Tampere University

Manuel Alfonso Albini

ASSESSMENT OF ANAEROBIC DIGESTION OF BIOBASED PACKAGING MATERIAL

Master Thesis
Faculty of Engineering and Natural Sciences
Jukka Rintala, Donya Kamravamanesh
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ABSTRACT

Manuel Alfonso Albini: Assessment of anaerobic digestion of biobased packaging material
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The environmental pollution related to the production and waste management of conventional plastics have led the packaging industries to search for bio alternatives. An alternative to petroleum-based plastics are the bioplastics. Bioplastics can biodegrade under conditions existing in natural environment. One of these conditions is the anaerobic environment. Which is based on the organic recycling, one of the end-of-life options for the waste management of bioplastics. This current research investigated the anaerobic biodegradation of seven different commercially available biomaterials, and two non-biodegradable plastics which were used as control. The study was carried out in different times using two different set-ups. The anaerobic biodegradation was also performed in the presence and absence of kitchen waste to simulate the situation where discarded food packages are often contaminated with food waste. The biochemical methane potential batch test allowed the estimation of the conversion of carbon into gaseous products. There was no evidence that any of the biomaterials inhibited the anaerobic digestion process. It was concluded that, of the seven tested biomaterials, only three plant-based material showed substantial biodegradation under anaerobic conditions. The two compostable material studied here showed a smaller rate of biodegradation under mesophilic condition and an apparent conversion into methane.

Keywords: biodegradable packaging material, biopolymers, food waste, Biomethane production potential, anaerobic digestion, mesophilic conditions.

Preface

I would like to thank the following people, without whom I would not have been able to complete this research!

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Author

Manuel Alfonso Albini

CONTENTS

LIST OF FIGURES

LIST OF TABLES

LIST OF SYMBOLS AND ABBREVIATIONS

AD	Anaerobic Digestion
BC	Bacterial cellulose
BOD	Biological Oxygen Demand
BioPET	Biopoly (ethylene terephthalate)
BioPP	Bio Polypropylene
BMP	Biochemical Methane Production Potential
CE	Current Era
CoA	acetyl moiety of acetyl-co-enzyme A
COD	Chemical Oxygen Demand
CO_2	Carbon dioxide
CH_4	Methane
EFSA	European Food Safety Authority
EU	European Union
FAO	Food and Agriculture Organization
FID	Flame Ionization Detector
FCM	Food Contact Material
FLI	Food Loss Index
GC	Gas Chromatography
HDPE	High-density polyethylene
LDPE	Low-density polyethylene
LLDPE	linear low-density polyethylene
N_2O	nitrous oxide
PBAT	Poly(butylene adipate-*co*-terephthalate)
PBS	Poly(butylene succinate)
PE	Polyethylene
PHAs	Polyhydroxyalkanoates
PLA	Poly(lactic acid)
PLC	Poly(caprolactone)
PP	Polypropylene
TCD	Thermal Conductivity Detector
TS	Total Solid
VS	volatile solid
VSS	Volatile Suspended Solids

INTRODUCTION

Due to their low cost and versatility, plastics present a wide range of applications (Koch and Mihalyi, 2018). Packaging and Building & Construction by far represent the largest end-use markets. In particular, plastic packaging account for the greatest amount of global plastic consumption. More than 99% of plastic packages are made from petroleum-based polymers (Zhao et al., 2020).

The petroleum-based polymers used to produce packages represent a threat to the environment because of their chemical and mechanical properties and in particular to their durability. They persist for hundreds of years in the environmental (Zhao et al., 2020). Packaging production increases the CO_2 emission and more than 50% of the plastic packages are used only once before disposal. The recycling process of plastics faces different challenges, such as the presence of pigmented plastic, multilayered and mixed plastics, when used in food packaging they are contaminated with residual food, and marginal market (Zhao et al., 2020). These challenges decrease the percentage of plastics waste that is recycled but in the other hand increase the amount of plastic that is disposed in landfill. The disposed plastics have a huge impact on the living organism, especially of those present in the marine environment. This is because the living organisms confuse plastic waste with food and this leads to their suffocation after swallowing it (Folino et al., 2020).

Plastic durability, their resistance to degradation as well as the environmental pollution related to their production and waste management have led the packaging industries to search for bio-alternatives, able to improve the sustainability. An alternative to petroleum-based plastics are the bioplastics.

Currently, bioplastics represent only 1% of the about 360 million tonnes of plastic produced annually (Plastic Europe, 2019). The bioplastics market is continuously growing, and the production capacity is expected to increase from around 2.11 million tonnes in 2019 to approximately 2,43 million tonnes in 2024 (European Bioplastics, 2019). Consumer awareness, demand for sustainable products, growing need to reduce fossil dependency, improved properties and new functionalities are the reasons behind the success of bioplastics (European Bioplastics, 2019). Europe and Asia are the major bioplastics manufactures. Europe has the largest bioplastic market and does more research and development than any other region (European Bioplastics, 2019). Bioplastics are used in different markets, even though packaging remain the largest field of application. They are mainly used for the production of rigid and flexible packaging material. This could be because depending on the material, they have the same properties as conventional plastics and offer additional advantages, such as reducing the environmental pollution or having different waste management options. For almost every conventional plastic material and corresponding application there is a bioplastic alternative (European Bioplastics, 2019).

Bioplastics present different option in terms of waste management. In addition to recovering energy and mechanical recycling, industrial composting (organic recycling) became a waste management option. In fact, bioplastics present the ability to biodegrade under conditions exiting in natural environments. The environmental conditions affect the rate of decompositions. Organic recycling includes industrial composting and anaerobic digestion. The biodegradation process in an industrial composting facility is called composting and lead to the conversion of the bioplastics into CO_2, water, and biomass. While the biodegradation that occurs in anaerobic digestion is named biomethanisation, because the process leads to the production of bio-methane. To be suitable for organic recycling, products and materials need to meet the criteria of the European norm EN 13432 on industrial composting.

The evaluation of the biodegradability of bioplastics in anaerobic conditions is important because in the waste streams there is the presence of food packaging, not often sorted from food-waste. The presence of food waste makes the mechanical recycling of plastics difficult. Instead, if bioplastics are used for the manufacturing of food packages this make them suitable for organic recycling, together with the food-waste. Due to the fact that they can be used as substrate in anaerobic digestion (AD) plant. This represent an advantage because the deterioration of organic matter under anaerobic conditions lead to the production of methane that can be use as energy source, as well as the nutrient-rich digestate residue, which is used as fertilizer. Moreover, this makes the collection and sorting process of food packaging easier.

The aim of the research was to assess the biodegradability and compostability, under anaerobic conditions, of some commercially available food packages, manufactured by using bioplastics. The biodegradability and compostability were evaluated by performing biomethane production potential (BMP) tests, for which mesophilic conditions were used. In addition to the bioplastics, other bio-based packages were assessed. More importantly, food waste was used as feedstock in the digester to simulate the conditions were the food packages are discarded with food residues. The results of the study were intended to provide information to the consumers and to the stakeholders on the degradability of the selected bio-based packages and to evaluate the possibility to use them as substrate in an AD plant to produce biogas.

1. BACKGROUND

According to the Food and Agriculture Organization (FAO) (FAO, 2017), the world´s population is projected to reach more than 9 billion by mid-century and may peak at more than 11 billion by the end of the century. This growth is predicted to mainly occur in developing countries, such as sub-Saharan Africa, East and Southeast Asia and in the world’s cities (FAO, 2017). Along with the population growth the market demand for food and feed would continue to grow significantly.

In 2050 to feed a world population of 9 billion people the food production should raise, especially those of key commodities. In order to assure nutrition security, the annual cereal production, for instance, would have to grow from 2.068 billion (today production) to 3.009 billion. Meat production instead from 259 million to 470 million tonnes, so an increase of over 200 million tonnes. Oil crops must increase from 149 million tonnes to 282 million tonnes (FAO, 2017).

As of now, food loss and food waste are important problems representing a waste of resources used in production such as land, water, energy, and inputs. More importantly some 795 million people still suffer from hunger, and more than two billion from micronutrient deficiencies or forms of over-nourishment (FAO, 2017).

Food losses are the decrease in edible food mass that takes place at production, post-harvest, and processing stages in the food supply chain (Parfitt et al., 2010). Food losses occurring at the end of the food chain (retail and final consumption) are called “food waste”, which relates to consumer’s and retailer’s behaviour (Parfitt et al., 2010).

Every year, about 670 million tonnes of food is lost or wasted in high-income countries, and 630 million tonnes in low- and middle-income countries. In total1.3 billion tonnes of food is wasted every year. (FAO, 2017).

The Food and Agriculture Organization (FAO) has established the Sustainable Development Goals to reduce the global food waste at the retail and consumers levels to half its present content by 2030, also reducing the food loss along with production and supply chains, including post-harvest losses (United Nations, 2015).

The Food Loss Index (FLI) has been used to calculate the percentage of food loss globally and by region. FAO through the FLI estimated that only in 2016, 13.8 percent of food produced was lost from the farm excluding the retail stage. As shown in Figure 1 this value has been much higher (20.5 %) in Central and Southern Asia (FAO, 2019). In medium and high-income countries, such as Europe or Norther America, food losses are lower (15,5%), but still present. The cause can be the quantity produced; in many cases it exceeds the demand. In terms of food groups, roots, tubers and oil-bearing crops report the highest level of loss (25%), followed by fruits and vegetables (21%) (Figure 1). Fruits and vegetables have high levels of loss because of their highly perishable nature. Meat & animal products, cereals & pulses present respectively 11% and 9% of food loss (FAO, 2019).

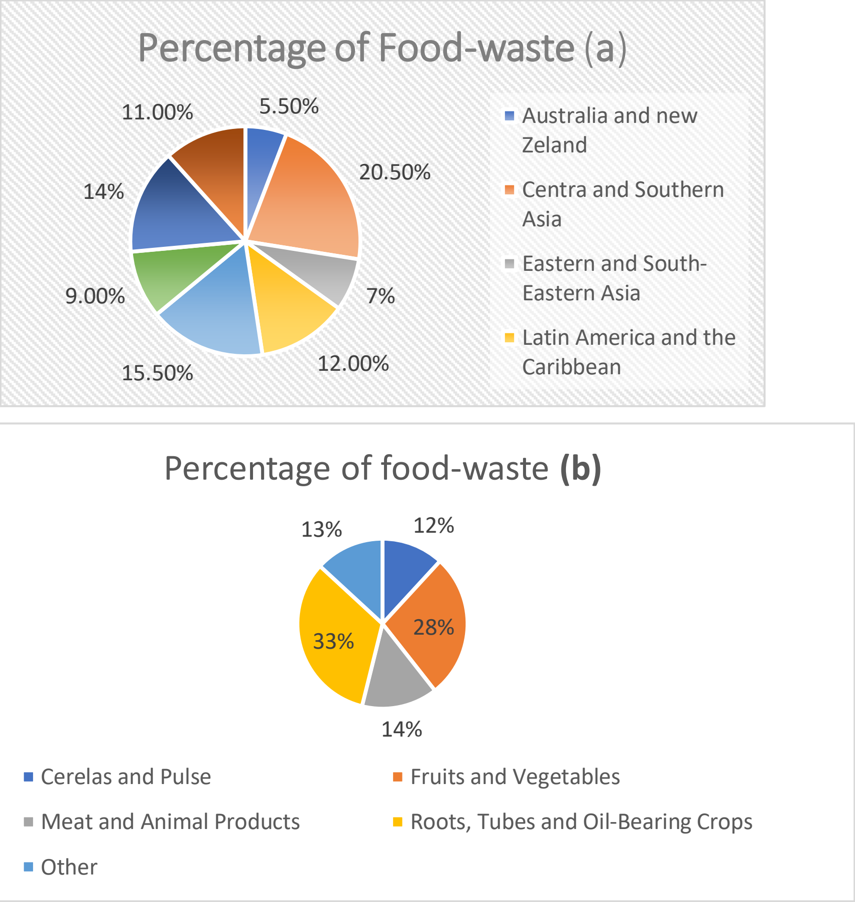


Figure 1. *Food loss from post-harvest to distribution in 2016, Percentage globally and by region. Food loss from post-harvest to distribution in 2016 (Graph a); Percentage by commodity group. (Graph b). Source: FAO, 2019*

Throughout the food supply chain, several efficient strategies can be applied. These actions, however, should not be addressed to a single part of the chain, since the efforts applied in one part affect the subsequent ones.

In low-income countries, measures should have a producer perspective, e.g. by improving harvest techniques, storage facilities, and cooling chain. In industrialized countries, measures, to avoid having a marginal role, should have a consumer perspective. Consumers need to be informed since their behaviour is the main responsible for high levels of food waste.

Packaging has a vital role to play in containing and protecting food as it moves through the supply chain to the consumer. It already reduces food waste in transport and storage, and innovations in packaging materials, design, and labelling provide new opportunities to improve efficiencies.

1.1 Food Packaging

Packaging surrounds, enhances, and protects the goods, from processing and, manufacturing, through handling and storage, to the final consumer (Robertson, 2006). Generally, four basic functions are attributed to packaging (Robertson, 2013). These four are interconnected and together allow to obtain functional packaging.

1. **Containment** refers to the fact that without containment food, such as free-flow food, cannot be moved to one place or another. The package must contain the product to function successfully. Without it, the product would lose in the environment, and pollution would be spread.
2. **Protection:** the package is a protective barrier capable of preserving the quality of the food. It protects from three major classes of external influences (physical, chemical, and biological).
3. **Convenience:** packaging plays an important role in meeting the demands of consumers, which requires more and more convenience. This function should take in consideration, other two aspects: the package should be produced into consumer-sized dimension (apportioning function of packaging), it refers to the fact that a package should contain a portion size that is convenient for the intended consumer; the package should allow a unitizing function, which is important during transportation since it permits to optimize the handling.
4. **Communication:** the package is the face of the product, so it may be designed to enhance the product image and/or to differentiate the product from the competition. A distinctive or innovative packaging can boost sales of a product in a competitive environment. The package also communicates important information to the consumers, such as how the product has been produced, whit which ingredients, which nutrients (macro and micronutrients) the food furnish and even how the dispose the packaging at the end of its used.

Materials designed to be in contact with food are called Food Contact Material (FCM). For a correct choice of the FCMs it is important to have adequate knowledge of the properties of the materials (chemical-physical, mechanical barrier, and optical properties), since they play a key role in the selection, always taking in consideration which type of food must be contained (Limbo and Piergiovanni, 2010).

Generally, properties are classified in chemical and physical properties (Table 1), and together these define the technological suitability or function of a material.

Table 1. *General properties of packaging material in food industry. source: Siracusa, 2016.*

Thermal properties	Mechanical properties	Barrier properties	Optical properties	Chemical properties
• Melting point temperature • Glass transition temperature • Heat of fusion (crystalinity/amorphous phase determination) • Heat capacity • Thermal conductivity	• Tensile strengh (stress/strain curve) • Tear strength • Bursting strength	• Permeability and gas/moisture transimission rate • Gas and moisture diffusivity • Gas and moisture solubility	• Transparency • Opacity • Gloss • Haze	• Atomic costituent • Chemical bonds between atoms • Intermolecular forces • Spatial arrangements • Biodegradability • Biotoxicity • Biofilm adhesion

In Europe, FCMs are governed by both national and European measures. Regulation (CE) n. 1935/2004 (framework standard) defines the general requirements to which all materials and objects must comply. In particular, the 3rd article of the Regulation, contains the general requirements for the packaging, it is specified that:

Materials and articles, including active and intelligent materials and articles, shall be manufactured in compliance with good manufacturing practice so that, under normal or foreseeable conditions of use, they do not transfer their constituents to food in quantities which could:

- endanger human health;
- bring about an unacceptable change in the composition of the food;
- bring about a deterioration in the organoleptic characteristics thereof.

