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SUSTAINABLE ALTERNATIVES FOR PLASTICS IN FOOD PACKAGING

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To my family

especially to Jacqueline Rodríguez

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Abstract

The flexibility, lightness, transparency and low cost of plastics were the characteristics that have most supported the expansion of polymeric food packaging and gradually replacing traditional materials such as glass and aluminum. Despite the surprising technical and aesthetic potential, the extensive use, production, end-of-life treatments and even worse, dispersion of plastics into the environment have brought to light great environmental problems associated with them. Therefore academia, governments, industries and the growing group of conscious consumers are eager to find and prefer more sustainable alternatives that can begin to take the first steps towards an upcoming replacement of plastics in the food packaging sector and of single-use products. The present research considers an overview of the main concepts of food packaging and the analysis of different groups of sustainable solutions: bio-based, biodegradable, recycled or bioplastic coatings for paper solutions. Each group examines its breakthrough and the current challenges and disadvantages that hinder its evolution on a large scale. The research also takes in account several innovative and emerging cases on the market, as an example and stimulus for the food packaging sector. Therefore, the thesis offers a discussion from the economic point of view of this sector with its main success factors, strengths, weaknesses, opportunities, and obstacles.

Keywords: sustainable food packaging, eco-design, bioplastics, recyclable plastics, biobased plastic coatings, circular economy

[eng]

La flessibilità, la leggerezza, la trasparenza e il basso costo delle materie plastiche sono state le caratteristiche che più hanno sostenuto l'espansione del packaging alimentare polimerico e andando gradualmente a sostituire i materiali tradizionali come il vetro e l'alluminio. Nonostante le sorprendenti potenzialità tecniche ed estetiche, l'esteso uso, produzione, conferimento, trattamenti a fine vita e ancor peggio, dispersione nell'ambiente delle plastiche hanno portato alla luce grandi problemi ambientali ad esse associati. Ecco perché il mondo accademico, i governi, le industrie e il crescente gruppo di consumatori consapevoli sono desiderosi di trovare e preferire alternative più sostenibili che possano incominciare a fare i primi passi in vista di una prossima sostituzione delle plastiche nel settore dell'imballaggio alimentare e dei prodotti mono-uso.
La ricerca considera una panoramica dei principali concetti di packaging alimentare e lo sviluppo di ogni gruppo di soluzioni sostenibili: soluzioni bio-based, biodegradabili, riciclate o coating bioplastici per carta. Ogni gruppo esamina la sua svolta e le attuali sfide e svantaggi che ne ostacolano l'evoluzione su larga scala. Nella ricerca sono inoltre analizzati diversi casi innovativi ed emergenti sul mercato, come esempio e stimolo per il settore dell'imballaggio alimentare. Pertanto, la tesi si conclude con un'analisi e una discussione anche dal punto di vista economico di questo settore con i suoi principali fattori di successo, punti di forza, debolezza, opportunità e ostacoli.

Parole chiave: imballaggi alimentari sostenibili, eco-design, bioplastiche, plastiche riciclabili, rivestimenti in plastica biobased, economia circolare

[ita]

La flexibilidad, ligereza, transparencia y bajo coste de los plásticos han sido las características que más han soportado la expansión de los empaques de alimentos en este material y el gradual reemplazo de otros tradicionales como el vidrio y aluminio por este. A pesar de su sorprendente potencial estético y técnico; su uso extensivo, producción, tratamientos de disposición final vinculados y aún peor, su dispersión en el medio ambiente han puesto a la luz los problemas asociados a este. Es por ello, que la academia, los gobiernos, el sector privado y cada vez más grande el grupo de consumidores conscientes, se encuentran particularmente interesados por encontrar y preferir alternativas más sostenibles que puedan comenzar a dar los primeros pasos hacia una próxima sustitución de plásticos en el sector de los envases alimentarios y de productos de uso único. El presente estudio considera una vista general de los principales conceptos relacionados al empaque de alimentos y el análisis de los diferentes grupos de alternativas sustentables para este segmento, tales como: materiales plásticos bio-basados, biodegradables, reciclados y películas de bio-plástico para recubrimiento de papel. En cada grupo se discuten los últimos avances científicos, así como los actuales desafíos e inconvenientes que obstaculizan su desarrollo a gran escala. Además, la investigación toma en cuenta varios casos innovativos y emergentes en el mercado como ejemplo y estímulo para el sector. Es por ello, que la tesis ofrece un análisis sobre el punto de vista económico de este sector planteando los principales factores de éxito, fortalezas, debilidades, oportunidades y obstáculos que presenta actualmente el mercado.

Palabras clave: Empacado de alimentos sostenible, eco-design, bioplásticos, plásticos reciclables, películas de plásticos de base biológica, economía circular

[spa]

Introduction

In the past 60 years, plastics have been the most developed family of materials on the market. Plastics have grown rapidly both in volume and in diversity to meet the demands of different sectors. This expansion has been seen notably in every materialization of the current frantic modern life. Today, plastic's world production is estimated at around 359 MT per year and is expected to double in the next 20 years (Ellen MacArthur Foundation, 2016). [ita]

Currently, about half of plastic's global production is employed in the packaging industry (European Bioplastics, nova-Institute, 2020), where food packaging has the largest application of nearly 60% (Matthews et al., 2021). Moreover, because of its short life and frequent use and disposal, these are often the ones mostly found in municipal solid waste. However, it is calculated that only about 14% of global plastic packaging is collected for recycling, 40% is landfilled, and 32% leaks out of the collection system (Ellen MacArthur Foundation, 2016).

Recycling represents a sustainable solution for plastics. Nevertheless, the current situation uncovers an inefficient plastic collection and sorting system, which must be considered to drive a circular plastic economy for food packaging applications (Attaran et al., 2017; Rujnić-Sokele & Pilipović, 2017; Rahimi et al., 2017; Rai et al., 2021).

Due to the current problem of pollution produced by plastic - in their majority of single use – the impact on food packaging has incremented and it has become extremely necessary to look for plastic materials' sustainable alternatives that reduce this pollution (European Bioplastics & nova-Institute, 2020).

The material used for food packaging must primarily protect the food during its lifetime and therefore act as a physical and chemical barrier to the outside environment. Food packaging should keep certain characteristics that will protect food, acting as a barrier against gases, water (in all states), and microorganisms. Additionally, as food contact material, they should not pose any risks for human health (Piergiovanni & Limbo, 2010; Yam & Sun Lee, 2012; Andrady, 2015).

Since its introduction, food packaging has provided food safeguards and has enabled food loss (Yam & Sun Lee, 2012). In addition, the various presentations, technology, shapes, and designs have made it possible to contribute to the industrial, economic, and social growth of this sector. Plastic food packaging has been a momentous factor in this development especially due to its innate properties, such as flexibility, transparency, lightness, and low cost (Piergiovanni & Limbo, 2010).

However, along with the vast portfolio of beneficial plastic's properties, one is found in particular, durability. That is to say, the same intermolecular property that makes it join between monomers and form long polymers is the one that in turn generates a long life for this material (Robertson, 2012; Andrady, 2015). This property also makes it resistible in different environments and not edible by micro-organisms due to these strong molecular bonds. That is why most conventional plastics, those that come from petrochemical sources, do not possess the ability to biodegrade.

Fortunately, scientific advances in recent decades, especially since the 90s, have allowed the development of other ways of producing plastics. Renewable sources are now part of the solution to obtaining more sustainable plastics. As a result, these are opening new possibilities for economic revaluation of used packaging through biodegradation, composting and recycling (Nakajima et al., 2017; Habel et al., 2018; Hatti-Kaul et al., 2020; Reichert et al., 2020).

The food packaging industry has also been affected by the pollution results from a linear plastic management system, requiring new findings on more sustainable alternatives.

Based on material science, product development and design, this research study seeks to respond to the latest developments and updates on sustainable alternatives for food packaging made of plastic material. The proposals presented consider the analysis corresponding to the material's life cycle, from its collection to its final disposition as a selection guideline.

