-32[h]	-61[h]	-5 to 5[h]	-10 to 5[h]
Molar mass (g/mol)	n.m.	n.m.	n.m.	n.m.	20,000-80,000[e]	n.m.	n.m.
Density (g/cm^3)	n.m.	1.24[b]	1.25-1.27[c]	1.26[d]	1.1[e]	1.26[f]	1.25[g]
Cristallinity rate (%)	n.m.	0-40[h]	n.m.	34-45[h]	67[h]	60-80[h]	30-80[h]
Maximal tensile strength (MPa)	1-23[a]	45[b]	30-40[c]	30[d]	14	16[f]	39[g]
Young's modulus (MPa)	0.1-3.5[a]	n.m.	n.m.	720[d]	0.19[c]	n.m	2.8-3.5[g]
Elongation at break (%)	0,5-80[a]	<5%[b]	560-710[c]	330[d]	> 500[c]	6[f]	2[g]

n.m., not mentioned.

[a] TPS from corn whole plant or from wheat flour (T55 type), with glycerol (from 0 to 25% w/w) and/or citric acid (from 0 to 14% w/w) used as plasticizer(s).

[b] Value for Luminy LX 175 grade (Total Corbion).

[c] Value for Ecoflex® F Blend C1200 grade (BASF).

[d] Value for PBE003 (Natureplast)

[e] Value for Capa™ 6800 grade (Perstorp).

[f] Value for M VERA GP1012 (Biofed)

[g] Value for ENMAT Y1000P (Tianan)

[h] Value from Shruti, V.C., Kutralam-Muniasamy, G., 2019. Bioplastics: Missing link in the era of Microplastics. Science of The Total Environment 697, 134139. https://doi.org/10.1016/j.scitotenv.2019.134139