The labelling, advertising and presentation of a material or article shall not mislead the consumers.

The framework provides:

- For special rules on active and intelligent materials (they are by their design not inert);Powers to enact additional EU measures for specific materials (e.g.

for plastic); The procedure to perform safety assessments of substances used to manufacture FCMs involving the European Food Safety Authority (EFSA);

- Rules on labelling including an indicator for use (e.g. wine bottle, a soup) or by reproducing the appropriate symbol;
- For compliance documentation and traceability.

For FCM, such as ceramic materials, regenerated cellulose films, plastics (including recycled plastic), as well as active and intelligent materials, there are specific EU measures. There are also specific rules on some starting substances used to produce FCMs (Limbo and Piergiovanni, 2010).

In packaging, an important issue is the migration phenomenon, which is a mass transfer from an external source into food by the sub-microscopic process. To ensure food safety, in many countries, regulations are made. For example, for plastic EFSA has established a migration limit. This limit specifies the maximum amount of substances allowed to migrate to food. Generally, if a material is considered safe for food contact, it receives a specific symbol, a wine glass and, a fork, which is the international symbol of “food safe”.

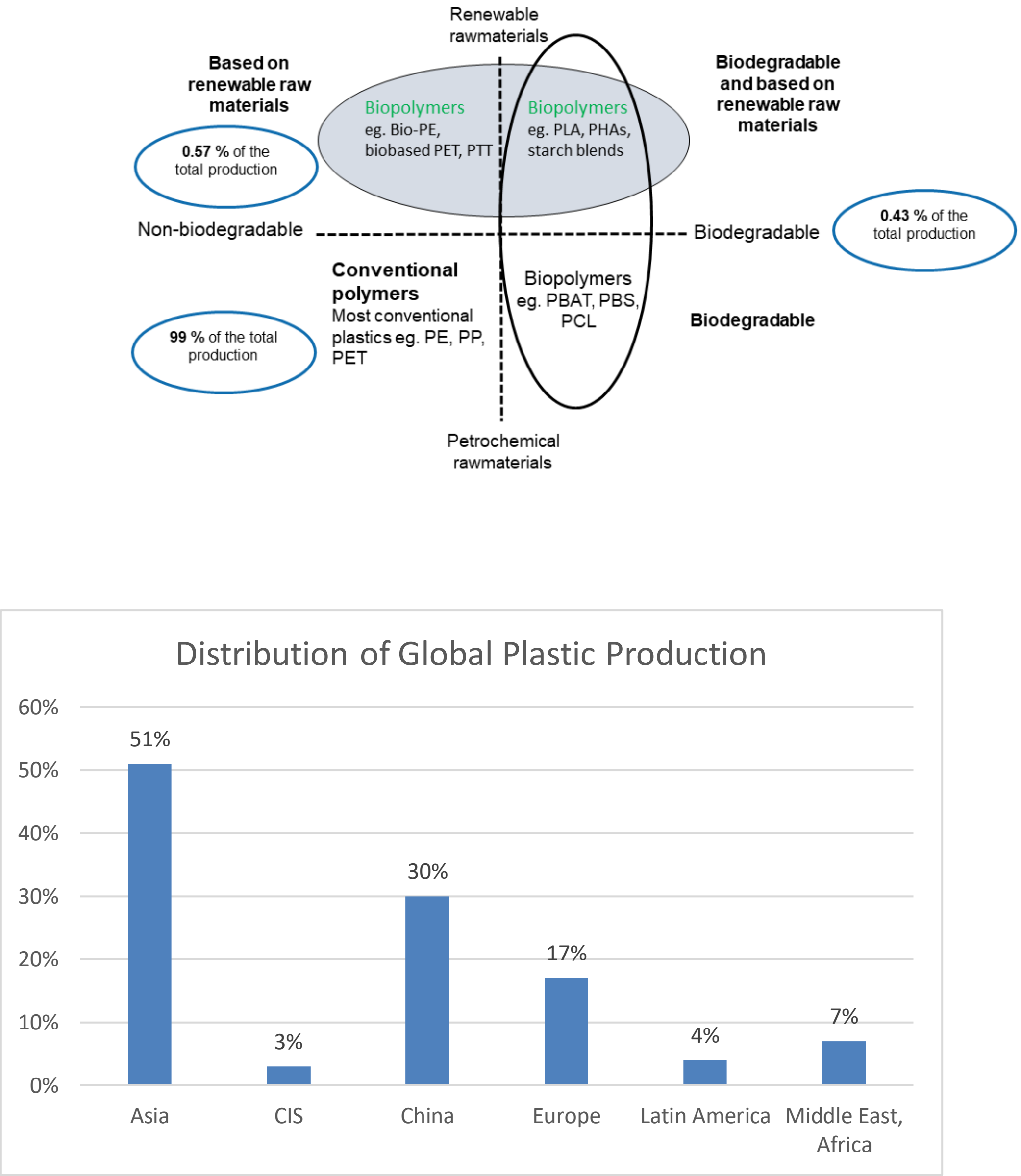


Figure 2. *a) Different type of polymers, biodegradable and non-biodegradable. Source: Endres and Siebert-Raths (2011). b) Distribution of Global Plastic production. Source: PlatisticsEurope, 2019.*

In 2018, global plastic production almost reached 360 million tonnes (Figure 2a) (Platistics*Europe,* 2019). Out of which, 61.8 million tonnes of plastics were produced in Europe (Platistics*Europe,* 2019). Of the total production convention polymers (polymers obtained from petrochemical-monomers) represent the 99% while the bioplastic has a total market share of only one percent (Bioplastics Market Data, 2019) (Figure 2**a***)*. This one percent is shared between biopolymers that are biodegradable and based on renewable raw materials and biopolymers which are based on renewable raw materials but are non-biodegradable. The biobased and biodegradable polymers only have a share of approximately 0.43% of the total mentioned one percent in the market while the rest of the produced biopolymers are mainly non-biodegradable (Figure 2a) (Plastics*Europe,* 2019; Bioplastics Market Data, 2019).

In 2018, plastic production almost reached 360 million tonnes (Plastics*Europe,* 2019). Packaging, in 2018, was the market with the highest demand for plastic, accounting to 39,9%. With the highest production numbers for Polyethylene (PE) (10M tonnes) and Polypropylene (8-9 M tonnes) (PP) which were mainly used in food packaging (sweet and snack wrappers, containers, hinged caps) or to produce trays, agricultural film, films, etc (Plastics*Europe,* 2019).

In 2019, Europe has strengthened its position among the global bioplastic producers, thanks to the research and development in this industry. Globally, Asia is the main producer, with 45 percent of the entire production. Its market is growing and will keep growing in the future thanks to more and more rising demand, products emerging, applications and biopolymers that are more sophisticated. In its annual report, European Bioplastic highlights that the materials with the highest relative growth are the innovative biopolymers, such as bio-based polypropylene (bioPP) and Polyhydroxyalkanoates (PHAs). The growing popularity of these materials must be attributed to their characteristics; these polyesters are 100 percent biobased and biodegradable and feature a wide array of physical and mechanical properties depending on their chemical composition. Bioplastics find application in a wide range of market, even though packaging remains the largest field of application with more than 53 percent (1.14 million tonnes) of the total bioplastic market in 2019. However, the portfolio of applications continues to diversify. Other reason behind the popularity of these products are the increasing demand of sustainable products from consumers and brands.

1.2 Primary Packaging Material

Traditionally, the most used materials for food packing were glass, metals (aluminium, foils and laminates, tinplate, and tin-fresh steel), paper, paperboards, and plastics.

Today, the range of materials used to produce packaging is wide. Nowadays, strategies are based on combinations of different materials to exploit functions or aesthetics.

Table 2. *Conventional packaging materials in food packaging: characteristics and applications. Source: Marsh and Bugusu, 2007*

Material	Characteristics/Properties	Applications
Glass	Amorphous Brittle and beaks Impermeable to gases and moisture Odourless Chemical inertia High thermal resistance Optically isotropic and transparent reusable and recyclable heavy and bulky low cost material costly to transport	Food and Beverage industry Chemicals, Pharmaceutics, and Cosmetics Industry. Alcoholic beverage packaging is the major application.
Steel, aluminium, tin and chromium	Low toxicity Resistance to high temperature Good flexibility Malleability and formability Surface resilience impermeable to moisture and gases resistant to corrosion cannot be welded limited structural strength lightweight good permeability recyclable economic	Food and beverages industry Health& beauty products Household and Industrial sector used as composites materials or as pure alloy Excellent physical protection and barrier properties to gases, moisture, light decorative potential
Plastics	Low cost (economic starting resource) Easy processability Chemically resistant Wide range of physical and optical properties Variable permeability Low molecular weight Flexibility Lightness Recyclable Reusable Crystalline or semi-crystalline structure	Packaging industry Building &Construction sector Automotive, Electrical &Electronic Industry Agriculture sector Household and sport industry.
P**aper**	Crystalline or semi-crystalline structure made from renewable resources Poor barrier properties Not heat sealable Recyclable Usually is treated, coated, laminated or impregnated with other materials to improve functional and protective properties low cost	Food and Beverages Industry Paper and paperboard are used in corrugated boxes, milk cartons, folding cartons, bags and sacks, and wrapping paper. tissue paper, paper plates, and cups.

Even though these conventional materials present diverse properties that make them suitable for different applications (Table 2), it is important to understand that they have also a negative impact on the environment. Most packaging is designed as single-use and are typically thrown away rather that been reused or recycled (Bodamer and David, 2016). Yet, their physical structure results in preservation on much longer time scale. The trouble with food packaging begins as its creation. Each form of packaging uses lot of resources such as, energy, water, chemicals, petroleum, minerals, wood and fibres to produce. For instance, the manufacturing of paper/paperboard produced large volume of toxic wastewater (National Academics of Sciences, 2019), aluminium production creates a toxic sludge that is caustic and may contain radioactive elements or heavy metals, making its management complicated. The pollution of the seas and oceans is increasing due to the amount of plastic waste generated each year. According to the European Commission in the European Union (EU) 150-500 thousand tonnes of plastic litters enter the oceans every year. This represents only a small portion of global marine litter. In Europe the plastic waste ends up in vulnerable marine areas, such as the Mediterranean Sea and parts of the Arctic Ocean. Other than harming the environment, plastic wastes cause economic damage to activities such as tourism, fisheries and, shipping.

Packaging materials can be harmful for the environment and human health once discarded. Due to their characteristics, plastics packages are responsible for negative externalities such as greenhouse gas emission or ecological damage (Nkwachukwu et al., 2013).

The practice of burning plastic waste leads to the release of pollutants, which can be transported through the air and deposited onto land or into water. These pollutants (such as dioxin, mercury, and furans) persist for long periods with the tendency to bio accumulate. Problems associated with bioaccumulation include cancer, deformed offspring, reproductive failure, immune diseases, and subtle neurobehavioral effects. Not only the wildlife is exposed to these problems but humans too because of the consumption of contaminated fish, meat and dairy products (Nkwachukwu et al., 2013).

Incineration of plastic involves air emission of sulphur dioxide, nitrogen dioxide, chlorine, dioxin, and fine particulates and emission of greenhouse gases of CO_2 and nitrous oxide (N_2O). Also, the ashes which remain after incineration need to be disposed of. Most of these gases are toxic (i.e. interfering with the normal biochemical processes of the body or exclude O_2 from the victim) (Nkwachukwu et al., 2013).

1.3 Biodegradable Packaging Material

An interesting alternative to the use of plastics are bioplastics. The term bioplastic does not refer to only a singular material, but it refers to a whole family of materials with different properties and applications. According to European Bioplastics, a plastics material is defined as bioplastic if it is either biobased, biodegradable, or features both properties. Biobased packaging materials are made from renewable raw materials and can be classified according to their origin.

The term biobased means that the material or product is (partly) derived from biomass (plants). It is important to highlight that not all biomaterial is biodegradable, since biodegradability is a property that depends on the chemical composition of the polymers. In particular, the type of chemical bond defines whether and in which time the microbes can degrade a material of interest (Siracusa, 2018).

Bioplastics however represent a series of advantages (Chen, 2014), which are:

- Lower carbon footprint. because the biopolymer/plant used to produce the bioplastic capture the CO_2 during the photosynthesis process and once degraded the sequestration is reversed.
- They do not use scarce crude oil.
- Increase source efficiency: as the biomass can be cultivated on an annual basis.
- Saving fossil resources and replacing them step by step.
- Reduce the amount of litter produced and favour composting.

There are other advantages related to the use of bioplastics to produce packaging materials, but these depend on the polymer and process used. For example:

- Improved printability, the ability to print a highly legible text or image on the plastic;
- Bioplastics can be engineered to offer a much more acceptable surface feel than conventional plastics.
- Less likelihood of imparting a different taste to the product contained.
- A bioplastic may have much greater water vapor permeability than standard plastic.
- Bioplastics can be made clearer and more transparent.

Despite the mentioned advantages, certain disadvantages limit the use of bioplastics. First, the land, used to cultivate the biomass, is a major hurdle in their functionality success of these materials. Properties of certain bioplastics like thermal instability, difficult heat saleability, brittleness, low melt strength, high water vapor, and oxygen permeability of PLA limit their use as films in food packaging applications (Rhim et al., 2009). Due to their hydrophilic nature biopolymer obtained from starch or cellulose possess a low water vapor barrier, which is responsible for poor processability, brittleness, vulnerability to degradation, limited long-term stability and poor mechanical properties (Cyras et al., 2007).

These disadvantages, on the other hand, have led to new research to improve the functionality of biomaterials.

“biobased does not equal to biodegradable”.

For almost every conventional plastic material and corresponding application there is a bioplastic alternative, since bioplastics have the same properties as conventional plastics. Among the properties that these materials possess the most important are the biodegradability and compostability properties. Both these properties describe the breaking down of the organic materials in a specific environment. According to European Bioplastics, biodegradation is a chemical process in which materials are metabolized into carbon dioxide (CO_2), water and biomass with the help of microorganisms. Composting is a type of enhance biodegradation under managed environmental conditions, such as temperature, humidity, and microorganism present (Kale et al., 2007). It is also referred as aerobic biodegradation. When speaking of biodegradability, it is important to specify the environment where the process is happening as everything is biodegradable given time.

Biobased and/or biodegradable packaging materials can be classified into four categories according to their method of production (Figure 3):
Category 1: Polymers extracted from biomass;
Category 2: Polymers produced by chemical synthesis from biomass monomers;
Category 3: Polymers produced directly by natural or genetically modified organisms;
Category 4: Polymers whose monomers are obtained from petrochemical-based monomers.