The thesis is structured in five chapters. The first chapter comprises the main concepts related with food packaging in order to give a complete overview and thesis content insights necessary for the reader to understand the following chapters (II, III, IV) which are specialized in each sustainable category solution. The second chapter address the latest research about biodegradable bioplastics suitable for food packaging applications. The third chapter is focused on the recyclable plastics enabled for food packaging. Then, the fourth chapter is centered on the bioplastics for paper coating' applications, and the last chapter of the study focuses on the bioplastic food packaging market analysis.

The research addresses the challenges, limitations, and opportunities of each recognized sustainable alternatives for food packaging in plastic. Additionally, the present thesis develops each of these state-of-art solutions, considering its latest breakthrough in research, properties, design, and marketing of these.

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Chapter I.

SUSTAINABILITY AND FOOD INDUSTRY

1.1 Sustainability and packaging

1.1.1 Sustainability principle

According to Cambridge Dictionary, *sustainability,* refers to "the idea that goods and services should be produced in ways that do not use resources that cannot be replaced and that do not damage the environment" (Cambridge Dictionary, n.d).

The sustainability principle is widely applied to diverse areas such as cultural, technological, and political projects (James, 2014), as its etymology demonstrates, it is the "ability" to "sustain" over a period of time (Merriam-Webster, 1999). An ability that can be applied to everything, from business practices to energy and agriculture, the meaning can evolve and change to fit specific needs.

The sustainability concept had its beginning in Germany when it appeared in a handbook of forest management published in 1713 (Grober, 2007). Since the 20th century, the concept has been enriched and strongly used because of the global attention and concern about the overflowing levels of greenhouse gases emissions, the abuse of non-renewable resources as principal fuel for industries, transport and domestic use. In addition to the colossal waste accumulation in rural landfills and oceans. All of them, act as a severe detrimental of human life, ecosystems environment conservation.

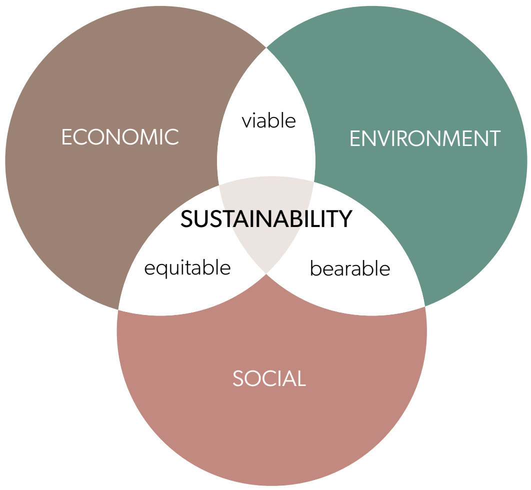
Nowadays, the most widely recognized definition of sustainability is based in *Our Common Future* (1987) report released by the World Commission on Environment and Development[1] , which states:

"Sustainable development is development that meets the present without compromising the ability of the future generations to meet their own needs".

This definition is reinforced by the 2005 World Summit on Social Development. Resulting in a common understanding about which accepts to achieve a sustainable development are necessary to maintain a balance between economic, environmental, and social pillars (Circular Ecology, n.d; Worldenergy, 2014).

[1] World Commission on Environment and Development (WCED). Conference organized by the United Nations. https://sustainabledevelopment.un.org/milestones/wced

Figure 1



The Three Pillars of Sustainability
Note. Figure 1 shows a visual representation of three principal components of sustainability. Which simultaneously considers and balances economic, environmental, and social goals from a microeconomic standpoint.
This perspective corresponds to the idea of the triple bottom line, concept developed by Elkington (1998, 2004) in his book *Cannibals with Forks: The Triple Bottom Line of 21st Century Business.*
Source. (Circular Ecology, n.d).

1.1.2 Packaging industry

Packaging is the technology and art of preparing a commodity for convenient transport, storage, and sale. Packages in the contemporary market are designed to protect goods from the hazards of handling and environmental conditions; to provide a manageable unit of the packaged product for the producer, distributor, and consumer; and identify the goods in a way that appeals to the potential purchaser. Packages must also be easy to manufacture and fill, while being inexpensive compared to the final packaged product ("Britannica Academic", n.d.).

Currently China is the world's largest packaging market with a global market share projection of roughly 28% by 2022; North America follows with a 22% of the global market share, and Western Europe project arrives at to 17%.

Globally, packaging is a diversified USD 850 billion plus (2018) industry with healthy growth prospects of 3 percent per annum (Feber et al., 2019). In Italy, the packaging sector has registered a positive trend in the last years, reaching a turnover of EUR 33,2 billion in 2018 (Lascone, 2020).

1.1.3 Sustainable packaging industry

Despite the favorable economic health of the industry in terms of turnover, the sector is challenged by the new circular economy approach, requirements, and regulations (European Commission, n.d.). The criteria for ranking packaging based on its sustainability is an active area of development. Several groups are publishing general guidance and metrics. Governments standards organizations, consumers, retailers, and packagers are considering different types of criteria. Nevertheless, the main goal is shared, to improve the long-term viability and quality of life for humans and the longevity of natural ecosystems. It means sustainable packaging must meet the social, environmental, and economical needs without compromising future generation's ability to meet their own needs. Therefore, sustainability must be contemplated as an active process of continuous improvement (Norton et al., 2013, p. 342).

For instance, the European Organization for Packaging and the Environment[2] (Europen, n.d.), recognizes the essential contribution to the sustainable production and consumption that packaging makes by contributing to the reduction of product waste and to the protection of resources, while acknowledging that packaging consumes resources along all stages of the supply chain. The following table 1 summarizes Europen's vision regarding the sustainable packaging.

Table 1. *Europen's Vision of Packaging's Contribution to Sustainable Development*

How packaging can contribute to sustainability?

Such packaging should:

- be designed holistically with the product in order to optimise overall environmental performance
- be made from responsability sources materials
- be designed to be effective and safe throughout its life cycle, to protect the product
- meet market criteria for performace and cost
- meet consumer choice and expectations
- be recycled or recovery efficiently after use

Note. Table 1 shows a visual representation of Europen's vision of packaging's role in sustainable development (Europen, n.d, as cited in Robertson, 2012).

[2] Europen-The European Organization for Packaging and the Environment. Is an industry organization presenting the opinion of the packaging supply chain in Europe on topics related to packaging and the environment, without favoring any specific packaging material or system. https://europen-packaging.eu/sustainability/packaging-environment.html

Many companies also recognize the value and responsibility of developing sustainable packaging solutions for their businesses. According to McKinsey & Company[3] (Feber et al., 2019), the most cutting-edge firms in the sector have already made bold sustainability declarations and commitments for years to come, about:

- Reduction of packaging material in terms of size, weight, and thickness
- Increased recyclability, reusability, or compostability of packaging material
- Increased use of recycled plastic in packaging material
- Design for recycling, circular design
- Customers education
- Proper waste management

The main objective for both key parts is to optimize the packaging throughout its life cycle. Optimization refers to the responsible and sustainable management of resources, feedstocks, and waste generation. Approach based on the holistic design of the packaging. To define sustainability in the packaging industry is to analyze the industry through a holistic perspective taking care all stages of the supply chain.

Currently, worldwide methodologies are applied to assess the sustainability of processes, products, and services. Such as Life Cycle Assessment (LCA) is defined by UNE-EN ISO 14040[4] as a technique for assessing environmental aspects and potential impacts associated with a process, product, or a service. While Life Cycle Inventory (LCI), is a methodology built based on the inventory of input and output flows for a product system (Webb & Kosseva, 2013, Chapter 15, p.265).