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Table 3: M

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Biodegradable plastic nature	Size and shape	Temperature (°C)	Operational parameters and supplementary data	Time (days)	Methane potential	Percentage of methane conversion	Reference
Mater-bi (PCL + Amidon, Novamont)	Pieces of plastic bag <1mm	35	Plastic: 1g. Inoculum: 5mL of pig slurry mixed with synthetic medium for methanogens and acclimated to mesophilic anaerobic condition	90	33 L CH_4 kg^{-1} VS	6%	Vasmara and Marchetti, 2016
PLA (Ingeo)	Pieces of plastic cup <1mm				0 L CH_4 kg^{-1} VS	0%	
PHBV (0,5% HV, ENMAT Y1000P)	31,25 mm x 6,2 mm x 2,1 mm rectangular prism	37	Plastic to inoculum ratio: 0.5 g VS g^{-1} VS . Inoculum: digestate from a mesophilic digester. Method: ASTM D5210-90	42	630 L CH_4 kg^{-1}	83 %	Ryan et al., 2017(a)
PHBV (0,5% HV ENMAT Y1000P)	420-840 µm	37	Plastic to inoculum ratio: 0.5 g VS g^{-1} VS. Inoculum: digestate from a mesophilic digester. Method : ASTM D5210-90	20	600 L CH_4 kg^{-1}	79%	Ryan et al., 2016
PLA (Fabri-Kal)	Plastic cup ground to 3mm	37	Plastic: 1 g. Inoculum: 10mL of anaerobic inoculum	60	2 L CH_4 kg^{-1} VS	0%	Vargas et al., 2009
PHB (ENMAT Y3000, TianAn)	0,15 mm	35	Plastic: 125 mg. Inoculum: 50 mL of lab inoculum fed with nutritive media and powdered milk	40	199 L CH_4 kg^{-1} ThOD	50%	Benn and Zitomer, 2018
PHB (MIREL F1006, Metabolix)					233 L CH_4 kg^{-1} ThOD	59 %	
PHB (MANGO materials)					316 L CH_4 kg^{-1} ThOD	80%	
PHB (Mirel M2100, Metabolix)					316 L CH_4 kg^{-1} $ThOD^{1}$	80%	
PLA (Ingeo 2003D, Natureworks)					1 L CH_4 kg^{-1} ThOD	0%	
PBS (Sigma-Aldrich)	125-250 µm	37	Plastic: 10 g. Inoculum: mesophilic digestate from an anaerobic digester (37 °C) treating cow manure and green waste	277	-	0%	Yagi et al., 2014
PCL (Sigma-Aldrich)					-	3-22 %	
PLA (Unitika)					-	29-49%	
PHB (Sigma-Aldrich)				9	-	90%	
PHB (MW 540 000 $g.mol^{-1}$, Biopol BX G08,)	25 mm of diameter 100 µm of thickness film	37	Plastic: 0.2 g. Inoculum: sludge from a laboratory anaerobic reactor treating wastewater from a sugar factory. Method: ASTM D 5210-91	9	-	100%	Abou-Zeid et al., 2001
PHBV (MW 397 000 $g.mol^{-1}$,Biopol BX P027)				42	-	29%	
PCL (MW 50 000 $g.mol^{-1}$, Polyscience Inc.)					-	7.5%	
PHA (PHA-4100, Metabolix USA)	1-2 mm wide pellets	37 ± 2	Plastic to inoculum ratio: 4 g L^{-1}. Inoculum: sludge from a semi continuous anaerobic digester fed with food waste, olive and cheese waste. Method: ASTM 5511-02	11	-	102%	Greene, 2018
			Plastic to inoculum ratio: 8 g L^{-1}. Inoculum: sludge from a semi continuous anaerobic digester fed with food waste, olive and cheese waste. Method: ASTM 5511-02		-	95%	
PLA (Natureworks)			Plastic to inoculum ratio: 4 g L^{-1}. Inoculum: sludge from a semi continuous anaerobic digester fed with food waste, olive and cheese waste. Method: ASTM 5511-02	20	-	< 5%	
PHB (MW 540 000 $g.mol^{-1}$, Biopol BX G08,)	19 mm of diameter film	37	Plastic: 35-40 mg. Inoculum: sludge from an anaerobic laboratory reactor fed with wastewater from sugar industry. Method: ASTM D 5210-91	8	-	101%	Abou-Zeid et al., 2004
PHBV (MW 397 000 $g.mol^{-1}$, Biopol BX P027)				42	-	29%	
PCL (MW 50 000 $g.mol^{-1}$, Polyscience Inc.)					-	16%	
1,4-butanediol/adipic acid (MW 40 000,GBF)					-	1.1%	
1,4-butanediol (50 mol %) adipic acid (30 mol %)/Terephtalic acid (20 mol %) (MW 47 600, Hüls AG)					-	5.5%	
PHB (MW 540 000 $g.mol^{-1}$, Biopol BX G08)			Plastic: 35-40 mg. Inoculum: sludge from an anaerobic digester of a municipal WWTP. Method: ASTM D 5210-91	8	-	100%	
PHBV (MW 397 000 $g.mol^{-1}$, Biopol BX P027)				42	-	31%	
PCL (MW 50 000 $g.mol^{-1}$, Polyscience Inc.)					-	17%	
1,4-butanediol/adipic acid (MW 40 000, GBF)					-	11%	
1,4-butanediol (50 mol %) adipic acid (30 mol %)/Terephtalic acid (20 mol %)					-	11%	
PHBO (90% PHB, 10% HO)		35	Plastic: 100 $mg.L^{-1}$. Inoculum: digestate from an anaerobic digester treating WWTP sludge.	60	-	88%	Federle et al., 2002
PCL			Plastic: 10 $mg.L^{-1}$. Inoculum: digestate from an anaerobic digester treating WWTP sludge	122	-	0.2%	
PCL	1cm^2 film pieces	37 ± 2	Plastic to inoculum ratio: 0.5 g VS g^{-1} VS. Inoculum: digestate from a mesophilic anaerobic digester fed with food wastes and manure	30	15.8 ± 21.1 L CH_4 kg^{-1} VS	6.5%	Nunziato et al., 2018
PCL 40% TPS 60%					133.3 ± 17,5 L CH_4 kg^{-1} VS	32.3%	
PCL 60% TPS 40%					74.2 ± 15.7 L CH_4 kg^{-1} VS	18.5%	
PHBV (PHB/HV; 92/8, w/w)	5x60 mm film	35	Inoculum: anaerobic digested sludge from a WWTP. Method: ASTM D5210	20	-	85%	Shin et al., 1997
Cellophane	20x40 mm film				-	80%	
PLA (lab)				100	-	0%	
PBS (Elson Green)					-	0%	
PBES (MW 100 000, Sky Green)					-	0%	

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Table 3: Mesophilic biochemical methane potential and operational parameters described in the literature (2/3)