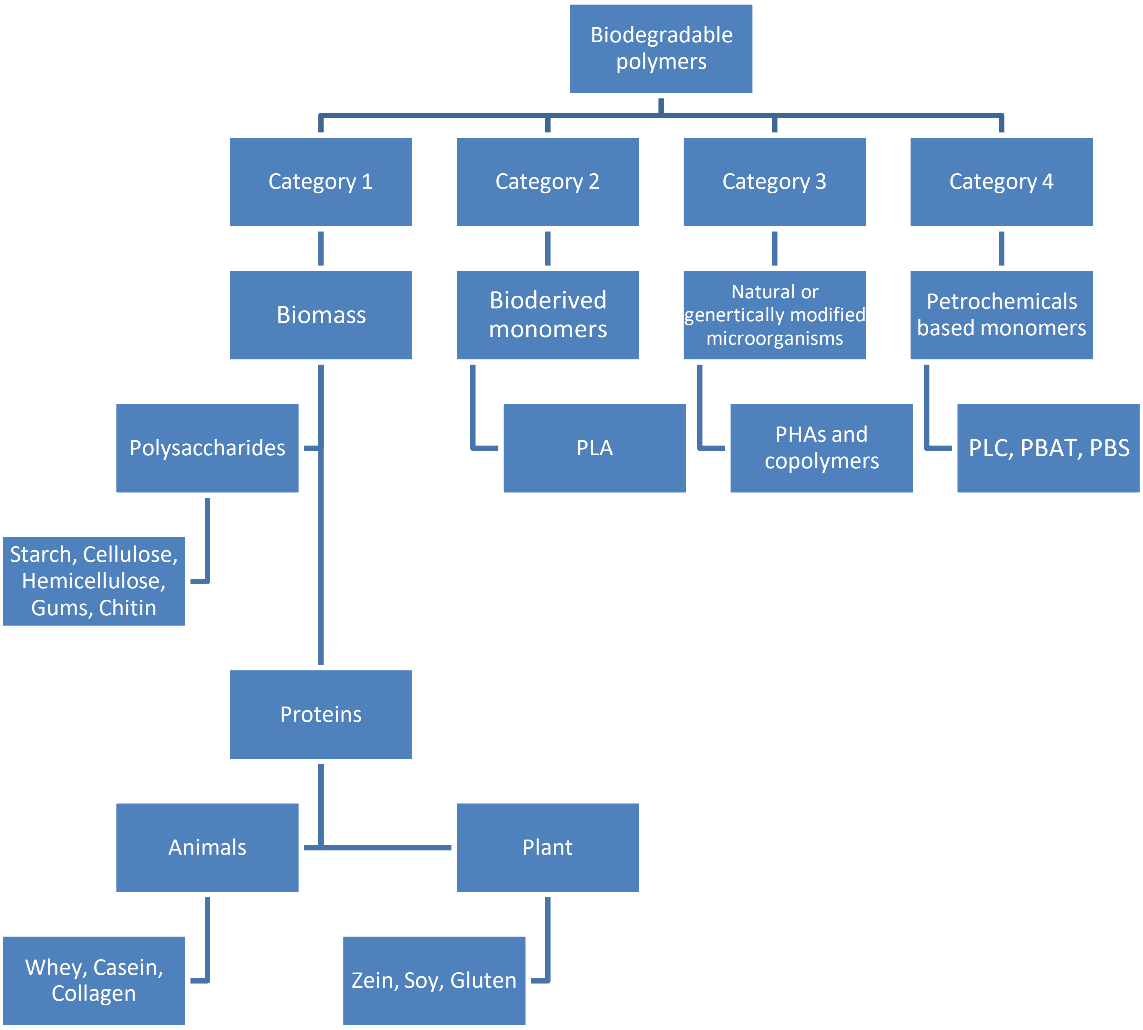


Figure 3. *Classification of biodegradable packaging materials based on their origin and method of production. Source: Robertson, 2016*

The first category of bioplastics include polymers that are extracted from marine and agricultural products (Table 3*)*; examples are polysaccharides such as cellulose, starch and chitin and proteins such as collagen and soy. Usually, they are used alone or in a mixture with synthetic biodegradable or biobased polymers. Due to their properties, such as high crystallinity and strong intermolecular interactions, these materials cannot be processed easily using shear and heat. To increase the molecular mobility, a plasticizer such as water, glycerol or lactic acid are used.

Table 3. *Characteristics of biopolymers: properties, production, biodegradability and compostability conditions: Source: Folino et al., 2020.*

Biomaterial	Properties	Production	Biodegradable	Compostable
Characteristics of biopolymers extracted from biomass				
Starch and derivates	Semi-crystalline polysaccharides extracted from tubes or/and cereals	To improve barrier properties and mechanical properties starch is modified chemically and plasticizers are used as reinforcement. Bioplastics based on starch are produced by using blend (starch + hydrophilic petroleum -based polymers)	Yes	Yes Home composting: ≤35 ◦C Industrial Composting:50–60 ◦C pH: 5.5-8 t: 90-150 Days U: % p/p > 50
Cellulose	Linear polysaccharides Crystalline structure insoluble in water and organic solvent	Different derivates can be obtained by dissolution of insoluble cellulose: - regenerated cellulose film - cellulose acetate - micro fibrillated cellulose	Yes 100%	Yes Home composting: ≤35 ◦C Industrial Composting:50–60 ◦C pH: 5.5-8 t: 90-150 Days U: % p/p > 50
Protein	Amino acid biopolymers High permeability to water Low permeability to oxygen	Properties are modified by using plasticizers or by blending with other materials. Difficult to process due to their properties	Yes	Home composting: ≤35 ◦C Industrial Composting:50–60 ◦C pH: 5.5-8 t: 90-150 Days U: % p/p > 50
characteristics of polymers obtained from bioderived monomers				
Poly (lactic acid) (PLA)	Linear, aliphatic polyester Optical copolymer Amorphous or semi-crystalline structure Good mechanical properties High thermal plasticity	Synthesized from lactic acid monomers. Lactic acids are produced by the fermentation of glucose (from biomass (corn, wheat, etc) or from lactose	Yes 60-70%	Industrial Composting:50–60 ◦C pH: 5.5-8 t: 90-150 Days U: % p/p > 50
Biopolyethylene (BioPE)	Has the same characteristic of conventional PE and can be use in the same application	Obtained by the dehydration of bioethanol which is made by the fermentation of various feedstocks It can be used to produce all the PE types: High-density polyethylene (HDPE), Low-density polyethylene (LDPE) and linear low-density polyethylene (LLDPE).	Can be recycled but is not biodegradable	No
Biopoly (ethylene terephthalate) (bioPET)	Has the same characteristic of conventional PET	The monomers used for its production is the terephthalic acid which can be obtained by fermentation of different biomass	Can be recycled but is not biodegradable	No
Characteristics of polymers obtained from naturally or modified organism				
Poly(hydroxyalkanoates) (PHAs)	Microbial polyester Optically active Linear aliphatic polyester	Produced in the form of intracellular particles by commonly found microorganism	Yes	Yes Home composting: ≤35 ◦C Industrial Composting:50–60 ◦C

				pH: 5.5-8 t: 90-150 Days U: % p/p > 50
Bacterial cellulose (BC)	Is identical to the cellulose produced by plant but whit the advantages that is not combine with lignin, hemicellulose, pectin.	Produced by bacteria belonging to the genera *Acetobacter, Agrobacterium, Alcaligenes, Gluconaceto bacter, Rhizobium or Sarcina*	Yes	Home composting: ≤35 ◦C Industrial Composting:50–60 ◦C pH: 5.5-8 t: 90-150 Days U: % p/p > 50
Characteristics of polymers produced from petrochemicals monomers				
Poly(caprolactone) (PCL)	Flexible Aliphatic semi-crystalline polyester Miscible with other polymers	Obtained by the ring-opening polymerization of ε-carpolactone	Yes	Home composting: ≤35 ◦C Industrial Composting:50–60 ◦C pH: 5.5-8 t: 90-150 Days U: % p/p > 50
Poly (butylene adipate-*co*-terephthalate) (PBAT)	Aliphatic-aromatic copolyester Mechanical properties are similar to PE	Synthetized from 1,4-butanediol, adipic acid by adding special additives and optimizing the processing conditions, transparent cling films can be obtained	Yes	Yes Home composting: ≤35 ◦C Industrial Composting:50–60 ◦C pH: 5.5-8 t: 90-150 Days U: % p/p > 50
Poly (butylene succinate) (PBS) and Copolymers	Copolymers Balanced mechanical properties	Synthetized by polycondensation of succinic acid and butanediol	Yes 80%	Home composting: ≤35 ◦C Industrial Composting:50–60 ◦C pH: 5.5-8 t: 90-150 Days U: % p/p > 50

Biodegradable polymers have not been extensively utilized in food packaging applications due to their limitations in certain properties, processability and due to the high production costs (Robertson, 2016). The cost of bioplastics is much higher than that of conventional polymers. Taking PLA as an example, the cost is 3-5 times more than the cost of PET (Vimal K., 2017). The advantages of bioplastics are meaningless if the material is too expensive. One way of reducing the cost is the use of technologies that can help to minimize the energy required for material synthesis and processing. An example is recycling, which is an efficient method of energy minimization that releases less CO_2 than producing a new product; by doing so, it is possible to reduce the extensive utilization of a limited feedstock and replace it with low cost recycled material (Vimal K., 2017). In terms of properties, biodegradable plastics exhibit low thermal stability and decreased mechanical and barriers properties, which reduces material applicability (Bonhomme et al, 2003). To enhance the use of biodegradable polymers in packaging applications, these can be reinforced with various materials to produce biocomposites. The reinforcing materials can be particles or fibrous. The incorporation of reinforcing materials improves barrier, mechanical and thermal properties, which ultimately improves the material processability (Vimal K., 2017).

1.4 Biodegradation process

Biodegradation is the breaking down of polymers by the action of microorganisms. The process is heavily influenced by the chemical and physical properties of the material (Tokiwa et al., 2009). These properties are the surface characteristics (hydrophobic or hydrophilic, surface area), the first-order structures (molecular weight, molecular weight distribution, chemical structure), and the higher-order structures (crystallinity, crystal structure, modulus of elasticity, glass transition temperature, melting temperature) of polymers (Tokiwa et al., 2009).

Depolymerization and mineralization are the two key steps that characterize the biodegradation of polymers. During the depolymerization process, the long polymeric chains are broken down because of temperature, water, and sunlight to shorter oligomers, dimers or monomers. These monomers pass through the cell walls of microorganisms to be used as a substrate. Once in the cell, they are degraded by the action of microbial enzymes. Two main types of enzymes are involved in microbial depolymerization processes: extracellular and intracellular depolymerases (Shah et al., 2008). Extracellular enzymes act outside the cells to break the longer units down into shorter molecules, preparing them for further degradation by intracellular enzymes.

The final products of the mineralization process are minerals, salts, water, and gases such as CO_2 and CH_4. The CO_2 generated from the renewable carbon sources, once released it is submitting to photosynthetic fixation generating renewable carbon again. Thanks to this cycle the carbon flux in the synthesis and degradation of polymer is balanced. So, it presents the advantage to not accumulate in the atmosphere.

1.4.1 Biodegradation under anaerobic conditions

The biodegradation can occur in two ways, aerobically and anaerobically, this offers two types of biological waste treatment. In aerobic degradation, the energy stored in the organic matter is released as heat. To avoid that the high temperature inhibits the growth and activities of the microorganisms and to provide oxygen the biomass is turned. In anaerobic degradation, the energy stored in organic matter is mainly released as methane, and due to the lack of oxygen in the process, lees heat and less microbial biomass are produced.

Anaerobic digestion is a microbiological process that can be divided into four main steps: hydrolysis, acidogenesis, acetogenesis, and methanogenesis. These are performed by the combined action of three physiological groups of microorganisms: hydrolytic-acidogenic bacteria (and most likely fungi), syntrophic acetogenic bacteria and methanogenic archaea.

- Hydrolysis/Acidogenesis: in this step polymeric compounds are broken into soluble monomers (lipids, proteins, carbohydrates, etc) by the action of extracellular enzymes (either secreted to the bulk solution or attached to the cell wall). This phase is influenced by the structure of the polymer compound.
- Acetogenesis and Syntropy: acetogenesis is performed by acetogens and is characterized by the reduction of carbon dioxide (CO_2) to the acetyl moiety of acetyl-co-enzyme A (CoA) through the acetyl-CoA pathway. Acetogens can use a wide variety of carbon sources, electron donors and acceptors and grow as autotrophs or heterotrophs. The reactions can only proceed if the partial pressure of these products is kept low, for example through consumption by methanogens. This type of symbiosis, that exists between acetogens and methanogens, is called syntropy.
- Methanogenesis: in this final step methanogens catalyse the conversions of the products obtained in the acidogenesis/acetogenesis into methane. Based on the substrate and pathway used, methanogens can be classified into hydrogenotrophs and methylotrophs. The first one used formate or hydrogen as an energy source, and CO_2 is reduced to methane. Methylotrophs, instead, are more versatile in terms of substrates that they can use. Example are hydrogen, CO_2, acetate, methanol, and methylamines.

To obtain an efficient biogas production all the microorganisms must work in a synchronized manner. This is possible if all the requirements of all the agents involved are satisfied. The biomass used as feedstock has an impact on the process as well as contains different molecules that can inhibit the growth of the microorganisms.

During the process different parameters can be controlled such as temperature, stirring period, contents of organic acids, pH, alkalinity, hydrogen concentration, the volume of gas formed, and gas composition. Through these is possible to improve the production of biogas.

1.4.2 Assessing biodegradability

To assess the biodegradability of biopolymers and organic matter different protocols have been developed. These protocols are based on tests that are performed under controlled conditions to determine if a product is biodegradable or not. The aerobic biodegradability is measured as the amount of CO_2 produced from the conversion of a carbon substrate because of a microbial attack. The amount of CO_2 evolved is quantitatively confronted to the value reached by cellulose, taken as a control in the same operating conditions and time (Robertson, 2016). Example of protocol is the ISO 17088 *Specification for compostable plastics* that evaluates the biodegradability of materials under laboratory conditions and not in the real commercial conditions. According to the ISO 17088:2012 standard, plastic can be considered biodegradable if a significant change in the chemical structure (organic matter is transformed in carbon dioxide, water, inorganic compounds, and biomass) not resulting in visible or toxic residues under composting conditions. Another example is the European standard ENI 13432 *Requirements for packaging recoverable through composting and biodegradation- test scheme and evaluation criteria for final acceptance of packaging* is based on a reaction with water. The evaluation of the biodegradation, measured as CO_2 produced, is made after 90 days, under compositing condition.

The European standard 13432 also defines the minimum requirements that packaging must meet in order to be processed by industrial composting. EN 13242 requires, disintegration (fragmentation and loss of visibility in the final compost), biodegradability (is the capacity of the compostable material to be converted into CO_2 under action of microorganisms),absence of negative effects on the composting process, the amount of heavy metals, and the final compost must not be affected negatively (no reduction of agronomic value and no Eco-toxicological effects on plant growth).

If a packaging satisfies these requirements it can be certificated with the standard. Plastics certified according to EN 13432 can be recognised by the “seedling” logo.

The Biochemical Methane Production Potential (BMP) is a parameter used to determine the biogas potential of solid organic substrates (biowaste, energy crops, agricultural waste, manure, etc). In an industrial context, it is also used for assessing design, economic and managing issues for the full-scale implementation of the anaerobic digestion process.

The result of a BMP test is the methane or biogas produced from a given weight of a certain substrate. Different techniques can be used to measure the gas production: volumetric methods (typically acidic water displacement), manometric (determination of pressure variation by transducers), gas-chromatographic methods (flame ionization (FID) or thermal conductivity detectors (TCD).

The gas-chromatographic method requires the collection and injection of a sample volume of gas (e.g. 100 µL) collected with a gas-tight syringe into the GC. The gas-chromatograph will produce peaks based on the amount of methane injected. The obtained peak area should be compared with the peak obtained from the injection of a standard gas mixture of known composition. The volume of methane produced is obtained by multiplying the headspace volume by the (%) of CH_4 in the headspace as determined by GC analysis.

When presenting the BMP result it is important to give a clear description of inoculum source, activity, and volatile solid (VS) or volatile suspended solid (VSS) content, medium composition, waste (water) composition or description, and dilutions used.

BMP tests can be used to obtain further information on the substrate studied like the hydrolysis rate if hydrolysis is limiting the anaerobic conversion process.