One of the biggest concerns referred to sustainability in the packaging industry, is the waste generation and all its implications. In Italy, packaging waste represents approximately 26-28% of municipal solid waste generated annually (Consorzio Nazionale Imballaggi, 2018). In 2017, the 27 European Union (EU) Member States reached a total volume of 77.5 million tonnes of packaging waste materials. With a mean rate of 67.5% for recycling and 81.7% for recovery. Despite these favorable rates, the recycling and recovery ratio change among materials. For instance, in 2017, Italy recycled only around 40% of the total plastic packaged waste (Eurostat, 2020).

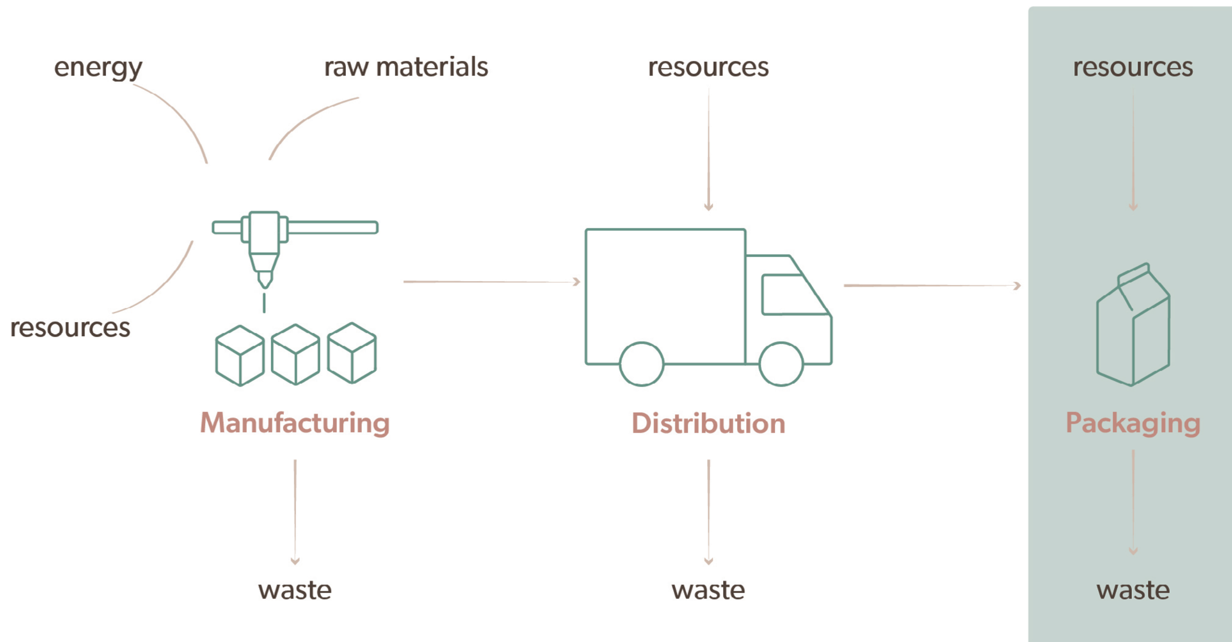
As explained before, applying sustainability as an initial constrain in the packaging industry, it opens a large spectrum of application centers. These application centers, showed in the following figure 2, can be grouped into three significant segments. These segments are the industrial process, the distribution, and storage associated, and the lifecycle of the packaging as a product by itself.

In the present study, due to the material and design science perspective, the application of the sustainability principle will focus on the packaging as a product. Which implies a product development design perspective throughout the packaging life cycle.

[3] McKinsey & Company. US-based management consulting firm. https://www.mckinsey.com/

[4] ISO 14040 is an international standard in environmental management and Life Cycle Assessment (LCA) issue by ISO (the International Organization for Standardization). https://www.iso.org/obp/ui#iso:std:iso:14040:ed-2:v1:en

Figure 2



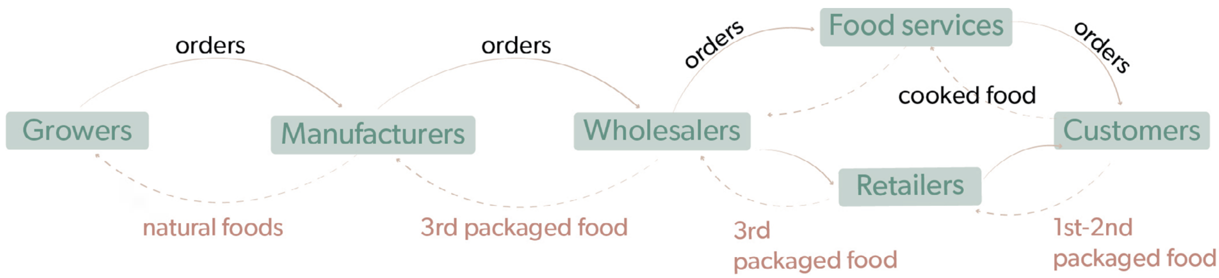
Environmental Impact Centers in the Packaging Industry
Note. Where waste contemplates the substances (in all states) discarded after its application. Which consider water and raw materials released to air, land, and water.

1.2 Sustainability and food industry

1.2.1 Food industry

The food industry is a massive business and a unique industry because almost everybody depends on, is influenced by, and is impacted by it. Therefore, the supply of safe, affordable, and plentiful food is essential to the well-being of a nation. The food industry comprises all the supply chain (Figure 3). Farmers, growers, processed plants, wholesalers, retailers, and food services until the product arrives at the final customer (Alberti, 2016; Dudbridge, 2011, Chapter 1).
The food and beverage industry is the EU's biggest manufacturing sector in terms of jobs and value added, in the last 10 years, EU food and drink exports have doubled, reaching over EUR 90 Billion and contributing to a positive balance of almost EUR 30 Billion (European Commission, n.d).

Figure 3



Overall View of the Food Industry Supply Chain

Note. Figure 3 shows a graphic representation of the food industry supply chain based on the book Handbook of lean manufacturing in the food industry
Source. (Dudbridge, 2011).

1.2.2 Sustainable food industry

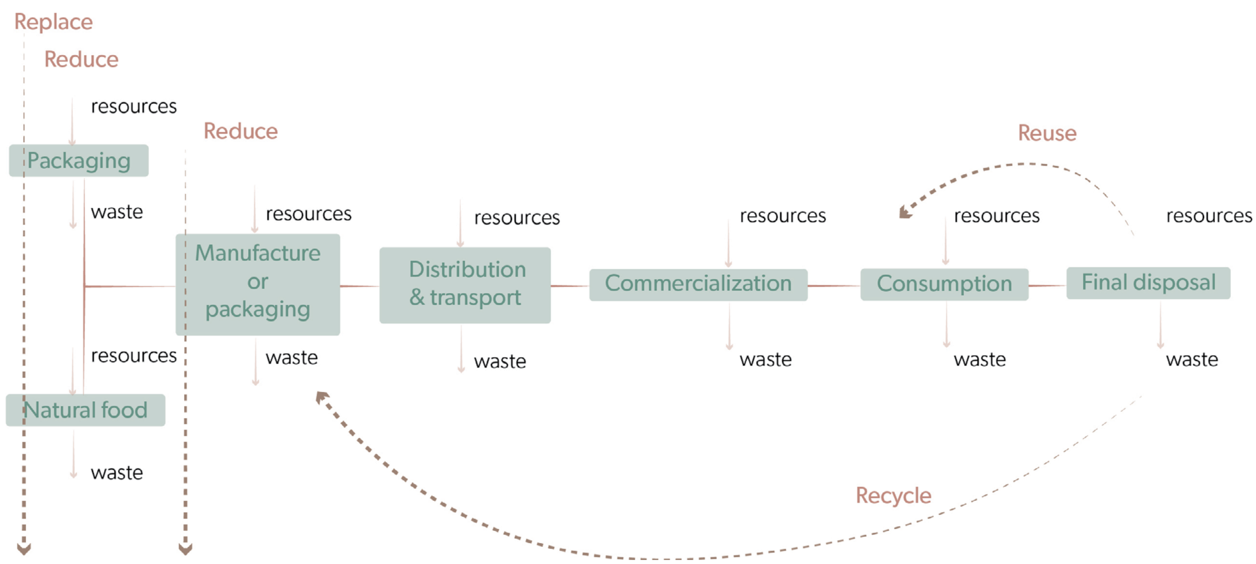
In the last decades, the environmental changes, raised pollution indexes, social inequality and poverty, the increasing number of the world's population, and the wasteful and careless utilization, have increased the public's awareness about sustainability, especially in the food industry. Which have also led to boost fair farming and cultivation, claim clean manufacturing processes, promote fair commerce and businesses, and change traditional discard methods to ecological ones, within the biggest concerns.