Biodegradable plastic nature	Size and shape	Temperature (°C)	Operational parameters and supplementary data	Time (days)	Methane potential	Percentage of methane conversion	Reference
PHB	Granular form	?	Plastic to inoculum ratio: 10 g VS g^{-1} VS. Inoculum: digestate from a WWTP anaerobic digester. Method: ASTM D 5210-91	23	-	100%	Majone et al., 1995
Mater-bi (Starch + PE, AF08H, Novamont)	2x15 cm strips	35	Inoculum : Mixture of sewage sludge treating domestic sewage and paper sludge (3:1 ratio)	40	-	32%	Day et al., 1994
Mater-bi (Starch + PE, AF10H, Novamont)					-	30%	
PHBV (ICI)					-	55%	
PCL (Tone, Union Carbide)					-	5%	
Ecostarplus (starch + PE)					-	12%	
PLA (Argonne A)	6x5 cm film				-	10%	
PLA (Argonne B)					-	15%	
PHB	Powder	35	Plastic: 5 mg. Inoculum: anaerobically digested domestic sewage sludge	16	0.14 mmol CH_4	87%	Budwill et al., 1992
PHBV (13% HV)					0.16 mmol CH_4	96%	
PHBV (20% HV)					0.17 mmol CH_4	83%	
PLA	Granules	37	Plastic: 30 mg. Inoculum: anaerobic sludge from a WWTP. Method: ASTM D 5210	100	-	60%	Itävaara et al., 2002
Mater-bi (60% starch, 40% hydrophilic resin)	Entire bag	35 ± 0.5	Plastic to inoculum ratio: 0.5 g VS g^{-1} VS. Inoculum: liquid digestate from an anaerobic digester fed with manure, agro-wastes and residues	15	144 ± 18.4 L CH_4 kg^{-1} VS	-	Calabro et al., 2019
Cellulosic plates	Plate			44	311 ± 37.6 L CH_4 kg^{-1} VS	-	
PCL-Starch blend (55% PCL, 30% Starch, 15% aliphatic polyester)	?	35	Plastic to inoculum ratio: 2g VS L^{-1}, Inoculum: 20 mL digestate from an anaerobic digester treating sewage sludge. Method: ASTM E1196-92	139	554 L CH_4 kg^{-1} VS	83%	Cho et al., 2011
PBS				100	11 L CH_4 kg^{-1} VS	2%	
Mater-Bi (PCL+Starch+Glycerin, ZI01U, Novamont)	Film	35	Inoculum: anaerobic sludge from an anaerobic digester. Method: ASTM D 5511-94	81	-	21%	Scandola et al., 1998
	Pellets				-	10%	
PCL (Tone, Union Carbide)	Powder	35	Inoculum: 2mL of digestate from an anaerobic digester treating sewage sludge. Method: ISO 14853 Supplementary data: Cellulose was only degraded to 66% in 28 days.	28	-	0	Massardier-Nageotte et al., 2006
PLA (Natureworks, Cargill)	2x2 cm film 20 µm of thickness				-	0	
PBAT					-	0	
Mater-bi (Starch + PCL, Novamont)					-	23%	
PHBV (8.4% HV, ICI)	46.4 µm	35	Plastic: 1% w/w, Inoculum: 10% w/w anaerobic sludge from a WWTP of a sugar factory	30	-	95%	Reischwitz et al., 1997
PLA (Biopolymer-4043D, Nature Works)	< 2x2 cm	35 ± 2	Inoculum: sludge from a WWTP. Method: ISO 14853	56	-	0%	Narancic et al., 2018
PCL (CAPA 6500, Perstorp)					-	3%	
PBS (PBE 003, NaturePlast					-	0%	
PHO (Bioplastech R, Bioplastech)					-	12%	
PHB (ENMAT Y1000, TianAn)					-	102%	
TPS (Bioplast TPS, BIOTEC)					-	98%	
PLA/PCL (80/20)					-	0%	
PLA/PBS (80/20)					-	0%	
PLA/PHB (80/20)					-	0%	
PLA/PHO (80/15)					-	2%	
PHB/PHO (85/15)					-	92%	
PHB/PCL (60/40)					-	38%	
PHB/PBS (50/50)					-	15%	
PCL/PHO (85/15)					-	4%	
PCL/TPS (70/30)					-	36%	
Poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) 93% HB, 7% HHx	5x5x1 mm Film	38	Plastic to inoculum ratio: 0.7-0.8 g VS g^{-1} VS. Inoculum: Digestate from a mesophilic anaerobic digester fed with sludge and fats	80	483.8 ± 35.2 L CH_4 kg^{-1} VS	77 ± 6%	Wang et al., 2018
Poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) 93,5% HB 6,5% HHx	5x5x1 mm Flake			40	337.5 ± 100.3 L CH_4 kg^{-1} VS	54 ± 16 %	
Starch (from wheat)/PVOH	Foam	37	Substrate to inoculum ratio: 1 g VS g^{-1} VS. Inoculum: digestate from a mesophilic anaerobic digester	10	270 L CH_4 kg^{-1} VS	72.1%	Guo et al., 2011
Starch (from potato)/PVOH					265 L CH_4 kg^{-1} VS	68.6%	
Starch (from maize)/PVOH					248 L CH_4 kg^{-1} VS	75.4%	