Biogas can be produced from the organic fraction of any material, such as crop residue, textile wool, industrial food waste, fruit waste, etc. Today, in biogas facilities the tendency is to use feedstocks that are easily utilizable by the microorganism responsible for the anaerobic digestion process. There is this tendency because the characteristic of the feedstock (such as availability, composition, and degradability) affect the entire biogas production, leading to a low biogas yield, high retention time, and high investment cost. Anaerobic digestion includes different steps. Hydrolysis presents a low rate of degradation for hard-to-digest biomass (Fernandes et al., 2009) such as lignocellulose and keratin-rich waste. Moreover, some toxic by-products can be formed during this step (Nerves et al., 2009). In the Acidogenesis if the feedstock has a low buffering capacity and the organic loading rate is high, the accumulation of volatile fatty acids can result in a pH drop, which would inhibit the methanogens. The final step, methanogenesis, is the limiting step for easily degraded and those with low buffering capacity feedstocks (Rozzi and Remigi, 2004).

The major reasons why some feedstocks are not suitable for biogas production are: they cannot be digested by microorganisms, digestion by microorganisms is difficult to achieve, digestion could be achieved but in a very slow way, and the presence of inhibitor or their production during degradation of the biomass.

To facilitate the digestion different pre-treatments can be applied. For instance, indigestible materials can be treated by a combined gasification-fermentation process. Moreover, hard-to-digest compounds such as recalcitrant lignocellulose or keratin can be pretreated by thermal, chemical, physicochemical, or biological methods. Feedstocks which contain inhibitor can be subjected to a steam explosion, or non-pre-treatment methods developed to facilitate their digestion, such as innovative digester and/or bio-augmentation (Patinvoh et all, 2017).

1.5 Food packaging in municipal waste system

The life cycle of plastic packaging products includes different steps, which are production, converters demand, manufacture, and consumption. At the end of their life, the end-user products become waste, which is collected and treated. Post-consumer waste can be recycled, use for energy recovery, or discarded in landfills. According to the report made by Plastics*Europe*, in 2018 only 29.1 Mt of plastic waste was collected, out of which 32.5% was recycled, 42.6% was used for energy recovery, and 24.9% was deposited in landfills.

Typically, plastics-based materials are recycled mechanically. However, this process cannot be applied to all the materials. Other options are pyrolysis process (to obtain syngas or liquid fuel), feedstock recycling, or chemical recycling (depolymerization of plastic to obtain monomers).

Mechanical recycling includes, sorting, shredding, washing, and drying, and at the end the material can be melted (to obtain pellet) or products directly. In Europe the recycling rate of plastic packaging is between 26-52% (Plastic*Europe, 2019*). This wide range is the result of differences in collection schemes, available infrastructure, and consumer behaviour.

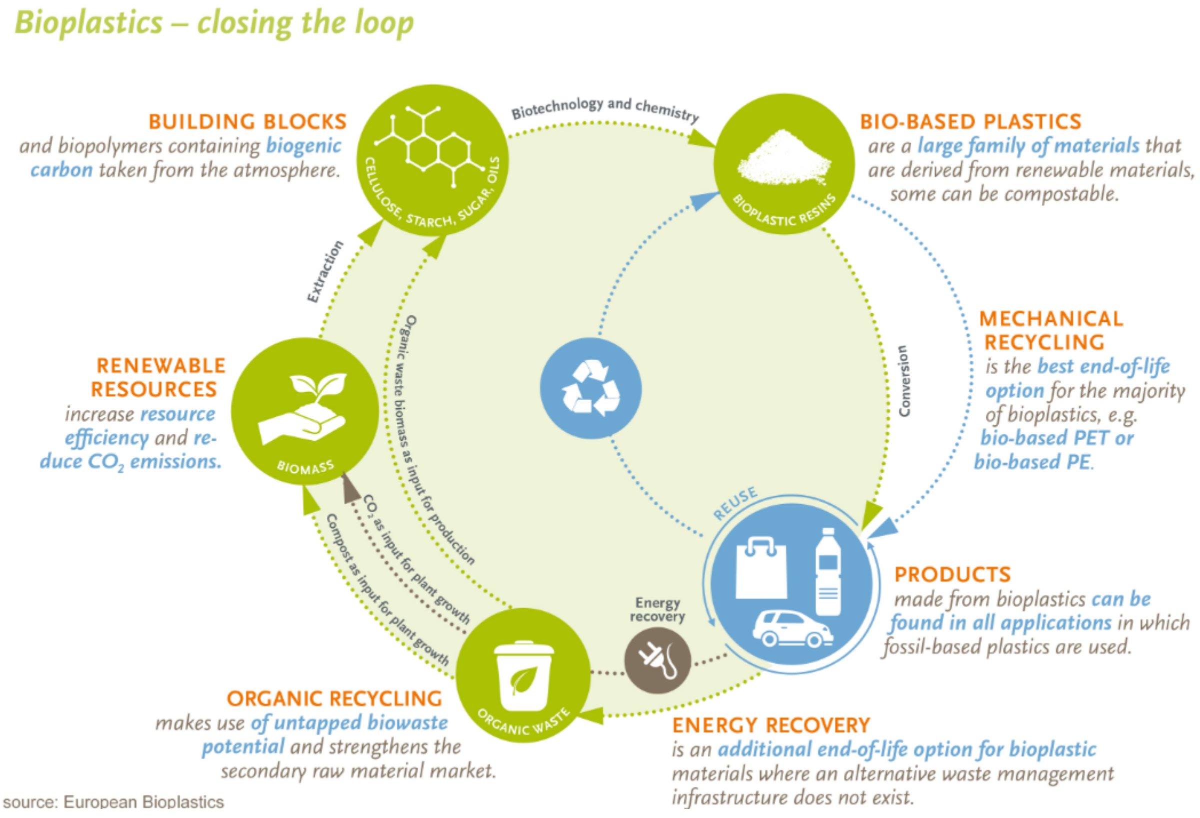
The mechanical recycling process is influenced by different challenges. The key challenge is the quality of the recovered and recycled material. In many cases post-consumer waste presents a low level of purity because plastics are composites materials (mixed with different plastics), laminated with multiples layers, present additives or fillers or they can be contaminated in the recovery process. To obtain a cleaner recycled plastic product is important to improve the collection, recovery and separation technology, and above that create dedicated recovery system for all the types of plastic. Materials such as PET and HDPE present higher recycling rates if confronted with films.

Another important aspect is the design of the packaging. As mention before packaging products are obtained using different polymers and varieties of additives, and fillers, which are a barrier to recycling. So, improving the design process may reduce this variety and enable better recycling.

To improve separation efficiency and effectiveness and to handle mixed plastic wastes to still produce a high-quality recycled product new technology will be needed.

1.6 Sustainable food packaging in a circular economy context

In the circular economy context, the European Commission aims to reduce the generated waste in general to a minimum amount. This is possible if the materials, composing a product, instead of being discarded is kept within the economy wherever possible. In this context what used to be considered as “waste” is now seen as a valuable resource. Bioplastics are materials that are designed to have the same properties and functions of conventional plastics but with the advantages to reduce the environmental impact and to improve the waste management. So, in a circular economy context, bioplastics can play an important role to make this vision into a reality.



source: European Bioplastics

Figure 4. *Bioplastics in a circular economy context. Source: European Bioplastics*

According to European Bioplastics (Figure 4), at the end of their life, bioplastics are suitable for different options. These options are mechanical recycling (product discarded finds a new applications), organic recycling (bioplastics and biowaste are used as feedstocks in biogas facilities), and energy recovery (bioplastics are used to create renewable heat and electricity through incineration).

What make bioplastics suitable for this economic model are their properties and characteristics, the biodegradability and compostability capacity. Between the different end-of-life options that a circular economy loop offers, the organic recycling is the one that really takes in consideration these characteristics. In order to be suitable for industrial composting or Anaerobic Degradation (AD), these products need to meet specific requirements (EN 13432 or waste directive such as 94/62/EC,) (European Bioplastics, 2017).

Organic recycling considers the possibility of recycling the biomaterials together with the organic content. This is an important advantage since biobased packages are capable of stimulate the microbial activity during composting process. At the same time, they could help to solve the problematics related to the sorting of packaging and reduce the amount of waste generated.

1.7 Anaerobic digestion of Bioplastics

Bioplastics have the ability to biodegrade under different environments (Voinova et al., 2007). Most commonly examined degradation environments were aerobic, soil, fresh, or marine water (Shruti et al., 2019). Folino et al., in 2020 published an article named "Biodegradation of Wasted Bioplastics in Natural and Industrial Environments: A Review". The review investigates both the extent and the biodegradation rates under different environments and explored the state-of-the-art knowledge of the environmental and biological factors involved in biodegradation. The article includes tables in which are summarized the studies carried out on bioplastics' degradation in different environment conditions. Approximately 83% of the performed experiments took place in aerobic environments and the 43% of those experiments presented an extent of biodegradation over 60%. Most of the aerobic-condition experiments took place in compost (39%), followed by soil (34%), and aquatic (27%) environments. Only 17% of the total experiments took place in anaerobic conditions. More than half of the bioplastics studied in anaerobic environment achieved over 65% of biodegradability (Folino et al., 2020).

Table 4 summarizes several studies carried out on bioplastics' degradation in anaerobic condition. PHB was found to be suitable for AD as 90% of the material was degraded in nine days of digestion in the anaerobic sludge, while PBS remained stable under the same conditions (Yahi et al., 2014). PHA-based polymers generally showed a biodegradability over 75% in less than two months (Hedge et al., 2019), while starch-based biopolymers showed a lower performance (Calabrò et al., 2020) compared to PHA. PHA bioplastics were well degraded under both aerobic ad anaerobic environment (Bàtori et al., 2018). Moreover, the use of biopolymers such as PHA and thermoplastic starch can improve the biodegradation rate of anaerobic digester as they fast degrade into polylactic acid (Hedge et al., 2019).

Table 4. *Studies carried out on bioplastics' degradation in anaerobic conditions. Source: Folino et al., 2020*

Source of Bioplastic	Name of Bioplastic		Type of Environment	Conditions	Scale	Biodegradation Indicator	Biodegradability (%)	Period of Biodegradability (Days
Bio-based	PLA-based	PLA	Sludge	Anaerobic, 37 ◦C	Lab-scale, 10 L	Produced CO2	29–49	277
		PLA	Sludge	Anaerobic, 55 ◦C	Lab-scale, 10 L	Produced CO2	80	30–50
		PLA	AD	Anaerobic, 52 ◦C	Lab-scale	Comparison with respect to theoretical BMP	90	36
		PLA powder	Sludge	Anaerobic, 55 ◦C	Lab-scale	Biogas production	90	60
		PLA	Sludge	Anaerobic, 55 ◦C	Lab-scale	n.a.	75	75
		PLA	Sludge	Anaerobic, 55 ◦C	Lab-scale, 10-L bottle	Produced biogas	85	60
	PHA-based	PHBs	AD	Anaerobic digestion—untreated PHB—35 ◦C	Lab-scale	Conversion to biogas	67	175
		PHBs	AD	Anaerobic digestion—untreated PHB—35 ◦C	Lab-scale	Conversion to biogas	91	175
		PHB	Sludge	Anaerobic, 55 ◦C	Lab-scale	Produced biogas	90	14
		PHB	Sludge	Anaerobic, 37 ◦C	Lab-scale, 10 L	Produced CO2	90	9
		PHB	AD	Anaerobic	Lab-scale	Weight loss—Biogas production	90	9
	Starch-based	Plastarch	AD	Anaerobic, 37 ◦C	2 L laboratory scale batch reactor	Produced CO2	26.4	50
		Mater-Bi plastic carrier bags	AD	Untreated bioplastic—35 ◦C	Lab-scale 1-L bottle	Weight loss	23–30	15–30
		Mater-Bi plastic carrier bags	AD	NaOH pretreated bioplastic—35	Lab-scale 1-L bottle	Weight loss	73	15
		Mater-Bi plastic carrier bags	AD	Untreated bioplastic—55 ◦C	Lab-scale 1-L bottle	Weight loss	28–41	15–30
Petroleum-based	PBS-based	PBS	Landfill	Anaerobic, 25 ◦C	500 mL glass bottle	Produced CO2	2	100
	PCL-based	PCL/starch	Landfill	Anaerobic, 25 ◦C	500 mL glass bottle	Produced CO2	83	139
		PCL	Sludge	Anaerobic, 37 ◦C	Lab-scale, 10 L stainless steel bottle	Produced CO2	3–22	277
		PCL	Sludge	Anaerobic, 55 ◦C	Lab-scale, 10 L	Produced CO2	75	40–75

2. MATERIALS AND METHODS

2.1 Feedstock component

2.1.1 Biobased packaging materials

For the experiment different biodegradable packaging materials were taken in consideration. The criteria used for the selection of the materials were: 1) the material need to be biobased (obtained from a renewable resource and/or produced by microorganism and/or bioderived monomers, etc.), 2) the material need to be compostable and biodegradable, if possible according to the provider, 3) the material has the potential to be used in food packaging and for food disposal and transportation.

The BMP of these materials was tested at two different times. In the first batch, three cellulose-based and two polysaccharide-based packaging materials were used (Table 5). The five biodegradable packaging products included a box (eggs box container), one cup, and three bags (two food-waste bags and sacks and a bag for the handling of food products). They were tested alongside non-biodegradable controls PET-plate and a thin plastic film (PP based). For the second part, corn-starch cutleries (fork, spoon and knife) and a sugarcane bagasse fibre plates have been added to the previously selected materials.

Each product was cut manually (with the help of a ruler and a cutter and/or scalpel) into 0.5*0.5 cm squares. The decision to shape the materials into tokens was made to increase the contact surface with the inoculum and to make sure to fit the materials inside the BMP bottles (60 ml and 1L, first and second batch, respectively).

2.1.2 Kitchen waste and Inoculum

The food-waste used as feedstock for the microorganism was obtained by collecting the kitchen waste in the household of the two researchers involved in the experiment. At the household food waste was added the content of used coffee filters.

The food waste was minced using a compact food processor (Bosch MCM3100WGB MultiTalent 3).

The inoculum was digested mesophilic municipal sewage sludge from the Viinikanlahtu sewage treatment plant (Tampere, Finland). The BMP of the inoculum was also determined separately along with the biomaterial and compared with those of the materials.

Table 5. *Bio-based packaging materials used in the BMP tests.*

Category	Products	Bio-degradability	Compostability	Polymer	Characteristics
Cellulose-based products	Cup Manufacturer: Bioware Bought at: Prisma (supermarket)	Yes	Yes According to Standard EN 13432	made from paperboard sourced from certified sustainably managed forests.	//
	Biowaste paper Bag Manufacturer: Rainbow Bought at: Prisma (supermarket)	Yes	Yes	made from paperboard	Dimension: 30x 22 x 12 cm
	Box Manufacturer: Kultamuna Bought at: Lidl (supermarket	Not given	Not given	made from paperboard	packaging container for a pack of 12 eggs The packaging is recyclable, but the label doesn't give information about its biodegradability or compostability
Biodegradable plastics	Plastic bag and sack (Bioska-506 bio film) Manufacturer: PlastiRoll Bought at: Prisma (supermarket)	Yes	Yes According to Standard EN 13432	coextruded biofilm. Blend made of starch and Polyvinyl alcohol (PVA)	Thickness: 14 to 18 mm Dimension: 390x500 home compost bag
	Plastic bag and sack (Bioska +) Manufacturer: Plastiroll	Yes	Yes	coextruded biofilm. Blend made of starch and PVA	Thickness: 18 to 20 mm Dimension:420x500 home compost bag that are made of a new faster compostable 506 biofilm

	Bought at: Prisma (supermarket)		According to Standard EN 13432		
Synthetic plastics	Plastic container Bought at: K-market	No	No	Made of Polyethylene terephthalate	//
	Thin plastic film Bought at: K-market	no	no	Made of PP	Thin film used to wrap gamma IV leaf products
Plant-based products	Cutleries (Knife) Manufacturer: Fuyit Bought at: Online from Germany	Yes	Yes	Made from corn-starch	Dimension: 16,00 cm
	Plates Manufacturer: Fuyit Bought at: Online from Germany	Yes	Yes	Made from sugarcane bagasse fibre	Dimension: 17,78 cm

2.2 BMP assays

The first batch experiment was carried out in duplicates in 120 ml infusion Pyrex bottles and the second in triplicates in 1L Pyrex glass bottles.