Food industry also causes environmental impacts related to the inefficient use of associated natural resources, such as water, oxygen, energy, and land. Additionally, the high amounts of food and packaging waste strongly contributes to ecosystems degradation, soil erosion, GHG emissions, and climate change.

The food sector has reported to utilize around 30% of the world's total energy consumption and 22% of total GHG emissions (Tolnay et al., 2020). Evidently, most of food products are not sustainable and is needed of drastic shifts in all steps of the supply chain.

Defining sustainability in the food industry, as any other industry, comprises a wide range of applications. As it is showed in the following figure 4, each box represents an application center throughout a sustainability perspective. Each application center implies reorganizations at all levels. Such as sources and feedstocks, transportation, mechanisms and technologies, and waste management (food and packaging).

Figure 4



Food Industry LCA Overview

Note. Representation of an overview of the whole food industry supply chain from an LCA approach and waste management hierarchy principle.
Source. (Norton et al., 2013, Chapter 14)

The present study contemplates the food packaging functions and properties required to fit each stage based on a sustainable perspective.

1.3 Food packaging

Food packaging lies in the core of the modern food industry. Nevertheless, for millennia, humans stored their food in containers they found in nature such as dried gourds, shells, hollow logs, and leaves. From its origin these containers have allowed food transport, handling, and protection.

The art and science of food packaging have evolved a long way from those origins. Today, different materials and techniques contribute to a safely protection, distribution, and consumption. Furthermore, food packaging can enhance food safety by preventing bacterial contamination. Being a key part to ensure the sanitation of consumers.

In addition, food packaging extends the shelf life of products and reduced food waste. Paradoxically, using more packaging to reduce food waste creates another waste problem. (Environmental Health Perspectives, 2012). In fact, in Italy, approximately 73% of the packaging produced correspond to food and beverage packaging (Lascone, 2020).

1.3.1 Food packaging functions

It is impossible to cover modern life's primary necessities and the challenges of a growing population without an efficient food packaging technology and system. Therefore, the design of a food packaging solution should admit the following fundamental functions (Andrady, 2015, Chapter 5, p. 126; Piergiovanni & Limbo, 2010, Chapter 1, P. 3-6; Yam & Sun Lee, 2012, Chapter 1, p. 5-6).

1. Containment

It is the oldest function and the original one, it refers to hold and support the principal product (food).

2. Protection

The packaging must act as a protective barrier from the environment to maintain the quality and safety. This barrier can be against mechanical stresses, influences of light, humidity, and oxygen. Also, from other possible chemical and biology contaminants forms coming from the exterior, and unwanted or fraudulent manipulations, protection must always be calibrated to the specific needs of the food and its distribution cycle.

3. Communication

The food packaging must drive of important messages to the consumer and stakeholders, from its manufacturing to the end of its life. This information is related to the commercial purpose (label, decoration, discount, gadget), to useful information for the consumer (nutritional information, usage advise, recipes). It is also about compliance with regulations (trademarks, marks, dates, metrological indications), and its identification (bar code, holograms).

4. Service

Is the most recent function and is centered on the modern user and their preferences and requirements. Some examples are the opening facilitation, the application of flexible materials, and the aptitude for treatment in microwave ovens.

5. Logistic

The food packaging also must help the product flow. The economization result from the optimization of the logistic aspect of the packaging is huge and justify substantial investments.

1.3.2 Types of packaging

Due to the capacity to hold other containment, packaging is divided into three main categorizations. This classification also applies to food packaging (Piergiovanni & Limbo, 2010, Chapter 1, p. 2; Esposito, 2019).

Primary packaging
Referred to the material or container in direct contact with the product; also called sales or representation packaging. It represents a sales unit for the final consumer.

Secondary packaging
Related to the containment system of one or more primary containers. Therefore, in direct contact not with the product but with the primary container. Also called multiple packaging, it is designed to constitute a grouping of primary packaging at the point of sale. If the product is removed from its secondary packaging, its characteristics and commercial value are not modified.

Tertiary Packaging
Set of several primary and secondary containers specifically designed for transport and handling, therefore, also called transport packaging. However, it does not refer to containers (which may contain multiple tertiary packaging) or to bulk packaging (large departmental boxes, for bulk goods such as large bags, drums, and trolleys), for which is used the quaternary expression packaging.

Figure 5 exemplifies the three categories of food packaging. The primary and secondary packaging are in direct contact with the consumer and both can be found at the point of sale. Due to the principal purpose of the secondary packaging, is promotional.

Figure 5

Types of Packaging
Source. (Piergiovanni & Limbo, 2010, Chapter 1, p. 2)

1.3.3 Food Packaging properties

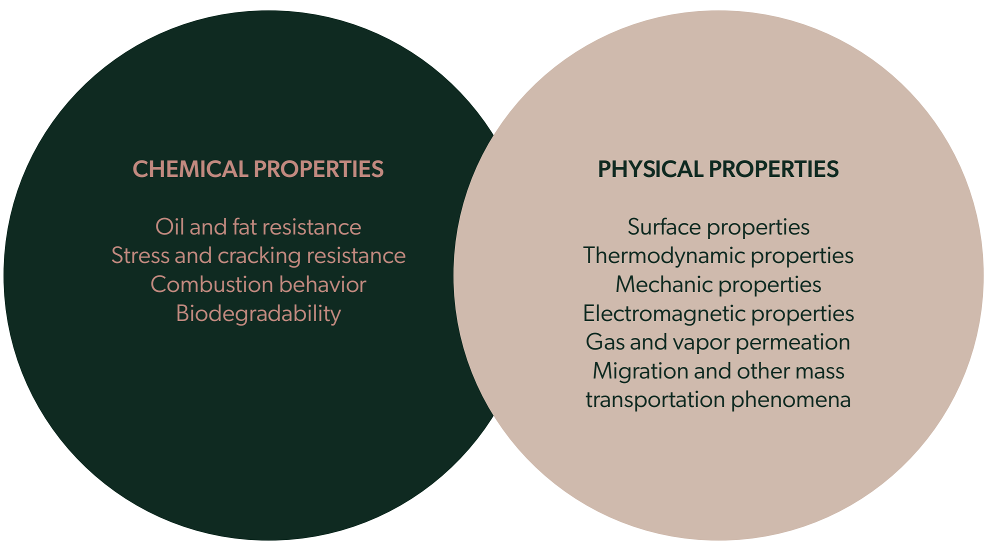
The food packaging properties are based on the type of family material applied and can be divided into chemical and physical properties. Both types of properties allow a special performance from the packaging in relation to the food it will contain. Figure 6 presents the most relevant material properties associated with food packaging solutions (Piergiovanni & Limbo, 2010, Chapter 2-4).

Properties that depend on the chemical nature of the material (atomic and molecular) are defined as chemical. The chemical structure of a material is defined by the chemical nature of its components and its organization. The modification of these chemical properties also causes profound changes in numerous physical properties.

The physical properties of packaging materials are related to phenomena that do not imply changes in the chemical structure, which are often reversible. These physical properties are always associated with defined physical quantities, which are conveniently measurable under objective and instrumental techniques.

The chemical and physical properties of a food packaging highly influence its containment, protection, service, communication, and logistic functions. Furthermore, they are critical to define the shelf life of the product.