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Biodegradable plastic nature	Size and shape	Temperature (°C)	Operational parameters and supplementary data	Time (days)	Methane potential	Percentage of methane conversion	Reference

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Table 3: Mesophilic biochemical methane potential and operational parameters described in the literature (3/3)

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Cellulose-based metallised film	1x1 cm film	37	Plastic to inoculum ratio: 0.25 g(VS).g(VS)$^{-1}$. Inoculum: digestate from a mesophilic digester treating municipal wastewater biosolids	65	-	74.5%	Zhang et al., 2018
Cellulose-based heat-sealable film					-	86.6%	
Cellulose-based high barrier heat-sealable film					-	84%	
Cellulose-based non heat-sealable film					-	80.4%	
Cellulose diacetate film					-	8.9%	
Starch-based film blend 1					-	18.3%	
Starch-based film blend 2					-	10.2%	
PLA film					-	18.8%	
PLA blend	Pellets				-	2.6%	
Starch (25% amylose) and PVA blend	Film	35	Plastic: 20g. Inoculum: digestate from a wastewater treatment plant. Method: ASTM D [illegible]10-92.	25	-	52 ± 1%	Liu et al., 2016
High-amylose starch (80% amylose)-PVA blend				20	-	54 ± 0.5%	
PBS (Enpol G4560, IRE Chemical Ltd, Korea)	5 x 5 mm thin film (100µm)	37	Plastic: 100 mg. Inoculum: mesophilic anaerobic sludge from a wastewater treatment plant. Method: ISO 11734	113	-	2.2%	Dvorackova et al., 2015
Starch:PVOH blends (90/10%)	5x5x1 mm film	38 ± 0.5	Plastic: 2g. Inoculum: supernatant from the second stage of a laboratory scale digester fed with a mixture of primary domestic sludge and food waste	100	140 L(biogas).kg^{-1} COD	-	Russo et al., 2009
Starch:PVOH blends (75/25%)					118 L(biogas).kg^{-1} COD	-	
Starch:PVOH blends (50/50%)					60 L(biogas).kg^{-1} COD	-	
PVOH (Dupont, Australia)					5 L(biogas).kg^{-1} COD	-	
PHA	PHA accumulated in activated sludge	37	Plastic: addition of 1mL of PHA accumulating sludge (30 g TS L^{-1}). Inoculum: 5mL of sewage sludge from a WWTP	15	250 NmL(CH4).g(VS)$^{-1}$	53 %	Soda et al., 2016
PLA (plastic cup)	2x2x0.5 mm	37 ± 0.5	Plastic to inoculum ratio: 2-4 kg VS m^{-3}. Inoculum: mesophilic digestate from a wastewater treatment plant anaerobic digester (37°C). Method: EN ISO 11734:2003	280	564 L(biogas).kg(VS)$^{-1}$	66%	Bernat et al., 2021
PHB (MW 539 000, Biopol BX G08)	200 µm powder	35	Plastic: 600 mg L^{-1}. Inoculum: domestic sewage sludge	30	-	80%	Puechner et al., 1995
Bioceta (Cellulose acetate)	5x5mm 90 µm of thickness fil		Plastic: 600 mg L^{-1}. Inoculum: domestic sewage sludge	60	-	22%	
Mater-bi ZF03U (PCL + Amidon, Novamont)	5x5mm 35 µm of thickness		Plastic: 600 and 400 mg L^{-1}. Inoculum: domestic sewage sludge		-	28%	
PCL (P787, Union Carbide)	5x5 mm 55µm of thickness and 250µm powder				-	0%	
Mater-bi (Novamont)	0,5-1mm film	35 ± 2	Plastic to inoculum ratio: 0.6-1 g TS g^{-1} TS. Inoculum: anaerobic sludge from an anaerobic digestion plant treating effluents from a brewery Method: ASTM D5526-94d.	32	220 L CH_4 kg^{-1} VS	-	Mohee et al., 2008
PBAT 93 000 g/mol (Ecoflex, BASF)	5x5 mm film 70 µm of thickness	37 ± 2	Inoculum: mesophilic anaerobic sludge (37 °C) from a municipal waste water-treatment plant	126	-	2.2%	Svoboda et al., 2018