Two different set-ups were studied in the first experiment. In one set-up the materials were in contact with just the inoculum. For the second batch effluent and food waste were added.

The BMP was performed based on the $gVS_{substrate}$/ $gVS_{innoculum}$ratio of 0.5. The amount of material in each bottle was calculated based on the Total Solid (TS) and Volatile Solid (VS) of the materials.

Before filling the reactors materials, food waste, and inoculum were weighed. Inoculum was added into each bottle at the beginning of the experiment. Then the required amounts of materials and food waste were added to each bottle along with 5ml of $NaHCO_3$ (42 g/L) and later were filled with distilled water to reach the desired volume of 60 mL of working volume. To make sure that the initial pH in each bottle was between 7 and 8,the pH was measured using a(pH 3210, WTW 82362 Wellhelm) and if needed the pH was set using 2M NaOH or HCl. A rubber septum (Plug, BUTYL) and an aluminium cap (20MM, Aluminium crimp cap, Blank, Chromacol) were placed to seal each reactor. The infusion bottles were sealed by using a crimper (Crimper 20mm, XDOCK). To assure anaerobic conditions the headspace of each bottle was purged with N_2-gas for 3-5 minutes. The bottles were kept at the temperature of 35 °C in an oven (Memmert, home & laboratory manager Labilo) for up to 90 days.

For the second batch the experiments were performed in the Pyrex glass bottles with a volume of 1L. To build the reactors different pieces were needed.

Before starting the experiment, the reactors were prepared by following the instructions present in the lab.

Once prepared, the reactors were filled up with the respective components of the experimental set-up. based on the TS and VS of the material with the ratio of 2. Total volume in one bottle was 700mL consisting of biobased packaging material, inoculum, buffer solution and distilled water. It was assumed that the density of all materials is about 1 mL/g, this assumption allowed to calculate the headspace volume, in this case it was of 300 ml. First the bottles were filled with distilled water and buffer solution (42g/L $NaHCO_3$. Once filled up and closed a water displacement method was used to check if the bottles were airtight. The head space of each bottle was purged with N_2-gas for 5 minutes to ensure anaerobic conditions. At the end, the reactors were placed in the water bath and connected to the gas bag. The biogas produced was collect in aluminium gas bags (Supel^{TM} Intert Foil Gas Sampling bags, screw cap valve, 5L). The experiment was performed in triplicates, except for the biowaste and coffee cups, at mesophilic temperature of (35 °C). Even though the temperature set was 35 °C inside the water bath the temperature was between 30-32 °C. The BMP analyses was continued for up to 60 days.

The experimental set-up was done according to the characteristics of the materials used with the constant inoculum to substrate ration of 2 for the material.

Infusion bottles (bottle volume: 120ml; liquid volume: 60ml): BMP of the biomaterials was measure in the presence of the inoculum (Table 6).

Sample	*g/bottle:*	*gTS/bottle:*	*gVS/bottle:*	*inoc/substr VS-ratio*	*TS (%):*	*VS (%):*	*VS/TS:*
Inoculum	30,0	1,93	0,93	2,0	6,44	3,09	0,48
Egg boxes	0,59	0,56	0,46	2,0	95,83	79,08	0,83
Coffee cups	0,49	0,47	0,46	2,0	95,62	95,31	1,00
Paper bags	0,49	0,47	0,46	2,0	95,35	94,68	0,99
Plastic bag and sack	0,47	0,47	0,46	2,0	98,89	98,53	1,00
Plastic bag and sack (+)	0,48	0,48	0,46	2,0	98,43	95,74	0,97
Thin plastic film	0,47	0,47	0,46	2,0	99,81	98,66	0,99
Non-biodegradable plates	0,53	0,52	0,46	2,0	99,94	88,39	0,88
Food waste	1,53	0,52	0,46	2,0	34,09	30,28	0,89

Table 6. *Set-up: 120mL Infusion bottles set-up (biomaterials and inoculum). Inoculum/Substrate ratio: 0,5*

Sample	*Inoculum (g)*	*Sample (g)*	*NaHCO3 (g)*	*H2O (g)*	*Total (g)*
Inoculum	30	30,0	6	24	60,0
Egg boxes	30	0,59	6	23	60,0
Coffee cups	30	0,49	6	24	60,0
Paper bags	30	0,49	6	24	60,0
Plastic bag and sack	30	0,47	6	24	60,0
Plastic bag and sack(+)	30	0,48	6	24	60,0
Thin plastic film	30	0,47	6	24	60,0

Non-biodegradable plates	30	0,53	6	23	60,0
Food waste	30	1,53	6	22	60,0

Infusion bottles (bottle volume: 120ml; liquid volume: 60ml): BMP of the biomaterials was measure in the presence of food-waste and inoculum (Table 7).

Sample	*g/bottle:*	*gTS/bottle:*	*gVS/bottle:*	*inoc/substr VS-ratio*	*TS (%):*	*VS (%):*	*VS/TS:*
Inoculum	30,0	1,93	0,93	2,0	6,44	3,09	0,48
Egg boxes	0,59	0,56	0,46	2,0	95,83	79,08	0,83
Food waste	1,53	0,52	0,46	2,0	34,09	30,28	0,89
Egg boxes+ food waste	2,12	1,08	0,93	4,0			1,71
Coffee cups	0,49	0,47	0,46	2,0	95,62	95,31	1,00
Food waste	1,53	0,52	0,46	2,0	34,09	30,28	0,89
Egg boxes+ food waste	2,02	0,99	0,93	4,0			1,89
Paper bags	0,49	0,47	0,46	2,0	95,35	94,68	0,99
Food waste	1,53	0,52	0,46	2,0	34,09	30,28	0,89
Paper bags+ food waste	2,02	0,99	0,93	4,0			1,88
Plastic bag and sack	0,47	0,47	0,46	2,0	98,89	98,53	1,00
Food waste	1,53	0,52	0,46	2,0	34,09	30,28	0,89
Plastic bag and sack + food waste	2,00	0,99	0,93	4,0			1,88

Plastic bag and sack (+)	0,48	0,48	0,46	2,0	98,43	95,74	0,97
Food waste	1,53	0,52	0,46	2,0	34,09	30,28	0,89
Plastic bag and sack (+) + food waste	2,02	1,00	0,93	4,0			1,86
Thin plastic film	0,47	0,47	0,46	2,0	99,81	98,66	0,99
Food waste	1,53	0,52	0,46	2,0	34,09	30,28	0,89
Thin plastic film + food waste	2,00	0,99	0,93	4,0			1,88
Non-biodegradable plates	0,53	0,52	0,46	2,0	99,94	88,39	0,88
Food waste	1,53	0,52	0,46	2,0	34,09	30,28	0,89
Non-biodegradable plates + food waste	2,06	1,05	0,93	4,0			1,77
Food waste	1,53	0,52	0,46	2,0	34,09	30,28	0,89

Table 7. *120mL Infusion bottles set-up (biomaterials + food-waste and inoculum). Substrate/Inoculum ratio: 1*

SAMPLE	**INOCULUM (G)**	**SAMPLE (G)**	**BIOWSATE (G)**	**NAHCO$_3$ (G)**	**H2O (G)**	**TOTAL (G)**
INOCULUM	30	0,00	0,00	6,00	24,00	60,0
EGG BOXES	30	0,59	1,53	6,00	21,88	60,0
COFFEE CUPS	30	0,49	1,53	6,00	21,98	60,0
PAPER BAGS	30	0,49	1,53	6,00	21,98	60,0
PLASTIC BAG AND SACK	30	0,47	1,53	6,00	22,00	60,0
PLASTIC BAG AND SACK (+)	30	0,48	1,53	6,00	21,99	60,0
THIN PLASTIC FILM	30	0,47	1,53	6,00	22,00	60,0

NON-BIODEGRADABLE PLATES	30	0,53	1,53	6,00	21,94	60,0
FOOD WASTE	30		1,53	6,00	22,47	60,0

BMP in Pyrex glass bottles (bottle vol. (ml): 1000; Liquid vol. (ml): 700; Headspace (ml) :300): the BMP of the biomaterials was measure in the presence of food-waste and inoculum (Table 8).

SAMPLE	**G/BOTTLE:**	**GTS/BOTTLE:**	**GVS/BOTTLE:**	**SUBST/INOC VS-RATIO**	**TS (%):**	**VS (%):**	**VS/TS:**
INOCULUM	350,0	10,61	5,77	2,0	3,03	1,65	0,54
PAPER BAGS	3,04	2,90	2,88	2,0	95,35	94,68	0,99
FOOD WASTE	12,88	3,35	2,88	2,0	26,02	22,38	0,86
TOTAL	15,93	6,26	5,77	4,0	121,37	117,06	1,85
COFFEE CUPS	3,02	2,89	2,88	2,0	95,62	95,31	1,00
FOOD WASTE	12,88	3,35	2,88	2,0	26,02	22,38	0,86
TOTAL	15,91	6,24	5,77	4,0	121,64	117,69	1,86
PLASTIC BAG AND SACK	2,93	2,89	2,88	2,0	98,89	98,53	1,00
FOOD WASTE	12,88	3,35	2,88	2,0	26,02	22,38	0,86
TOTAL	15,81	6,25	5,77	4,0	124,91	120,91	1,86
PLASTIC BAG AND SACK (+)	3,01	2,96	2,88	2,0	98,43	95,74	0,97
FOOD WASTE	12,88	3,35	2,88	2,0	26,02	22,38	0,86
TOTAL	15,89	6,32	5,77	4,0	124,45	118,12	1,83
SUGARCANE PLATES	2,92	2,90	2,88	2,0	99,27	98,79	1,00
FOOD WASTE	12,88	3,35	2,88	2,0	26,02	22,38	0,86

TOTAL	15,80	6,25	5,77	4,0	125,29	121,17	1,86
STARCH-BASED CUTLERY	3,76	3,72	2,88	2,0	99,04	76,65	0,77
FOOD WASTE	12,88	3,35	2,88	2,0	26,02	22,38	0,86
TOTAL	16,64	7,08	5,77	4,0	125,06	99,03	1,63
NON-BIODEGRADABLE PLATES	3,26	3,26	2,88	2,0	99,94	88,39	0,88
FOOD WASTE	12,88	3,35	2,88	2,0	26,02	22,38	0,86

Table 8. *BMP Pyrex bottles set-up*

SAMPLE	**INOCULUM (G)**	**MATERIAL (G)**	**BIOWASTE (G)**	**NAHCO3 (G)***	**H2O (G)**	**TOTAL (G)**
INOCULUM	350	0	0,00	67,00	283	700
PAPER BAGS	350	3,04	12,88	67,00	267,07	700
COFFEE CUPS	350	3,02	12,88	67,00	267,09	700
PLASTIC BAG AND SACK	350	2,93	12,88	67,00	267,19	700
PLASTIC BAG AND SACK (+)	350	3,01	12,88	67,00	267,11	700
SUGARCANE PLATES	350	2,92	12,88	67,00	267,20	700
STARCH-BASED CUTLERY	350	3,76	12,88	67,00	266,36	700
NON-BIODEGRADABLE PLATES	350	3,26	12,88	67,00	266,86	700
FOOD WASTE	350	0,00	12,88	67,00	270,45	700

2.3 Analyses and calculation

Totals solids/dry matter was analysed according to Standard EN 14346 and volatile solids/loss on ignition (VS) according to Standard EN 15169. For the sampling of the materials aluminium foil (VWR Disposable Aluminum Weigh Dish 75 ml) were used.

Standard EN 14346 requires burning the materials in an oven (Memmert, Tamro LAB)) at 105 °C overnight.

For VS determination, Standard EN 15169 requires flaming over a burner before ash in a muffle furnace (Heraeus Electronic) at 550 °C for 2 hours.

Before sealing the bottles, the pH was measured (pH 3210, WTW 82362 Wellhelm) to be sure that the initial pH was in the 7-8 range if not the pH was set using 1M NaOH or HCl.

The percentage of methane produced was measured using Gas chromatography (GC) (Perkin Elmer Clarus 500 GC-FID gas chromatograph) with helium as gas carrier. The flow rate of helium was 14.00 mL/m, instead the gas flow for air and H_2 were respectively 450 ml and 45.0 ml. Temperature of injector and detector were 100 °C and 250°C, respectively. Biogas at every measurement point, room conditions (Temperature, time, pressure) were noted. A gas standard with 50% methane and 50% carbon dioxide was used as standard. Typically, 5 injections of gas standard were made.

The methane content present in the headspace of the reaction bottles was measured regularly throughout the experiment. In the first part of the experiment, it was possible to sample the gas only once a week due to the restriction related to the Coronavirus epidemy. For the second part, the methane was measure three times a week for the first weeks and later it was enough to measure once/twice a week.

Gas samples (1 ml) were taken from the headspace of the reactors through the septum with a syringe with pressure lock (Pressure-Lock^{R} Series A-2 Syringe, 500 ul). The pressure lock was closed after the needle of the syringe had penetrated the septum and was inside the reactor headspace, making it possible to sample a fixed volume of gas at the actual pressure in the reactor. The syringe was redrawn, and the sample was injected directly into the gas chromatograph where the concentration of methane was measured.

Due to the amount of biogas produced it was necessary to release gas during the sampling to avoid build-up of high pressure in the reactor leading to leakage of gas or to the explosion of the reactors. The pressure was released under water by inserting a needle (BD Microlance^{TM} 3) in the rubber septum. The amount released was calculated from measurement of the methane content in the headspace of the reactor before and after the released.

Biogas volume inside the gas bags was measured using a water displacement method. The method consists of using a water column and a scale to quantify the amount of biogas produced by the anaerobic digestion of the biobased packaging material.

The parameters used to calculate the biogas production were:

Volume of gas (L): it is the gas accumulated in the bag during all experiment and measured in different time points.

- Production rate (L/d): $\frac{\boldsymbol{Volofgas}\ (\boldsymbol{L})}{(\boldsymbol{T2}-\boldsymbol{T1})}$
    - Expressed as litres of biogas produced per day. Calculated as the volume of biogas accumulated in the gas bags divided by the time (day).
- **Cumulative (L):** is the sum of gas, in litres, produced during the experiment.
- Concentration (%):
    - $\boldsymbol{50x}\frac{\boldsymbol{Average}\ (\boldsymbol{ofgasconc.obtainedbyGCforthesample})}{\boldsymbol{Average}\ (\boldsymbol{ofgasconc.obtainedfortheSTDgas})}$
- **Volume (L):** is the volume of biogas produced and accumulated in the BMP bottles and is calculated as
    - $\left(\left(Vol.(L)+Headspace(L)x\frac{Concentration(\%)}{100}\right)-\left(Headspacex\frac{Concentration(\%)}{100}\right)\right)xpressure$
- Biogas production rate (L/d):
    - $Prod.rate\left(\frac{L}{D}\right)x\frac{Concentration\ (\%)}{100}xpressure$
- Where the factor (0 °C - 1 bar) is a specific parameter that take in consideration the conditions of the room where the experiments have taken place, it considers the temperature and the pressure. Calculated as
- $\left(\frac{273{,}15}{8273{,}15+T(°\mathrm{C})}\right)x\frac{p(bar)}{1}$

- **Methane Production Potential (L):** of a particular material was calculated as average of methane (L) produced for duplicates/triplicates samples excluding the values of methane obtained for the effluent.