Figure 6



Packaging Material Properties
Source. (Piergiovanni & Limbo, 2010, Chapter 2-4)

Chemical properties

The most essential chemical properties for the materials used in food packaging can be identified in their oxidation and combustion behavior. The response to the biological agents and corrosion

resistance is important to consider material's reaction to both physical and chemical aggressive agents.
Chemical properties are measured by largely empirical or simply comparison methods. In other cases, some physical properties are measured whose variations are correlated to the chemical transformation. The specific chemical properties of packaging material are used to assess and identify the product's exact composition. Nevertheless, most of the time, they are measured to test the suitability for a specific use. Assessing the chemical properties related to the packaging is crucial to ensure the aseptic properties of the material and the consumer's safety. These properties are generally measured under controlled conditions of exposure over the so-called *Abuse Test* performed according to standard methods presented in table 2.

Table 2. *Assessment of some material properties through ASTM[5] methodological*

Standard	Description
ASTM D543	Chemical resistant test (evaluating of the effects of controlled exposure to chemicals on weight, size and appereance)
ASTM D570	Water absorption (evaluating of weight gain following immersion in water)
ASTM D3929	Evaluating of stress cracking (resistance to agressive chemicals and the environment)
ASTM F119	Penetration of grease (measurements of the penetration time of oil under pressure)

Source. (Piergiovanni & Limbo, 2010, Chapter 2, p.14)

Physical properties

The physical properties are classified into five categories: surface, thermal, mechanical, electromagnetic, and diffusion properties.

The surface properties of a material are critical to the success of important technical operations such as adhesion and printing; and for the optimization of functional characteristics, such as resistance to water and oils, and brilliance.

On the other hand, a material's thermal properties describe its behavior in response to the thermal stresses. Which can be during a heat exchange process or because of a temperature variation. The main thermal properties affecting the packaging materials sector are thermal conductivity, thermal capacity, the thermal expansion coefficients, the temperature service, the transition temperatures, and the calorific value.

[5] ASTM International. American Society for Testing and Materials
https://www.astm.org/

The mechanical properties include those physical properties that describe the behavior of a solid exposure to the application of a force. Which can be the weight of the body itself or by external stress. The knowledge of the mechanical performance of a packaging or packaging material is fundamental to evaluate the suitability for a specific use and to discriminate between similar materials. Therefore, the mechanical performance is always included in the material technical specifications. It also includes the friction and resistance properties, the response to dynamic stress and hardness.

The electromagnetic properties include all the characteristics that describe the behavior of a material subject to irradiation with electromagnetic radiation (bright or not). Electromagnetic radiation is defined as the simultaneous propagation in the space of energy associated with electric and magnetic fields, which vary over time. This phenomena explains important packaging properties such as reflection, refraction index, transparency, opacity, brilliance, or gloss. Furthermore, this property can assess the behavior of a material irradiated with microwaves.

Evidently, the most crucial properties for food packaging are the diffusion properties. They are composed by the permeation of gases and vapors, the migration, and other mass transport phenomena.

The mass transport phenomena of aeriform (gases and vapors) through food packaging is extremely important since they are almost always related to events that affect the quality and safety of products. The entry of oxygen into a package can cause lipid oxidation, the appearance of unpleasant odors, a proliferation of microorganisms, loss, or variation of color. The release of carbon dioxide can cause the loss of effervescence or damage a packaging in a protective atmosphere. The entry or leakage of humidity is responsible for important variations in the consistency and possible microbial alterations. While having an adequate exchange of oxygen, carbon dioxide and water vapor it is indispensable in the packaging of fresh vegetables to support natural aerobic respiration and avoid sensory alterations.

The treatment of gas and vapor permeation through the thickness of materials, certainly does not exhaust the vast theme of mass transport phenomena affecting food packaging. From the point of view of food safety, even more important are those transport phenomena between the interface food/packaging. It may involve the transfer of substances from the packaging to food (migration or transfer) or food to packaging, defined as absorption or adsorption events.

1.3.4 Food Packaging Materials

Today, there is a wide range of materials utilized for different food packaging applications. Other packaging materials offer various advantages. For instance, glass preserves well the organoleptic properties of food. Cellulose derived materials such as paper and paperboard have a low-cost production and are easy to print on. They are also lightweight, which reduces the logistic costs. Steel and aluminum can also be bound to paper and plastic films, which enable more versatility in the typology of packaging. And plastics have revolutionized the packaging industry because of their mix of properties. They are highly moldable, lightweight, low-cost, easy to seal and durable

(Environmental Health Perspectives, 2012).
All food products could be packaged in metal cans or glass containers, but usually they can be more efficiently and economically packaged in one or more of a variety of structures such as cartons, pouches, bags, and wraps. In Italy, for instance, 70% of food and beverage packaging produced (tonnes) correspond to glass, 16% comes from cellulose materials, 12% are plastics, 1% is aluminum, and 1% steel (Lascone, 2018).

Materials also strongly influence the packaging cost. Design guidelines such as the geometry, thickness and weight will determine the total food packaging cost. Which arrives approximately to 20% of the total product cost (Dudbridge, 2011, Chapter 1, p. 10).

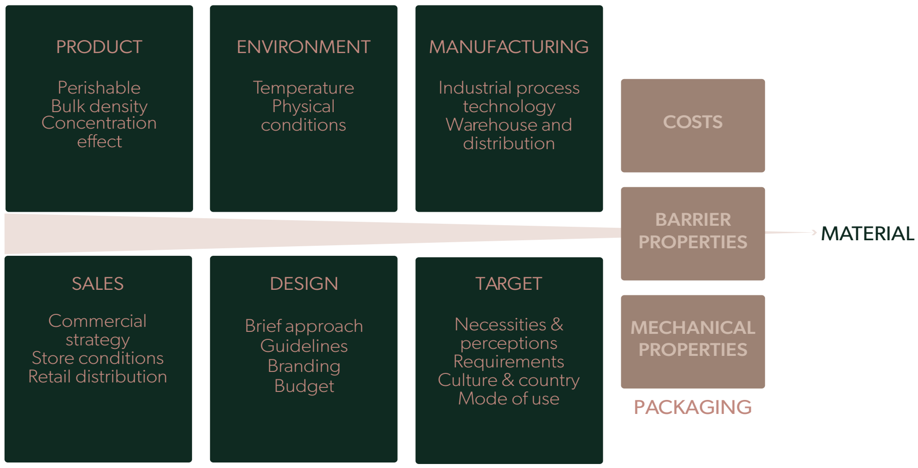
Food packaging material selection

There is no unique method for selecting proper material for a food packaging because the selection will also depend on the company strategy and closed variables, limitations, and available resources. Although, there are more than 100 different aspects to be considered when developing a new package, they may be conveniently grouped under six principal categories.

Figure 7 represents these six factors and the most critical variables they are composed of. To select a suitable material, product variables must be considered,meaning the type of food the packaging will content. The environment should also be considered, the location in where it will be handled, stored, and sold. Another important factor to contemplate is the industrial process and technology the product requires for its production.

Furthermore, the sale strategy is fundamental to define the type of packaging and the material required. The design approach will address the material selection through the brand guidelines and project budget. In addition, the material selection must consider the consumer for who is designed for, which means their perceptions, needs, and requirements.

Figure 7



Determining Factors to Packaging Material Selection
Source. (Robertson, 2009, Chapter 1, p.3; Robertson, 2012, Chapter 1, p. 4; Traitler et al., 2014, Chapter 3, p. 43-63).

These six factors are influenced direct or indirectly between themselves, and they will determine the type and degree of protection the food will need. Therefore, the barrier and mechanical properties of the packaging are required, and these can be employed one or a combination of materials (Robertson, 2009, Chapter 1, p.3; Robertson, 2012, Chapter 1, p. 4; Traitler et al., 2014, Chapter 3, p. 43-63).