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Biodegradable plastic nature	Size and shape	Temperature (°C)	Operational parameters and supplementary data	Time (days)	Methane potential	Percentage of methane conversion	Reference
PCL (Mn 58.1 kg.mol-1)	10x10x0,7mm film	55 ± 1	Plastic to inoculum ratio: 0.38 g COD g^{-1} of volatile suspended solid. Inoculum: thermophilic digested sludge from a digester	140	663 L biogas kg^{-1} TS	60%	Šmejkalová et al., 2016
PCL (Mn 38. kg.mol-1)	Powder			80	643 L biogas kg^{-1} TS	54%	
PCL (Mn 13 kg.mol-1)				70	676 L biogas kg^{-1} TS	57%	
PLA (Mn 44.5 kg.mol-1)	10x10x0,7mm film		Plastic to inoculum ratio: 0.15 g COD g^{-1} of volatile suspended solid. Inoculum: thermophilic digested sludge from a digester	120	677 L biogas kg^{-1} TS	74%	
PLA (Mn 3.4 kg.mol-1)	Powder			90	520 L biogas kg^{-1} TS	56%	
PLA (Mn 0,35 kg.mol-1)				30	625 L biogas kg^{-1} TS	84%	
PHB (Biopol)	2x2 cm pieces	52	Plastic: 3-5 g. Inoculum: Anaerobic solid waste treatment plant	20	-	73%	Itävaara et al., 2002
PLA				40	-	60%	
Mater-bi (60% starch, 40% hydrophilic resin)	entire bag	55 ± 0.5	Plastic to inoculum ratio: 0.5 g VS g^{-1} VS. Inoculum: liquid digestate from mesophilic anaerobic digester fed with manure, agro-wastes and residues shifted progressively to thermophilic condition	30	186 ± 11.8 L(CH_4)kg^{-1} VS	-	Calabro et al., 2019
Cellulose	1x1 cm film	55	Plastic to inoculum ratio: 0.5. Inoculum: sludge from a waste management company	35	280 L biogas kg^{-1} VS	18.3%	Shrestha et al., 2020
	2x2 cm film				260 L biogas kg^{-1} VS	17.1%	
	3x3 cm film				250 L biogas kg^{-1} VS	16.3%	
PLA	1x1, 2x2, 3x3 cm rigid pieces				20 L biogas kg^{-1} VS	0%	
PLA (Luminy L130, Total Corbion)	Pellets	55	Plastic: 3 g. Inoculum: sludge from a thermophilic anaerobic digester treating food waste, plant residues and other organic waste product at 55-60 °C	110	224 ± 4 LCH_4 kg^{-1} PLA	-	Tseng et al., 2019
PLA (Biopolymer-4043D, Nature Works)	< 2x2 cm	55 ± 2	Plastic: 15 g. Inoculum: 1 kg of digestate from a thermophilic reactor treating household waste. Method: High solid Anaerobic digestion (ISO 15985)	80	-	88%	Narancic et al., 2018
PCL (CAPA 6500, Perstorp)				127	-	95%	
PBS (PBE 003, NaturePlast				90	-	12%	
PHO (Bioplastech R, Bioplastech)				50	-	6%	
PHB (ENMAT Y1000, TiTAN)				127	-	92%	
TPS (Bioplast TPS, BIOTEC)				127	-	81%	
PLA/PCL (80/20)				121	-	90%	
PLA/PBS (80/20)				121	-	84%	
PLA/PHB (80/20)				80	-	104%	
PLA/PHO (80/15)				66	-	90%	
PHB/PHO (85/15)				66	-	87%	
PHB/PCL (60/40)				80	-	104%	
PHB/PBS (50/50)				121	-	78%	
PCL/PHO (85/15)				66	-	85%	
PCL/TPS (70/30)				80	-	68%	
PBS (Enpol G4560, IRE Chemical Ltd, Korea)	5x5 mm thin film (100µm)	55	Plastic: 50 mg. Inoculum: mesophilic anaerobic sludge from a wastewater treatment plant acclimated to thermophilic temperature	113	-	20.2%	Dvorackova et al., 2015
	5x5 mm thick film (1,02mm)				-	20.1%	
	Powder (320 µm)				-	18.1%	
	5x5 mm thin film (100µm)		Plastic: 50 mg. Inoculum: mesophilic anaerobic sludge from a wastewater treatment plant shifted to thermophilic temperature with addition of a PBS acclimated inoculum from a previous experiment		-	23.3%	
	5x5 mm thick film (1,02mm)				-	22.0%	
	Powder (320 µm)				-	10.3%	
PLA (Unitaka, Japan)	Crushed film (>500µm)		Inoculum: mesophilic digestate from an anaerobic digester (37 °C) treating cow manure and green waste acclimated to 55 °C. Addition of 20 mL of acclimated sludge to PLA thermophilic digestion during the pre-incubation	60	936 L biogas kg^{-1}	97.5%	Yagi et al., 2012
	1x1 cm film, 25µm of				880 L biogas kg^{-1}	94.5%	

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thickness		
15x34 cm film, 25µm of thickness	893 L biogas kg^{-1}	96%
39x82 cm film, 25µm of thickness	827 L biogas kg^{-1}	89%
Powder 125-250 µm	827 L biogas kg^{-1}	89%