3. RESULTS

3.1 TS & VS

Characteristics of the materials used for the experiments are shown in

Table 9. Among the selected materials the ones that present the highest values of TS and VS are those extracted from biomass, the bio-based materials obtained from polysaccharides such as starch, cellulose, etc. For instance, the plastic bag is coextruded biofilm made from a blend of starch and Polyvinyl Alcohol (PVA) designed as shopping bags that can be also used to dispose the food waste in the household. These bags present a TS of 98%. Other examples are the plant-based materials, the cutleries obtained from corn-starch and the plates made from sugarcane bagasse fibre present a TS of 99%. The PET based plate used as control presented a high value of TS (99%), with the difference that the plate cannot be used as feedstock in an AD process to obtain biogas.

The experiment was carried out in different moments, so it was necessary to prepare diverse food waste and to use different inoculum. The inoculums were obtained from a local wastewater facility, but since the compositions of the sludge changes the TS and VS were measured every time. The inoculum used during the first set of experiments represented a higher TS value than the inoculum used for the second set of experiments.

The food waste used as feedstock also represented different TS values since there were obtained collecting the food-waste of two different households in different moments. The composition of the food waste in term of C, H, N in this case is influenced not only by the diet of the subjects involved by also to the fact that it is domestic food waste.

Table 9. *Characteristics of the biomaterials*

Material	Average TS%	TS% std dev	Average VS%	Average VS/TS	VS/TS std dev
Inoculum n1	6.44	0.095	3.09	0.48	0,005
Inoculum n2	3.03	0.018	1.64	0.54	0,0003
Food waste n1	34.08	0.167	30.28	0.88	0,014
Food waste n2	26,01	0,0318	22.39	0.86	0,006
Eggs boxes	95.83	0.030	79.08	0.82	8,853E-05
Paper bag	95.35	0.092	94.68	0.99	0,0001
Coffee cups	95.62	0.105	95.31	0.99	5,493E-05
Plastic bag and sack	98.89	0.134	98.53	0.99	9,227E-05
Plastic bag and sack (+)	98.43	0.029	95.74	0.97	0,0001
Cutleries	99.03	0.050	76.65	0.77	0,020
Sugar-based Plate	99.26	0.563	98.79	0.99	0,0002
Non-biodegradable plates	99.94	0.006	88.39	0.88	0,001

3.2 Methane production potential

The cumulative methane production of the tested biomaterials is reported in (Figure 5). The methane production potential of the biomaterial (Figure 5a) and the methane production potential of biomaterial in the presence of food waste (Figure 5b) were done in 120ml infusion bottles.

Figure 5a represents the results obtained for methane production potential from the experimental set-up where the biomaterials were solely investigated. The litres of biogas produced by the biowaste were around 369 L CH4/kg-VS. Among the materials the ones with the highest volume of methane produced are those obtained from biomass, those derived from plant-based polymer such as cellulose. In fact, paper bags and coffee cups produced respectively 312 and 318L-CH4/kg-VS. The amount produced by the egg boxes was 204L-CH4/kg-VS. For the other polysaccharide-based materials, the food bags and sacks, the values of methane obtained were not that high when compared with the cellulose-based materials. The first food bags produced only 105 L-CH4/kg-VS while the second 106 L-CH4/kg-VS. The anaerobic digestion of the non-biodegradable plates, used as control, resulted in no production of biogas (-0,1 L-CH4/kg-VS). The AD of the thin plastic film resulted 36 L-CH4/kg-VS, produced at the very end of the experiment.

Respectively Figure 5b shows the cumulative methane production of the biomaterial studied in the presence of food waste representing conditions where the biomaterial are discarded with the food residues. In this case the egg boxes were the material with the highest amount of methane produced when compared with the other substrates tested, showing value of 282 L-CH4/kg-VS. Even for the set of experiments which were done in the presence of food waste the PET plate and the thin plastic-film were the materials showing the lowest value for bio-methane production (PET: 68L-CH4/kg-VS, Plastic-film: 59 L-CH4/kg-VS). The paper bags and coffee cups showed values around 184 and 153 L-CH4/kg-VS. Regarding the two different plastic bags the values are respectively 114 and 112 L-CH4/kg-VS.

When we compare the results obtained, it is evident that the values obtained here are diverse, especially for the cellulose-based materials. The values of CH_4 produced are higher of those obtained when the biomaterials were solely investigated. This is true for most of the materials except for the egg-boxes, in this case the addition of food waste resulted in higher values of methane. Without the food waste the amount produced was 204L-CH4/kg-VS against the 282 L-CH4/kg-VS produced in the presence of food-waste.

For the coffee-cups there is a difference of almost 200 L in CH_4production,318 L-CH4/kg-VS against the 153 produced with food-waste. Such difference in production can be spotted for the paper-bag too (312*vs*184L-CH4/kg-VS).

Instead, for the polysaccharide-based materials, the addition of the food-waste did not really change the methane production. This can be notice when comparing the results obtained. For instance, the amount of methane produced by the first type of food sack was 105 L-CH4/kg-VS (when the biomaterial was tested lonely) against the 114 L-CH4/kg-VS when the biomaterial was tested in the presence of food-waste. The second version, instead, generated 106 L-CH4/kg-VS when tested lonely and 112 L-CH4/kg-VS in the presence of the food-waste. So, for the plastic bags the addition of the food-waste resulted in a difference in production of 6L.

The addition of the food waste also affected the BMP of the two plastics used as control. For instance, without food waste the amount of biogas obtained from the AD of the PET-plate were negative, meanwhile with food waste the plate produced 68,2 L-CH4/kg-VS. This can be seen also for the thin plastic-film (with food-waste: 59 L-CH4/kg-VS; no food-waste: 36 L-CH4/kg-VS). This is interesting considering that the L-CH4/kg-VS produced by the biomaterials and controls were calculated reducing the biogas production of the food waste and inoculum.

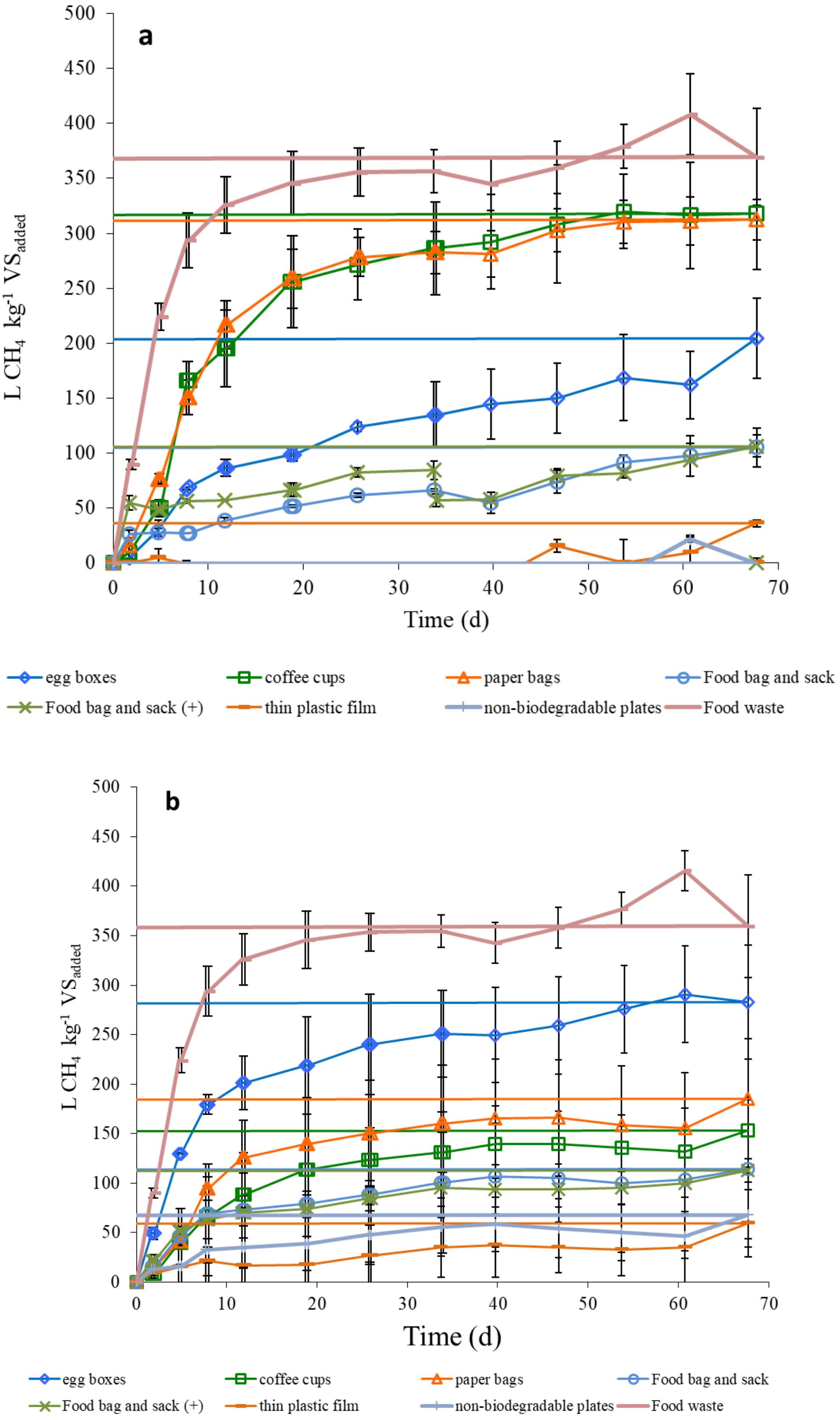


Figure 5. *Cumulative methane production of substrate (L CH4 Kg-1 VS added): (a) BMP of biomaterials in the only presence of the inoculum, methane production of inoculum reduced; (b) BMP of the bio-material in the presence of food-waste and inoculum, methane production by inoculum and food waste was reduced.*

Figure 6 instead presents the results obtained from the BMP test carried out in the 1L Pyrex bottles (Set-up 2).

The results obtained from experiments performed in one-liter scale showed that the total amount of methane produced by the food waste alone was 474L-CH4/kg-VS. The methane production plot for food waste represented in showed a decrease in biogas production at around 30th day of the experiment which could be related to the problems in temperature control of the water-bath.

Looking at the graph it is clear that the cellulose-based biomaterials where the substrates that produced the highest amount of methane. In particular, the paper-bags produced the highest amount showing a value of 464 L-CH4/kg-VS. Looking at the paper-bags curve it is possible to notice an increase in production after the 35th day. Sugarcane plates produced values around 442 L-CH4/kg-VS. The sugarcane-plate curve is different when compared with those of the other substrates. The plate´s plot present a lag phase that last for 7 days. The coffee cups produced 372,4 L-CH4/kg-VS. The two food bags produced respectively 262,5 and 303,4L-CH4/kg-VS. When confronting the plots of the two plastic-bags it is evident that the second type of compostable bag was the bag that produced the highest amount of methane.

Regarding the cutleries made from cornstarch their results have not been included in the results represented in Figure 6 because if we look at the data collected it is evident that the material has not been degraded. This is interesting since the manufacturer claims their biodegradability and compostability. The outcome is confirmed by the tokens recovered once the bottles were emptied at the end of the experiment, where there is no evidence of degradation. This was also confirmed by the surface analysis made with the microscopy.

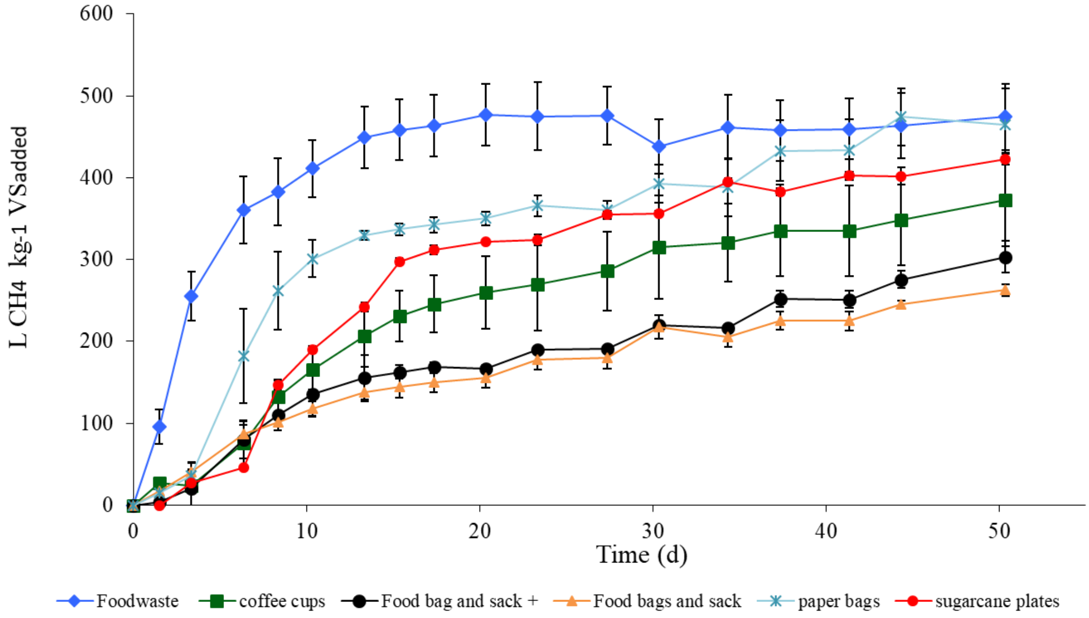


Figure 6. *Cumulative methane production of substrates (L CH4 Kg-1 VS added) from the second set-up: food waste and inoculum reduced*

The results obtained clearly demonstrate that the materials tested have the potential to be used to produced biogas. Even though the experiment was performed in two different periods and by using two different methods the data obtained shows more or less similar trend. It is important to consider that both methods are based on the same process, the anaerobic degradation operated by the coordinating action of different microorganisms. Cellulose-based materials in both experiments were the substrates with the highest degree of degradation and with the highest values of biogas produced. Among the cellulose-based, paper-bag and coffee cups are the materials that present the highest potential for biogas production. This is true for at least two of the three different set-ups used. In Figure 5b in fact it could be observed that the egg-boxes are the substrate with the highest volume of biogas produced followed by the paper-bags. It would be interesting to test the BMP of the egg-boxes using a 1L Pyrex bottle to see if the result will be different.

In Figure 5b and Figure 6 the plots of the biomaterials have a similar trend as the set-ups used for test the BMP are the same.

In all the tree different set-ups the food waste´s curves have the same growth, at least for the first 30 days.

3.3 Physical status and surface analysis of biodegraded biomaterials

At the end of the experiments, the BMP bottles were emptied to recover and analyse the materials. Once recovered they were visually inspected. The PET plate and the thin plastic-film used as control showed no visual sign of change.

Regarding the cellulose-based biomaterials, these showed different responses. The BMP tests on the paper-bag resulted in the completely biodegradation of the biomaterials, no tokens were recovered at the end of the experiments. The same outcome was obtained for the plates made from sugarcane bagasse fibre. The complete digestion of these two biomaterials was expected considering their thickness, in particular the one of the paper-bag (shape as a thin layer of cellulose).