1.3.5 Plastics in food packaging

The adjective *plastic* comes from the Greek *plastikos*, meaning easily shaped or deformed. Plastics is a generic term for macromolecular organic compounds obtained from molecules with a lower molecular weight or by chemical alteration of natural macromolecular compounds.
The utility of flexible sheet materials depends on the properties of a special kind of molecular structure: long, flexible molecules interlocked into a strong and nonbrittle lattice. These structures are built by the repeated joining of small basic building blocks called *monomers*, the resulting compound is called *polymer*, derived from the Greek roots *meros* meaning parts, and *poly* meaning many. Differences in monomer's chemical composition, in the structure of the polymer chains and interrelationship of the chains determine the various polymeric materials (Andrady, 2015, Chapter 3; Robertson, 2012, Chapter 2).

Synthetic plastics have a relatively recent history. The industrial production of the main resins dates from 1930-1940. However, in about half a century, a great number of different applications have been conquered like no other material (Piergiovanni & Limbo, 2010, Chapter 8, p. 206). In fact, during the period (1961-2012), plastics consumption grew by over 4800% (Andrady, 2015, Chapter 1, p. 20).

Thanks to its versatility, low-cost production, lightweight, and bio-inertness, plastics can be used to produced diverse food packaging applications. In Italy, the food packaging sector absorbs over half of the plastics materials used in packaging, which are mainly used to produce flexible packaging (Piergiovanni & Limbo, 2010, Chapter 8, p. 206).

Factors that influence plastic properties

There are special factors that determine plastic's properties and consequently, the packaging properties made of them. Plastic properties are determined by the chemical and physical nature of the polymers used in their manufacture. These properties are influenced by the molecular structure, molecular weight, degree of crystallinity and chemical composition of the polymer (Robertson, 2012, Chapter 2; Piergiovanni & Limbo, 2010, Chapter 8).
Due to the existence of many plastics, and new plastics are continuously synthesized, table 3 shows the main criteria commonly used to classify them. These criterias of classification are based on the factors that make unique polymer materials.

Table 3. *Main criteria adopted to classify plastics*

Classification criterion	Characteristics
Nature of raw materials	Natural, synthetic and partially synthetic
Polymerization mechanism	Addition, condensation
Tacticity	Isostatic, atactic, syndiotactic polymers
Molecular weight	Mono and polydisperse polymers
Behavior to heat	Thermoplastics and thermosets
Glass transition temperature	Rubbery and glassy polymers
Morphology	Amorphous polymers, crystalline, semicrystalline
Structural organization	Omo and copolymers, blends and alloys

Source. (Piergiovanni & Limbo, 2010, Chapter 8, p. 207).

Most plastics used in food packaging have a petrochemical origin, which is synthetic. Meanwhile, biopolymers, polymers of animal, vegetable or microbial origin, may find useful applications alone (natural) or in combination with synthetic ones (partially synthetic). Due to the importance and attributes of biopolymers from a sustainable approach, these plastics will be further discussed in section 1.4 and in the following chapters.

Polymerization processes

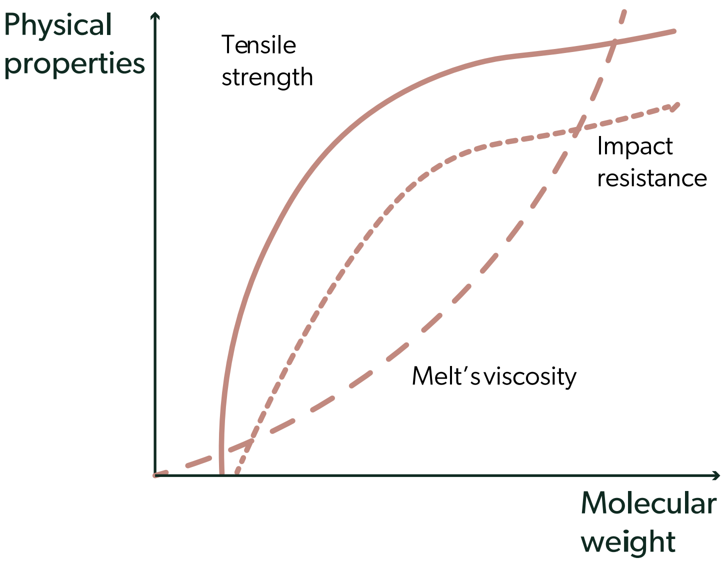
Thermoplastics can be made by joining a sequence of monomers. The composition of thermoplastics is done by a process that involves the joining of monomers to form polymers that have the same atoms as the monomers in their repeating units, which is called *addition.* Under normal conditions with the usual catalysts, if the spatial arrangements of the polymer branches are random, such polymers are called *atactic.* Some processes give products in which the branches are arranged in an orderly manner, these are called *isotactic polymers.*

Plastics are also prepared by *condensation* which involves two active sites joining to form a chemical bond, a small molecule being the result. In this case, the starting monomers are not identical to those of which the chains are to be composed.

Molecular Weight

The molecular weight is defined by the number of units that make up a single polymer macromolecule, which is known as the degree of polymerization or DP. When the DP is low (10-20), the polymer is presented in its liquid state at room temperature, while a polymer with a DP near 1000 is solid. High and very high molecular weight fractions influence melt viscosity, mechanical strength, and the fragility in solid state (Figure 8). Meanwhile, those with low molecular weight account for viscosity and adhesiveness of polymers.

Figure 8



Relationship between some physical properties and molecular weight of polymers

Source. (Piergiovanni & Limbo, 2010, Chapter 8, p. 209)

Behavior to heat

The polymers behavior to heat facilitates the differentiate *thermoplastics* polymers from *thermosetting* ones. The first are heated to temperatures above room temperature, they soften and finally meet at a temperature corresponding to the maximum freedom of movement for their macromolecules. Since this behavior is reversible, thermoplastics polymers can be easily hot-molded, forged into different shapes and sizes, and recycled. Most plastics used in food packaging are thermoplastics.

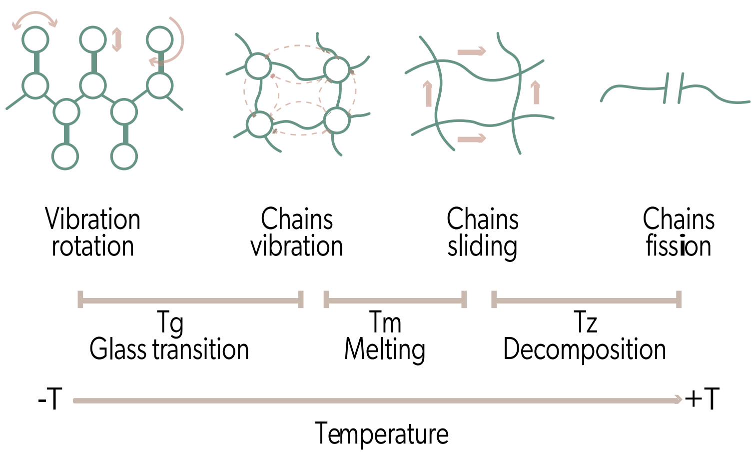
On the other hand, thermosetting polymers are characterized by the presence of unsaturated chains and a reticular structure. Therefore, any subsequent heating after its production, would have the effect of breaking the cross-links that stabilize the structure. In fact, they are much more rigid and robust that thermoplastics ones. Thermosetting plastics are rarely used in packaging, its use is limited to some closing accessories, to some adhesives and internal protection lacquers of metal boxes.

Glass transition temperature

Numerous physical and chemical-physical characteristics of polymers depend on the temperature and particularly on the glass transition temperature (Tg). The Tg is determined by the strength of the intermolecular bonds and the flexibility and length of the chains (Figure 9). In relation to the room temperature, it can be defined as *rubbery* or *glassy* to explain many behaviors.

The plastic materials that at room temperature are above the Tg, have a rubbery behavior. Which generally corresponds to a greater permeability to gases and easier workability. Additionally, those below the Tg, express lower diffusion and permeability coefficients and better mechanical resistance properties. Among the plastics used in food packaging materials, there are both rubbery and glassy polymers, which sometimes, blend together in composites structures (e.g. polyethylene/polyamide).