Table 4: Thermophilic biochemical methane potential and operational parameters described in the literature (part 1/2)

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Table 4: Thermophilic biochemical methane potential and operational parameters described in the literature (part 1/2)

Biodegradable plastic nature	Size and shape	Temperature (°C)	Operational parameters and supplementary data	Time (days)	Methane potential	Percentage of methane conversion	Reference
PCL (Mw 65,000, Aldrich)	125-250µm particule	55	Plastic: 10 g. Inoculum: mesophilic digestate from anaerobic digester (37 °C) treating cow manure and green waste acclimated to 55 °C and addition of diluted sludge acclimated to PCL and PLA	47	-	92%	Yagi et al., 2009
PLA (H-400, Mitsui Chemical)			Plastic: 10 g. Inoculum: mesophilic digestate from anaerobic digester (37 °C) treating cow manure and green waste acclimated to 55 °C and addition of undiluted sludge acclimated to PCL and PLA	75	-	91%	
				100	-	79%	
			Plastic: 5 g. Inoculum: mesophilic digestate from anaerobic digester (37 °C) treating cow manure and green waste acclimated to 55 °C and addition of diluted sludge acclimated to PCL and PLA	85	-	80%	
Mater-bi (PCL + Amidon, Novamont)	Small piece of plastic bags <1mm	55	Plastic: 1g. Inoculum: 5 mL of pig slurry mixed with synthetic medium for methanogens and acclimated to mesophilic anaerobic condition	90	303 L CH_4 kg^{-1} VS	55%	Vasmara and Marchetti, 2016
PLA (Ingeo)					267 L CH_4 kg^{-1} VS	56%	
PLA (Fabri-Kal, Inc., Kalamazoo)	Plastic cup ground to 3mm	58	Plastic: 1 g. Inoculum: 10 mL of anaerobic inoculum	56	187 L CH_4 kg^{-1} VS	40%	Vargas et al., 2009
PBS (Sigma-Aldrich)	125-250 µm	55	Plastic: 10 g. Inoculum: mesophilic digestate from anaerobic digester (37 °C) treating cow manure and green waste acclimated to 55 °C. Pre-incubation of the inoculum with 20 mL of sludge acclimated to PLA	100	-	1-3%	Yagi et al., 2013
PCL (Sigma-Aldrich)				45	-	84%	
PLA (Unitika)				80	-	82%	
PHB (Sigma-Aldrich)				18	-	83-88%	
PLA (Naturewoks 4043D)	Sheets	52 ± 2	Plastic to inoculum ratio: 0.5 g VS g^{-1} VS . Inoculum: digestate from a mesophilic anaerobic digester fed with food wastes and manure shifted to thermophilic temperature	36	409 L CH_4 kg^{-1} VS	90%	Hegde et al., 2018
PCL	1cm^2 film	52 ± 2	Plastic to inoculum ratio: 0.5 g VS g^{-1} VS. Inoculum: digestate from a mesophilic anaerobic digester fed with food wastes and manure shifted to thermophilic temperature (10 days)	30	44.4 ± 19.5 L CH_4 kg^{-1} VS	11,3%	Nunziato et al., 2018
TPS (70% starch from MP Biomedicals LLC and 30% glycerol)					320 L CH_4 kg^{-1} VS	77.1%	
80% PCL 20 % TPS					104 ± 9.1 L CH_4 kg^{-1} VS	26.2%	
PBAT 93 000 g/mol (Ecoflex, BASF)	5x5 mm film 70 µm of thickness	55 ± 2	Inoculum: mesophilic anaerobic sludge (35 °C) from a municipal waste water-treatment plant acclimated to thermophilic temperature (55 °C) for two weeks	126	-	8.3%	Svoboda et al., 2018
PLA (plastic cup)	2x2x0.5 mm	58 ± 0.5	Plastic to inoculum ratio: 2.4 kg VS m^{-3}. Inoculum: mesophilic digestate from a wastewater treatment plant anaerobic digester (37 °C) acclimated to 58 °C for 14 days. Method: EN ISO 11734:2003	60	835	90%	Bernat et al., 2021