Regarding the egg-boxes tokens recovered, it can be notice that there has been a changing in colour. After 70 days of experiment the tokens were discoloured, they went from a bright yellow to a pale colour. The loss of colour could be caused by the absorption of the digestate by the cellulose-fibres. Visually there were no sign of digestion, even though in the BMP test performed in the 120mL infusion bottles, the egg-boxes were one of the substrates that produced the highest amount of food waste.

The 1L BMP test of the coffee cups highlighted the presence of a coating. The presence of this transparent thin film was noticed only at the end of the experiment. This due to the digestion of the cellulose part of the cups and the setting of the coating film on the bottom of the bottles. The presence of the food-waste inside the bottles made it difficult to detect its presence. Further analysis should be made on the coating, to discover which polymer was used. It is also important to consider that no information about the coating are present on the label of the products, it is just specified that the cups are 100% compostable and biodegradable.

As demonstrated from the data collected, no sign of digestion was notable on the cutleries' tokens recovered.

The plastic-bag-tokens visually did not present a sign of degradation. It looked like they were disrupted than degraded. Visually a loss of colour can be detected. The original colour of the plastic-bags was respectively light green and brown. Both at the end presented a pale colour. The first one presented a pale yellowish color, while the second bag turned into a pale brown.

3.3.1 Microscopic examination

Six of the eight tested biomaterials were selected for preliminary microscopic examinations based on their appearance once recovered from the BMP bottles. Samples were taken from the washed and air-dried material removed from the digestate, with no special measures taken to preserve microbial films. The tokens were examined using Scanning Electron Microscopy.

The selected materials were the plastic-bags, the coating recovered from the anaerobic digestion of the coffee cups, the egg-boxes, the PET plate and the thin plastic film.

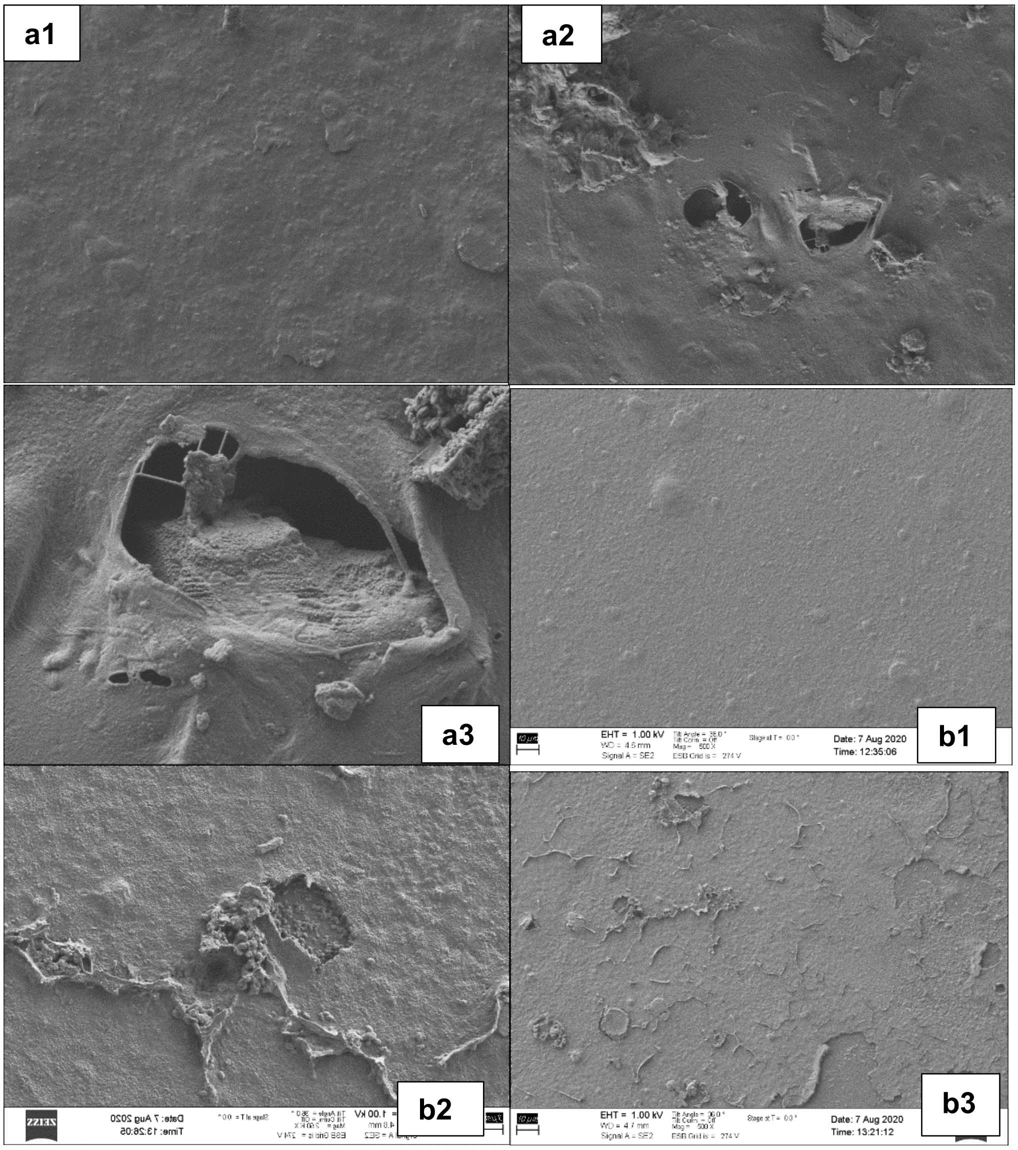
The surface analysis operated on the PET and on the Thin plastic-film highlighted the presence of cavity, that were no detected while doing the physical evaluation (Figure 7). For the PET, it is suspected that these holes are the result of the anaerobic degradation operated on a possible biodegradable coating present on the surface of the material (Figure 7, a1, a2, a3). The microscopic analysis of the thin plastic film showed physical degradation (Figure 7, b1 and b3). Further analysis should be made. It is suggested a characterization of the inoculum to have a better understanding on its enzymes-activity, especially on the biopolymers and on the petrochemical-based polymers.

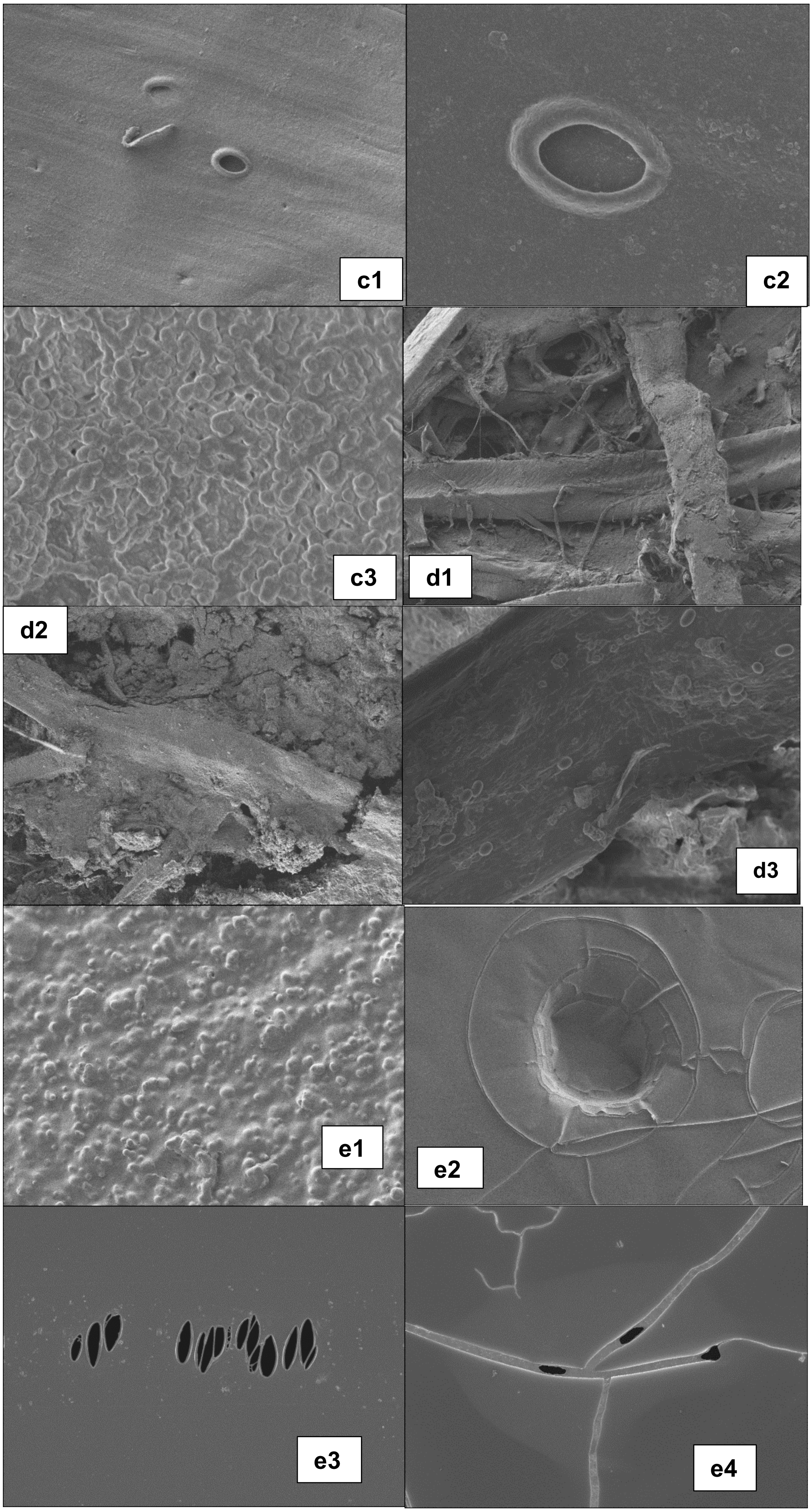
Regarding the coffee-cups, as mentioned previously the BMP assay resulted in the complete degradation of the biopolymer. What was left behind was a thin transparent film used as coating. For this reason, no confrontation was possible between the original materials and the biomaterial after the test. It was decided anyway to make the microscopic surface analysis of the coating (Figure 7).The evaluation brings out the presence of small cavity (Figure 7,c1, c2 and c3). There is no available information on the nature of the polymer used as a coating. Based on the holes detected it is not excluded a possible biodegradability. But considering the small number of holes and their size it is thought that a longer period is required for the complete degradation of the material.

Instead, the surface analysis of the egg-boxes detected a degradation. A degradation that was not visually detected. This is evident when confronting the images of the tokens before and after the BMP assay (Figure 7, d1 and d2). In the tokens recovered from the bottles it was possible to notice that cellulose fibres have been broken because of the inoculum growth (d2). The surface clearly showed signs of progressive attack in specific point (on the structure of the fibres, but also around), but it also showed the presence of bacterial colonies (d3).

Sign of anaerobic degradation on the food waste-bags can be detected by the presence of small pits and cracks on the surface (Figure 7, e2, e3, e4, and f3). The pits made by the inoculum are not deep, as indicate that the bags are compostable and biodegradable materials that required longer time to be degraded, especially at mesophilic conditions. Pits, cracks and holes, can be seen on the surface of the first kind of plastic-bags tested (e2, e3, and e4). By looking at pictures, e2 and e4, it is possible to understand how the inoculum degraded the material. First, it attacked the surface and its action resulted in cracks (e2). The digestion then continued inside the volume of the cracks (e4).

On the surface of the second type of food waste-bag tested the action of the inoculum led to the formation of cracks (Figure 7, f3). As for the pits, the cracks are superficial. This indicated that a longer time is required to obtain more visible sign of degradation.

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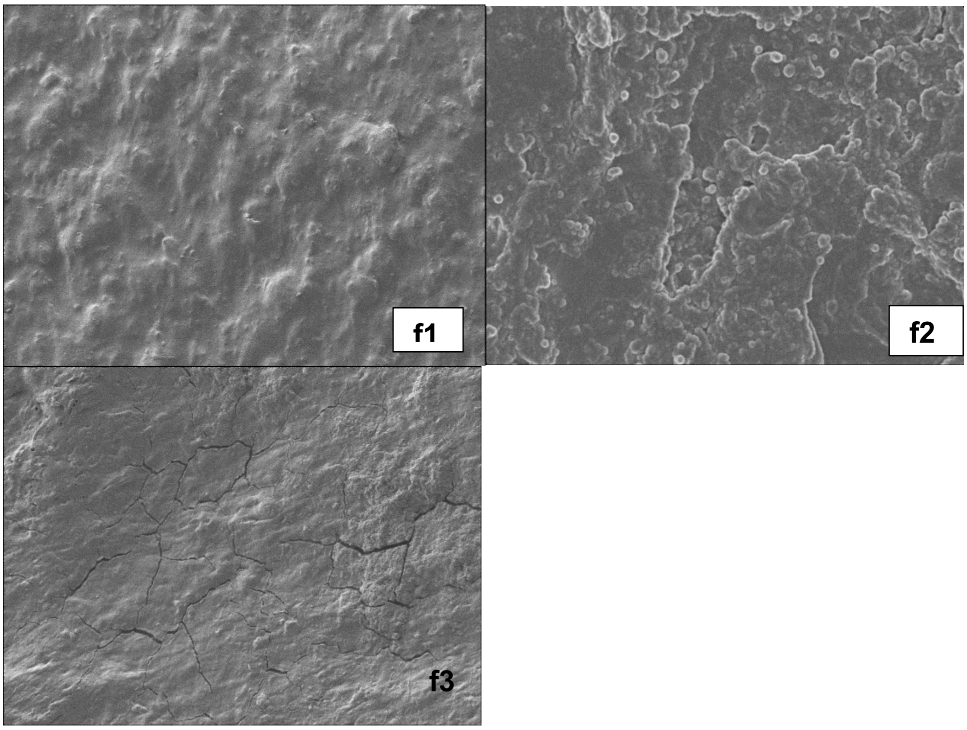

Figure 7. *Microscopy images :PET-plate (a1 : surface of the PET plate; a2-a3: surface showing a pit); Thin plastic-film (b1: surface of the thin film; a2-a3: partially digested surface after BMP test); Coating recovered from the AD of the coffee -cups (c1-c2: hole present on the surface of the coating; c3: partially digested surface of the coating); Egg-boxes (d1: image of the cellulose fibers before the AD: d2: structure of the cellulose-fibers partially degraded; d3: inoculum cells on the surface of the fibers); Food waste bag (e1: surface of the bag; e2: cracks resulted from the inoculum digestion; e2: holes on the surface of the food waste bag; e3: cracks resulted from the AD); Food waste bag+ (f1: surface of the bags; f2: inoculum cells on the surface ; f3: cracks resulted from the inoculum digestion). Source: Images and courtesy of Turkka Salminen, Tampere University Hervanta Campus.*

4. DISCUSSION

4.1 Food-waste: Characteristics, Potentials and challenges

Compared with traditional disposal method (landfill, incineration, and composting), anaerobic digestion is a promising technology for food waste management (Xu et al., 2018). Is the composition of the food waste that make it suitable for this process. The total solid content of food waste has a wide range, from dilute liquid (less than 2%) to solid (more than 90%). The organic content (VS/TS) ratio is generally around 90% indicating a high potential for biological treatment (Li et al., 2013; Zhang et al., 2014). The highest methane potential of each type of food waste is in the range of 0.3-1.1 m^3 CH4/kgVS$_{added}$, generally higher than other AD substrates such as lignocellulosic biomass, animal manure, and sewage sludge (Mao et al., 2015). Anaerobic digestion is a promising technology to convert food waste to energy. The use of the food waste as a feedstock in the anaerobic digestion is not widely applied. This could be due to technical, economical, and social challenges associated with anaerobic digestion process (Xu et al., 2018). In term of organic recycling the presence of food-waste represents an advantage because it makes the bioplastics compatible with the alternative end of life scenarios. If a bioplastic is used to collect food waste, the mixed waste result to be suitable for the composting process. The addition of the bioplastics to the mixture can increase the C/N ratio of the food waste to an optimal value, enhancing the digestion process (European Bioplastics, 2020). The availability of readily degradable bioplastics can enable the combined conversion of multiple waste feedstock in a single process, thus eliminating a significant fraction of materials being disposed of in landfills. Also diverting food waste has been identified to alleviate landfill burden, reduce methane emissions and create value-added alternatives (Hedge et al., 2018).