Figure 9



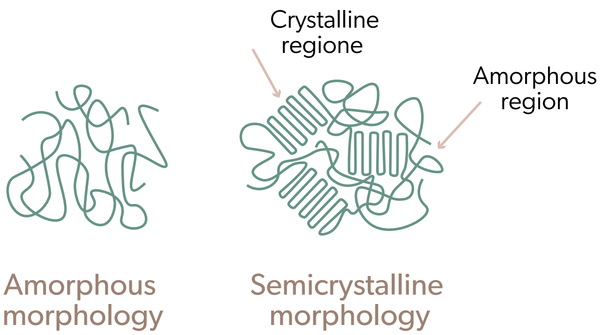
Thermal mobility of polymeric macromolecules

Source. (Piergiovanni & Limbo, 2010, Chapter 8, p. 211)

Morphology

The morphology of polymeric macromolecules allows distinction from crystalline, amorphous, and semicrystalline polymers (figure 10 and table 4). The spatial organization of polymeric macromolecules is mostly amorphous which means there is a disordered arrangement of macromolecules.

Figure 10



Polymeric macromolecules morphology
Source. (Piergiovanni & Limbo, 2010, Chapter 8, p. 212)

Table 4. *Properties of polymers in the amorphous and crystalline state*

Amorphous polymers	Crystalline polymers
Transparent	Opaque
Melting temperature non-identifiable	Specific melting point
Relatively weak and flexible	Relatively strong and fragile
High or medium gas permeability	Low or medium gas permeability
Moderate chemical resistance	Good chemical resistance
Few opportunities to intervene for modify its general properties	Many opportunities to intervene for modify its general properties

Source. (Piergiovanni & Limbo, 2010, Chapter 8, p. 213)

Molecular Structure

Polymers are molecular materials with the unique characteristic that each molecule is either a long chain or a network of repeating units. When plastics are made up of only one type of monomer, that is, of a single fundamental unit and it is repeated along the macromolecular chains, they are referred to as *homopolymers*. The *copolymers,* on the other hand, are obtained by jointly polymerizing two or more different monomers (comonomers).
Many polyolefins, the polymer most used in food packaging, are available in the form of copolymers. The presence of the comonomer along the chains often include ramifications on the main linear chains, influencing all the properties of the final polymer and its density.

Plastics typology in food packaging applications

There are different types of plastics used for food packaging solutions. Each particular class possesses different chemical and physical properties, which determine its applications. Also, each class requires different treatment, reprocessing methods, and types of collection (Piergiovanni & Limbo, 2010, Chapter 8, Side, 2002, Chapter 12).
The following table 5 represents the different types of plastics used in food packaging solutions. It shows the principal mechanical and barrier properties, and its applications.

Table 5. *Plastics typology and applications*

Symbol	Polymer Type	Properties	Type of food	Type of packaging
01	**PET** Polyester	•Low permeability to water and oxygen •Resistant to aromatic fats, oils, acids, and alkalis •Very high tensile and impact strengths •High hardness and rigidity •Fair temperature resistance	•As the sealing web for vacuum/gas •Flush packaging of cured meat, cheese, or fresh pasta. •Vacuum-metalized polyester film •Used for large pouch packaging of wine, syrup, and bulk tomato and fruit products	•Rigid bottles •Transparent bottles •Blended films
02	**HDPE** High-Density Polyethylene	•From flexible to rigid, depending on the density •Very low permeability to water and quite high to oxygen •Resistent to acids, alkalis, oils, alcohols and stress cracking •Not resistant to oxidizing agents and hot organic solvents	•Beverage (e.g. milk, water) •Cereals box liners	•Glasses •Trays •Semi-rigid bottles •Rigid bottles •Flasks •Drums •Blended films •Wrapping films •Boxes •Bags
03	**PVC** Polyvinyl Chloride	•Generally low permeability to water and oxygen •Transparent and soft •Resistant to diluit acids and alkalis, non-polar solvents, oils and fats, gasoline	•Wrap for: Fresh red meat Poultry Vegetables	•Glasses •Trays •Rigid bottles •Transparent bottles •Stretch films •Wrapping films •Sacks
04	**LDPE** Low-Density Polyethylene	•From flexible to rigid, depending to the density •Low permeability to water, very high to oxygen •Not very transparent •Resistance to acids and alkalis •Not very resistant to oils and alcohols •No resistance to oxidizing agents, hot organic solvents and surfactants	•Bread •Fresh vegetables and fruit •Stable food products •Squeezable food botles •Bags for frozen food	•Semi-rigid bottles •Flasks •Stretch films •Wrapping films •Blended films •Coatings •Bags •Sacks
05	**PP** Polypropylene	•Rather rigid and resistant •Excellent water vapor and oxygen resistant •Resistant to acids, alkalis,oils, alcohols and stress cracking •Not resistant to oxidazing agents and hot organic solvents •Poor gas barrier properties •Not very transparent	•Wrap bakery products •As lamination plies for snack chip pouches •Pastas •Margarine tubs •Microwaveable meal trays •Freash meat	•Glasses •Trays •Rigid bottles •Single-use cassettes •Boxes •Wrapping films •Blended films •Coatings •Bags
06	**PS** Polystyrene	•Rather rigid and very fragile •Very low permeability to water, medium to oxygen •Transparent and bright •Resistant to acids, alkalis, oils, and lower alcohols •Not resistant to oxidizing agents, organic solvents, stress cracking and UV	•Beverage (e.g. yogurt pots) •Foam hamburger boxes •Eggs cartons	•Glasses •Trays •Plates •Single-use cassettes

Note. Table 5 is a systematic representation of the plastics types used in food packaging applications.
Source. (Piergiovanni & Limbo, 2010, Chapter 8; Side, 2002, Chapter 12)

1.3.6 Regulations

In order to give a propitious scope in terms of the legislations that regulate food packaging, it is preferable to initiate with the definition of suitability from its functional and food approaches. Luciano Piergiovanni and Sara Limbo have given an appropriate definition in their book *Food packaging – materiali, tecnologie e qualità degli alimenti,* explaining the suitability for a food product, dividing the term in two complementary and independent components (Piergiovanni & Limbo, 2010, Chapter 4).

Functional suitability

Functional suitability refers to the capacity of a container or material to guarantee the conservation required for the product, to offer a pleasant or captivating image and to withstand normal transport or use conditions.

Food suitability

Food suitability concerns to the safety of the material destined to be in contact with food, which must not undergo modifications in their chemical, microbiological, or sensory nature.

To achieve the commercial success of a food product it is indispensable to maintain both, functional and food suitability. The food suitability of packaging materials and containers has been regulated in Italy since 1962 by a state law and, since 1973, by interventions of the Ministry of Health; only later it acquire priority importance at the European level in the harmonization of rules within the European Union.

The following indicates principal sections related to the food packaging, regulated by Italy and the EU legislations.

1. *Principle of "inertia" of the material and "purity" of food products*
   According to this principle, materials and objects must not migrate to food components in quantities representing a danger for human health or an unacceptable modification in the food composition or an alteration in their organoleptic characteristics.

2. *Positive labeling*
   The materials and objects intended to be in contact with food must be accompanied by a document certifying their suitability, the indication "for food" or an appropriate label, or by an indication that highlights any limitation of use.

3. *Standardization of procedures to verify compliance*

4. *Compliance of packaging*

The general rules regarding to the food suitability of an object intended to be in contact with food (FCM, Food contact materials), are regulated in the art. 11 of Law 283 of 30.4.1962, and have been further detailed and clarified in subsequent and more specific rules.
At the European level, the most recent and complete proposition is in the EC Regulation

1935/2004[6]. Below are the most essential measurements regulated regarding the food suitability in relation to the packaging:

Conformity of composition

All the components used to produce an FCM must be safe and listed in the assigned *positive list*. The *positive list* contains a list of features normed by the Italian law as safe to be in contact with food. In some cases, the limits of use per type of component measured by its weigh or area are excluded for specific uses. The declaration of its positive list must accompany all food packaging material.