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Biodegradable plastic nature	Size and shape	Temperature (°C)	Pretreatment		Time (days)	Methane production or biodegradability		Reference
			Condition	Effect		Without pretreatment	With pretreatment	
PHB (ENMAT)	0.15 mm	35 °C	35 °C, addition of NaOH until pH 12 for 24h	Reduced lag phase and improved methane production	40	199 L CH_4 kg^{-1} ThOD	398 L CH_4 kg^{-1} ThOD	Benn and Zitomer, 2018
PHB (Mirel F1006)			35 °C, pH 7 for 48h			233 L CH_4 kg^{-1} ThOD	359 L CH_4 kg^{-1} ThOD	
PHB (Mango materials)			55°C, addition of NaOH until pH=10, 24h			316 L CH_4 kg^{-1} ThOD	322 L CH_4 kg^{-1} ThOD	
PHB (Mirel M2100, Metabolix)			55°C, addition of NaOH until pH=12, 24h			316 L CH_4 kg^{-1} ThOD	357 L CH_4 kg^{-1} ThOD	
PLA (Ingeo 2003D Natureworks)			90°C, addition of NaOH until pH=10, 48h			1 L CH_4 kg^{-1} ThOD	86 L CH_4 kg^{-1} ThOD	
PLA	Commercial items	38 °C	Steam exposition, 3h 120 °C	Reduction of the lag phase from 21 days to 0 (under thermophilic conditions)	56	2 L CH_4 kg^{-1} VS / 0.4%	90 L CH_4 kg^{-1} VS / 19.3%	Vargas et al., 2009
		55 °C				187 L CH_4 kg^{-1} VS / 40%	225 L CH_4 kg^{-1} VS / 48.2%	
PLA	Commercial items	37 ± 1 °C	48h, addition of HCl pH= 2	No benefits	250	130 L CH_4 kg^{-1} VS	125 L CH_4 kg^{-1} VS	Battista et al., 2021
			48h, addition of NaOH pH= 12			130 L CH_4 kg^{-1} VS	101 L CH_4 kg^{-1} VS	
Crystalline PLA	2x2 cm cups	37 ± 1 °C	21 °C, pH > 11, 15 days	After 15 days of alkaline pretreatment, near total solubilisation was reached. No lag phase with pretreatment	70	756 mL(CH4)	1021 mL(CH4)	Hobbs et al., 2019
Polycaprolactone (Mw 65,000, Aldrich, USA)	<125 µm	55 °C	Size reduction	Smaller particle size powder degraded faster	25-35	91-98%		Yagi et al., 2009
	125-250 µm				50	89-9 %		
	250-500 µm				65	81-87%		
PLA film 25 µm of thickness (Unitaka, Japan)	> 500µm	55 °C	Size reduction	The crushed or small pieces of PLA film had slower anaerobic biodegradation rates than large pieces of PLA film due to flotation and static cling	60	936 L biogas kg^{-1} / 97.5%		Yagi et al., 2012
	1x1 cm					880 L biogas kg^{-1} / 94.5%		
	15x34 cm					893 L biogas kg^{-1} / 96%		
	39x82 cm					827 L biogas kg^{-1} / 89%		
PHBV (0.5% HV ENMAT Y1000P China)	3900 µm (pellets)	37 °C	Size reduction	A temporary inhibition is observed for particle size less than 840 µm, it is attributed to the accumulation of VFAs caused by the rate of hydrolysis and acetogenesis exceeding that of methanogenesis	40	600 mL(CH4).g^{-1} / 79%		Ryan et al., 2016
	420-840 µm				18	600 mL(CH4).g^{-1} / 79%		
	250-420 µm				25	600 mL(CH4).g^{-1} / 79%		
	150-250 µm				20	600 mL(CH4).g^{-1} / 79%		
	10 µm				25	600 mL(CH4).g^{-1} / 79%		
Mater-bi® (60% starch, 40% hydropilic resin)	Entire bag	35 ± 0.5 °C	5% TS NaOH, 24h	Mechanical pretreatment (reduction to 1x1 cm) and predigestion were also tested but do not show significative improvement	15	144 L CH_4 kg^{-1} VS mass loss = 27.5 ± 1.3%	203 L CH_4 kg^{-1} VS mass loss = 78.2 ± 5.9%	Calabro et al., 2019
Composite of PLA 76%, PBS 19% and CaCO3		52 ± 2 °C	Composite production with addition of two type of 5 % of CaCO3 (Omya TP39914 and Omya TP39968)		60	190 L CH_4 kg^{-1} VS 37 %	210 L CH_4 kg^{-1} VS 45 %	Hegde et al., 2018
Composite of PLA 76%, PBS 19% and CaCO3						190 L CH_4 kg^{-1} VS 37 %	230 L CH_4 kg^{-1} VS 49 %	
PHBV (0.5% HV ENMAT Y1000P China)	Rectangular prism 31,25 mm x 6,2 mm x 2,1 mm	37°C ± 2 °C			42	630 L CH_4 kg^{-1} / 83 %		Ryan et al., 2017(b)
80% PHBV and 20% oak wood flour			Addition of 20% oak wood flour		28	510 L CH_4 kg^{-1} / 73 %		
60% PHBV and 40 % oak wood flour			Addition of 40% oak wood flour		28	430 L CH_4 kg^{-1} / 60 %		
PHBV (ENMAT Y1000P China)	Rectangular prism 31,25 mm x 6,2 mm x 2,1 mm	37°C ± 2 °C	Neat PHBV		80	94 %		Ryan et al., 2017(a)
Maleated PHBV			Maleated PHBV		80	95 %		
80% PHBV 20% oak wood flour			Addition of 20% oak wood flour		50-63	84 %		
80% maleated PHBV 20% oak wood flour			Maleated PHBV + addition of oak wood flour			88 %		
80% PHBV 20% silane treated oak wood flour			Addition of silane treated oak wood flour			83 %		
PLA	Pellets	50 °C	Reduction of the glass transition temperature to 49 °C (bellow the temperature of digestion) by the addition of addives		270	90 %		Lee et al., 2016
PLA + 10 % poly(ethylene glycol) and poly(propylene glycol)					150	90 %		
PLA + 12 % poly(ethylene glycol) and poly(propylene glycol)					150	90 %		