For the BMP assays in this current study, food waste was used as co-substrate. The food waste was digested with the biomaterials because when discarded, food packages are commonly co-minge with food waste, which makes the mechanical recycling of these materials very difficult and this emphasis the organic recycling as an end-of-life alternative. The food wastes used belong to the consumption food waste type. This because it contained mainly kitchen waste, mixed with the content of the coffee filters. It presented nonedible portion of food (e.g. banana peels, egg shales) and uneaten food such as plate waste. It is reported that household and restaurant food waste present a TS% between 4,0-41,5, VS/TS(%) 88,7-95,1 and a methane yield($m^3/kgVS_{added}$) of 0,46-0,53 (Xu et al., 2018). The results obtained from the TS and VS analysis made on the food waste were similar to those reported from Xu et al., The analysis also showed that there was a difference in composition between the two. The first once in fact present higher TS than the second. This difference in composition could have vary the methane production. Further analyses should have been made. However, the results obtained from the BMP tests confirmed that the anaerobic digestion is a promising technology for the management of the food waste, as reported from Xu et al., 2018. The presence of the food waste in the digesters did not affect negatively the growth of the inoculum and the AD of the bioplastics .Above that, for the egg-boxes, the presence of the food waste resulted in the production of higher values of methane. The higher yield could be explained by synergistic effect obtained co-digesting the food waste with the sewage sludge (Xu et al., 2020). Similar result was obtained in the co-digestion of PLA with food waste (Hedge et al., 2018). The authors reported that the presence of the food waste increased the methane potential of 10%, indicating a synergistic effect, but resulted in a low degradation rate (29-49%). As well for PCL (3-22%) even after a long period (277 days) (Yagi et al., 2014).

In the literature there are example of studies carried out to test the AD of bioplastics, which reported the used of food waste as feedstock (e.g. Zhang et al., 2018; Bandini et al., 2020). In these studies, the food waste was co-digested with sewage sludge, as was made for this experiment. With the difference, that the BMP of the biopolymers was always studied in the presence of food waste. There are no literature values on anaerobic degradation of biomaterials to compare with this current study.

4.2 Anaerobic biodegradation of Bioplastics

The importance of evaluating the biodegradability of bioplastics under anaerobic conditions is mainly related to the presence of food packaging material not often sorted (although biodegradable) from food-waste, with the latter used as substrates in anaerobic digestion plant (Hedge at al., 2018) or to the use of compostable bags used to collect food waste in separate collection systems (Calabrò and Grosso, 2018) (Folino et al, 2020). Deterioration of organic materials and thus bioplastics, under anaerobic conditions, especially in an AD plant, offer some notable advantages. For instance, the limitation of odor emissions, the produced methane can be used as energy source (Yagi et al., 2014), as well as the nutrient-rich digestate residue, which can be used as fertilizer (Folino et al., 2020).

The results obtained from the BMP tests clearly demonstrate that the selected materials have the potential to produce biogas. Therefore, this make them suitable for the organic recycling. All the digester performed well during the experiment and it was clear that none of the materials caused any inhibition of the process at the loading rates used. The results from the BMP tests in the current study confirmed whether carbon conversion had occurred, and this indicate which of the test samples were anaerobically biodegradable. It is important to consider that the batch tests used are suitable for assessing whether a material is readily biodegradable, but do not necessarily reveal any long-term changes in the system biology that may affect its capacity and capability for ultimate degradation (Zhang et al., 2018).

As expected, the plant-based materials were those who produced the highest methane content. Paper-bag, coffee cups and the sugarcane plates were among the plant-based biomaterial those who stood out for their potential. The degradation of the cellulose-based materials has been studied in different environment (e.g. soil, simulated or field composting environments, anaerobic and aerobic conditions) (Folino et al., 2020). The results obtained from the studies carried out in these environments demonstrate that cellulose presents a very high rate of biodegradability. Generally, it has a range of biodegradability between 80 and 100 % (Folino et al., 2020). Similar results were obtained from the study. The results suggest that the polymeric structure of the plant-based materials was destroyed under anaerobic conditions, to an extent where the physical form was no longer recognizable. Different outcome was obtained from the egg-boxes. The AD of the egg-boxes showed a very low rate of degradation. It can be assumed that the process was influenced by the thickness of the packages. Which was higher when compared with those of the other plant-based materials. Rujnic´ and Pilipovic reporter that one of the factors affecting the rate of biodegradation is the thickness of the biodegradable material: the thicker the product, the longer its biodegradability. Another explanation could be that the microorganisms participating in anaerobic biodegradation did not have the ability to biodegrade higher molecular weight of cellulose. The microorganisms in the sludge may be able to degrade only smaller molecular weight of cellulose obtained in the hydrolysis phase. So, a longer period is required to obtain the same rate of degradation.

In the literature, some of the biopolymers that have been reported to biodegrade under anaerobic conditions are PHA, PHB, starch-blends, cellulose, PLA, PCL, PBS, and PVA (Folino et al., 2020). Regarding the starch-based food sack and bags tested, although the BMP tests resulted in the production of methane, both showed low physical breakdown in the simulation experiment. Similar results were obtained in the study made by Zhang et al. In the study, the authors looked at the anaerobic biodegradation of nine different bioplastics. Of the nine bioplastics tested, two were starch-based. The bioplastics were also tested at mesophilic conditions.
According to its chemical composition and design, each bio-polymer has a different rate of biodegradation, which is effected by the conditions of the plant (Rujnic´ and Pilipovic, 2017). Rujnic´ and Pilipovic reported that anaerobic microorganisms are able to degrade starch-based bioplastics, in mesophilic (≤35 ◦C) and thermophilic conditions (50–60 ◦C). Meanwhile a blend made of starch and PLA degrade at thermophilic conditions, a starch/PCL blend is digested at mesophilic conditions (Rujnic´ and Pilipovic, 2017).

The biodegradation of starch-based bioplastics has been studied in different environment and process conditions. In soil, was found a rate of degradation of about 95% (Jangong et al., 2019). The authors reported that the degradation increased by increasing the starch dosage. The process was also accelerated by the presence of water that led to a faster break down and allowed further attack of microorganisms. In general, starch-based plastic films, buried in field soil, were found to lose weight and degrade faster (one week) compared to other bio-based polymers. In composting environment, different rate of degradation was obtained. For instance, Javierre et al., reported a rate of ~85% after 90 days, while Mohee et al., a rate of 26,9% after 72 days. In anaerobic conditions, Folino et al concluded that starch-based biopolymers showed a lower performance when compared to other materials. Calabrò et al., underlined how starch-based bags are only partially degraded under normal hydraulic retention time.

The low rate of degradation could have been caused by the process conditions. The process conditions used for the tests were similar to those used for the “home composting”. In home composting, due to a low temperature (≤35 °C) and less frequent mixing, biomass degrades slowly. Moreover, under such conditions certain compostable materials certified for industrial composting (EN 13432) may not biodegrade sufficiently (Saalah et al., 2020). As what happened for the food bags tested. It is reported that mesophilic conditions do not allow a satisfying disintegration of the material, independently of the duration of the test (Zhang et al., 2018). Higher mass loss and disintegration have been found in thermophilic conditions (Calabrò et al., 2020). To improve the rate of degradation of the bags, thermophilic conditions could be tested.

4.3 Organic Recycling

Bioplastics are suitable for reuse, mechanical recycling, organic recycling, and energy recovery. Using compostable bio-based waste's bags, food packaging, and cutlery strengthens organic recycling as a waste management option and helps to increase waste management efficiency (Wojnowska et al., 2020).In order to be suitable for organic recycling, bio-based wastes need to meet the criteria of the European norm EN 13432 on industrial compostability (Wojnowska et al., 2020).The 13432 Standard states that for anaerobic degradation at least 50% of the substance needs to be converted into biogas over a two-month period to certify a packaging or a packaging material as compostable. A typical plant treating bioplastics operates with a hydraulic retention time of 15-30 days under thermophilic or mesophilic conditions (Bátori et al., 2018). Therefore, a bioplastic suitable for organic recycling should be able to degrade within these conditions (Bátori et al., 2018). Folino et al., reported that due to the process condition operated in the biogas plant the degree of degradation required by the EN 13432 is not always achieved. Of the seven selected biomaterials, the two carrier bags were the only one certified according to the EN 13432 standard. Even though the BMP tests were performed respectively for 50 and 70 days at mesophilic conditions, in any of set-up the carried bags reached the degree of degradation required. Above that, the tokens recovered from the digester at the end of the process were fully recognizable. Similar results were obtained in a study carried out to test the AD of compostable bags made of MaterBi® (Calabro et al., 2020). The compostable bags were degraded in mesophilic (35 ◦C) and thermophilic conditions (55◦C) for 30 days. The rates of biodegradation reached at the end of the experiment were 23-30% at 35 ◦C and 28-41% at 55◦C. Higher rates (78%) were obtained when the carried bags were pretreated with NaOH. The authors concluded that mesophilic temperature led to a significant increase of the methane yield but did not lead to a concurrent increase of the dry mass loss. Meanwhile, Napper and Thompson reported the complete degradation of a compostable bag (vegware) in aquatic environment (8.8−18.8 °C, sea temperature), within a 3-months period.

Considering that there is no information about the biopolymers that have been used for their production, a characterization of the physico-chemical structure of the plastic-bag should be made. Thus, to have a clear idea of which process conditions should be used to obtained a higher rate of degradation. In the literature is mention that these bags are usually made of resins composed of starch combined with biodegradable polymer such as PLA (Shan et al., 2020). The presence of other biopolymers could affect the rate of starch degradation. The biodegradability of a bioplastics is affected by the physico-chemical structure of the material but also by the conditions and properties of the test system (Folino et al. 2020).

A strategy that could be used to improve the rate of biodegradation under anaerobic environments is to use a thermal alkaline treatment (Benn et al., 2018), such as the addition of calcium carbonate at low concentrations (Hedge et al., 2018). Hedge et al., outlined that the alkaline pretreatment improved the degradation (>50%). Folino et al., reported that the biodegradability is not only affected by the physico-chemical structure of the material but also by the conditions and properties of the test system. These include volume and shape of the vessels, open or close bottles, mixing or shaking modes, oxygen supply, and test duration.

4.4 Management of Biodegradable plastics waste

Bioplastics is a large family of polymers that includes many different materials. Each of them should be treated by a different waste management option according to its characteristics (European Bioplastics, 2020). The impact of bioplastic on waste accumulation is not still completely evaluated (Vu et al., 2020). The end-of-life options for the management of bioplastics include biological waste treatment (e.g., composting, anaerobic digestion), recycling, incineration, and landfilling.

The main problem in bioplastic recycling is removing contamination in the recycling streams (Zhao et al., 2020). The techniques used currently to identify, and sort bioplastics include manual sorting based on labels, separation based on density, near-infrared detection. These techniques are mainly used for the recycling process of PLA (Niaounakis, 2019). However, there are always contaminates in the recycling system when plastics are recovered, which reduce recycling properties. Therefore, developing separate recycling streams for bioplastics is the only solution to improve the waste management efficiency (Niaounakis, 2019). Research on the recycling of bio-based materials, especially bio-blends and bio-composites, is still at a preliminary stage and lacks a deep understanding of the different factors affecting the performance, economy, and sustainability of recycled bioplastics (Wojnowska et al., 2020). According to Kawashima et al., chemical recycling and energy recovery via thermal treatment should be preferred due to the difficulties of post collection sorting.

Composting has been identified as the most relevant waste treatment technology available for biodegradable plastics. These because the process uses untapped food-waste potentials, strengthens the secondary raw material market, increase resource efficiency and reduce CO_2 emissions (European Bioplastics). From the organic recycling of bioplastics different products could be obtained, such as biogas (used as energy source), compost (that can be used as a soil amendment). In a circular economy context the compost is used as fertilizer, because it improves the plants growth, from which, thanks to the biorefinery, is possible to obtained bioplastics granulates, which are used to manufactured new products (European Bioplastic).Different problematics are faced at the composting facilities due to the fact that composting of bioplastics can be problematic. For instance, the existing plants processing bioplastics may be not effective in their management since they were not designed to process these materials. As a consequence, improvements in existing plants to process mixtures containing bioplastics and other organic materials should be identified and implemented. Pre-treatment methods are required to accelerate bioplastics biodegradation when anaerobically digested with food waste. The short composting period (<30 days) operated in the plant does not allow the fully biodegradation of the bioplastic.

4.5 Properties of Bioplastics

Many bioplastics (e.g. biodegradable polyester, starch and cellulose-based bioplastics, and drop-in) have been proven safe for food contact use (Zhao et al., 2020). Various requirement needs to be met when considering a material for food packaging application. Examples of common properties required are gas and water vapour permeability, mechanical changes, sealing and thermoforming capabilities, machinability, printability, resistance to light, water, and cost. These properties vary according to the different packaging applications and uses. In general, bio-PET and bio-PE (biobased but non-biodegradable) are mainly used for rigid packaging while biodegradable plastics (e.g. starch thermoplastic, PLA, etc.) are used for flexible packaging. Currently, among the two, the non-biodegradables have greater participation in food packaging because their properties and manufacturing processes are identical to conventional plastics. Above that, they can be recycled using the same streams used for conventional plastics. What limit the use of biodegradable bioplastics in food packaging applications are their properties. For instance, most of the biodegradable bioplastics have lower ductility, lower toughness, and lower flexibility than conventional plastics. Strategies used to improve these properties are improving performance and decreasing cost. A technique used to improve the mechanical strength of bioplastics is adding fibres. Not the synthetic one because they reduced the biodegradability. Lignocelluloses' fibres can be added to reinforce starch-based plastics. The adding of nanomaterials, such as nanoclay or chitosan, is another example of strategy used to increase the thermal stability of bioplastics developed from starch. Plasticizers (e.g. glycerol, xanthan gum) are usually added to the starch-based bioplastics to improve their strength, flexibility, and the ability to process it.

5. CONCLUSION

To assess the biodegradability and compostability, under anaerobic conditions, of some commercially available food packages, BMP methods were used. These were successful in quantifying the amount of methane produced by the degradation of the biopolymer. None of the biomaterials inhibited or destabilized the anaerobic digestion process. The co-digestion of the biomaterials with the kitchen waste clearly demonstrate that food waste is a suitable feedstock for the anaerobic digestion. Of the seven biomaterials tested at mesophilic condition, the plant-based polymers showed extensive biodegradation in the static BMP batch assay. Paper bag, coffee-cups and the sugar-based plates were those which not only produced the higher amount of methane but also were completely degraded under anaerobic conditions. The egg-boxes showed high methane conversion, especially when co-digested with food waste, but low physical change. Further analysis should be made on the physico-chemical characteristic of the certified compostable carrier bag to identify the better process conditions for their degradation.

Observation of the biomaterials using confocal microscopy showed the mechanism of attack of the inoculum on the biomaterials. These could be related to the properties and degradation behavior of each tested material.

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