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PLA + 1 poly(propylene glycol) + 2% additives	

Table 5. Strategies tested in the literature for enhancement of the methane potential and the kinetics of biodegradable plastics.

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Table 6. International standards available to assess the biodegradation of plastics under anaerobic conditions

Standard	ISO 14853 (2017)	ISO 13975 (2019)	ISO 15985 (2017)
Equivalent ASTM standard	ASTM D5210	-	ASTM D5511
Process simulated	Liquid process (UASB/fixed bed reactor)	Semi-liquid process (CSTR)	Solid state process
Temperature (°C)	35 (± 2)	35 (± 3) or 55 (± 5)	52 (± 2)
Inoculum	Digested sludge from a wastewater treatment plant (treating mainly household water). Alternatively, anaerobic sludge prepared in laboratory.	Digestate from an AD plant preferably treating domestic sewage. Alternatively, from AD plant treating livestock faeces or garbage	Digestate from a thermophilic dry reactor (>20% TS) treating organic fraction of municipal solid waste. Alternatively, liquid digestate dehydrated (>20% TS) by the use of centrifugation, press or dried at 58 °C
Inoculum preparation	Pre-incubation up to 7 days at 35 °C (± 2) and possible acclimation to the test material	Pre-incubation up to 5 days at 35 (± 3). In case of thermophilic experiment, mesophilic inoculum could be acclimated to 55 °C for one month. Possible acclimation to the test material	Pre-incubation up to 7 days at 52 °C (± 2).
Inoculum quality control	Organic carbon < 20 mg L^{-1}	TS < 15%, pH between 7.5-8.5	TS ≥ 20%; pH between 7.5 and 8.5; VFAs < 1g kg^{-1}; $N\text{-}NH_4^+$ between 0.5-2 g kg^{-1}
Reactors volume	Between 0.1 and 1 L	≥ 1.5 L	≥ 0.75 L
Number of replicate	3	3	3
Positive control	Cellulose, PHB or Poly(ethylene glycol)	Microcrystalline cellulose < 20 µm	Microcrystalline cellulose < 20 µm
Plastic shape and size	Powder (250 µm), film or fragment of items	Powders or films	Powder, pellets, film or fragment with surface ≤ 2x2 cm
Amount of plastic	20-200 mg organic carbon L^{-1} of inoculum	7-10 g VS L^{-1} of inoculum	15-20 g VS kg^{-1} of inoculum
Method of biodegradation calculation	Dissolved inorganic carbon and gaseous carbon are compared to the introduced organic carbon	Dissolved inorganic carbon and gaseous carbon are compared to the introduced organic carbon	Gaseous carbon are compared to the introduced organic carbon
Duration of the test	Maximum of 90 days	Maximum of 90 days	15 days extensible
Validity criteria	Biodegradation yield of the positive control > 70% after 60 days and the difference between the replicates < 20%	Biodegradation yield of the positive control > 70% after 15 days and the difference between the replicates < 20%	Biodegradation yield of the positive control > 70% after 15 days and the difference between the replicates < 20%

(adapted from Lagnet et al., 2020)

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Can anaerobic digestion be a suitable end-of-life scenario for biodegradable plastics? A critical review of the current situation, hurdles, and challenges

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Highlights:

● Biodegradable plastics will play a key role in the future management of biowastes;

● Anaerobic digestion of biodegradable plastics performs better under thermophilic than under mesophilic conditions;

● Pretreatment can significantly enhance the biodegradation rate of biodegradable plastics by anaerobic digestion;

●The agronomic potential and safety of digestates treating biodegradable plastics streams have not yet been reported;

●There is currently a lack of norms and certifications for biodegradability in anaerobic digestion systems.

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