Global Migration

The global migration limit is a pre-requisite for material inertia where the law establishes a limit for the possible interaction between food and packaging.
Today, all plastics objects are subjected to a global migration limit (OML, Overall migration limit) equal to 10 mg dm^{-2}(mass released per unit area of the packaging material) or 60 mg kg^{-1} (or ppm, packaging mass unit released per food unit mass).

Specific Migration

The specific migration limit (SML, specific migration limit) is settled whenever a particular substance, potentially migrating from a packaging, presents a risk to the consumers' health or even only to the organoleptic of the product. The limit is generally expressed in mg kg^{-1}, but can also be converted to mg dm^{-2} when referring to objects with a capacity greater than 10 L or less than 500 mL or to items that cannot be filled. The limits depend on the dangerousness of the substance and can be equivalent to the analytical zero.

Functional Barrier

The concept of functional barrier had already been proposed by Italian legislation in the Ministerial Decree of 21.3.1973[7], which in art.5 and European legislation in the recently Directive 2007/19/EC[8], which introduced the concept of a functional barrier of plastics material placed inside a packaging material that prevents or limits migration to the food.
If separated from a functional plastic barrier, unauthorized substances may also be used, if they meet specific requirements and the migration remains below to 0.01 mg kg^{-1} (10 ppb) in the product or in the food simulant.

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[6] Regulation in force (EC) No 1935/2004 of the European Parliament and of the Council of 27 October 2004 on materials and articles intended to come into contact with food and repealing Directives 80/590/EEC.
[7] Italian Ministerial Decree (2006, consolidation) concerning the hygiene requirements of packages, containers and tools destined into contact with food or substances for personal use.
[8] Commission Directive 2007/19/EC of 30 March 2007 relating to plastics materials and articles intended to come into contact with food and Council Directive 85/572/EEC laying down the list of simulants to be used for testing migration of constituents of plastics materials and articles intended to come into contact with foodstuffs.

Sensory suitability

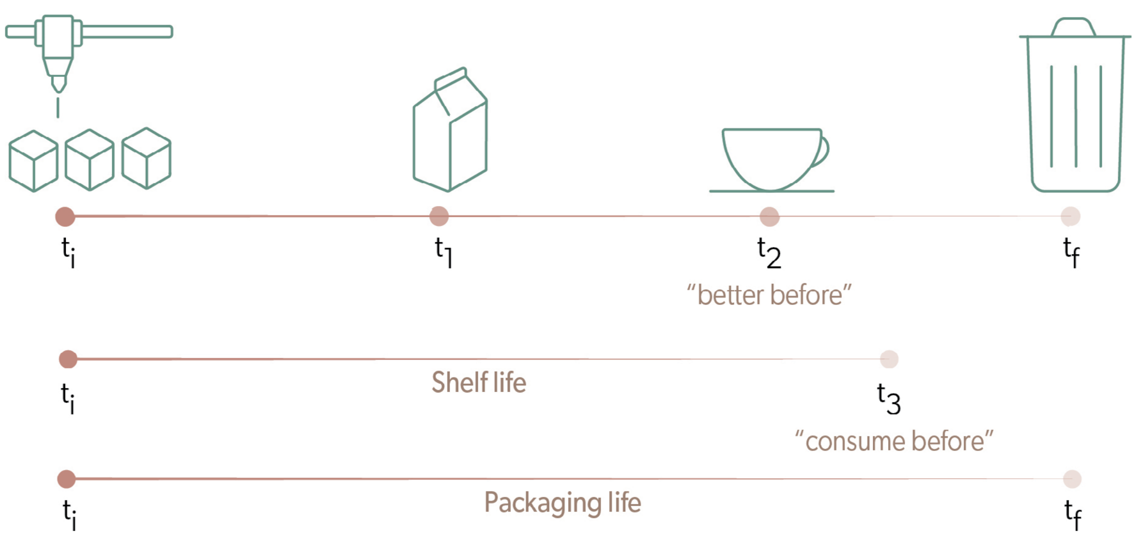
Since 1962 (Law283/62), the risk of sensory contamination of food from the transfer of packaging was foreseen and, among the general requirements of EC Regulation 1935/2004, the organoleptic food functions have been regulated. Nevertheless, this regulation does not consider the possibility of transferring odorous or sapid substances (by absorption or reaction) from the food to the material in contact. For this reason, many voluntary standardized bodies (ASTM, BSI[9], DIN[10], UNI[11]) have proposed standards and procedures to evaluate the sensory impact of materials and containers.

The legal regulations governing the production and use of food packaging represent a safe instrument of protection for the consumer. However, the interactions between food, packaging and the migration phenomena are substantially unavoidable events.
The risk for the consumer associated with these events can be considered very modest, since the provisions of the law ensure ample safety margins.

1.3.7 Lifecycle

Generally, the food product lifetime is defined as its shelf life. Nonetheless, from a sustainable perspective, the lifetime of a food product should be established and planned considering the whole product life, it means the shelf life of the food content plus the packaging lifetime. Figure 11 represents the difference in time between packaging life and shelf life of a food product.

Figure 11



Food Product Life Overview
Source. (Robertson, 2012, Chapter 12; Robertson, 2009, Chapter 1)

[9] BSI. British Standards Institution. https://www.bsigroup.com/
[10] DIN. Deutsches Institut für Normung. https://www.din.de/en
[11] UNI. Italian Organization for Standardization. https://www.uni.com/

Shelf life of packaged food

To understand the importance of calculating and regulating the shelf life of a food product, a brief mention of the food quality concept is required. Food quality is "the combination of attributes or characteristics of a product that have significance in determining the degree of acceptability of the product to a user" (Robertson, 2012, Chapter 12).

The overriding reason why it is vital and compulsory to determine the shelf life of a food product is because without this information is not possible to ensure commercially and legally, the sanitary conditions of the food content. Therefore, consumer health, due to most foods and beverages quality decreases with storage or holding time. For consumers, there will be a finite length of time before the product becomes unacceptable; this period in which is goes from production to unacceptability is referred to as shelf life. The shelf life of packaged food plays a crucial role in the organization of production, distribution, commercialization, and product consumption. Furthermore, the shelf life determines the packaging characteristics. Therefore, the definition of the "use by", "best before".

The Institute of Food Science and Technology (IFST[12]) has defined shelf life as "the period during which the food product will remain safe; be certain to retain desired sensory, chemical, physical, microbiological and functional characteristics. And comply with any label declaration of nutritional data when stored under the commended conditions"(Robertson, 2009, Chapter 1).

The shelf life of a product does not necessarily correspond to its real "life" since the loss of some characteristics (in particular, sensory ones) can be equivalent to the end of its marketability. And yet, it may not necessarily refer to the loss of the fundamental product characteristics, the hygienic-sanitary ones or nutritional efficacy. Therefore, shelf life is commonly defined as the time interval between the packaging of a product and when it becomes unacceptable under established environmental conditions. The complexity of defining the shelf life derives from the assortment of different possible situations. The same product can be packaged and stored in many ways.

Food engineering academy has grouped the different variables that can influence the shelf life of a food product together in three main factors: the product characteristics, including formulation and processing parameters (intrinsic factors), the environment to which the product is exposed to during distribution and storage (extrinsic factors), and the properties of the package (Robertson, 2012, Chapter 12; Robertson, 2009, Chapter 1).

Food variables

Variables related to the food characteristics which have a vital role in influencing its commercial durability are, for example, the microbial load, the enzyme kit, the PH and water activity values, the concentration of specific solutes, the presence of inhibitors and preservatives, of promoters and catalysts. Undoubtedly these factors, often defined as "intrinsic", are the most relevant in establishing the food storage possibilities (packaged or not).

[12] IFST. The Institute of Food Science and Technology. independent qualifying body for food professionals based in the UK and is concerned with all aspects of food science and technology. https://www.ifst.org/

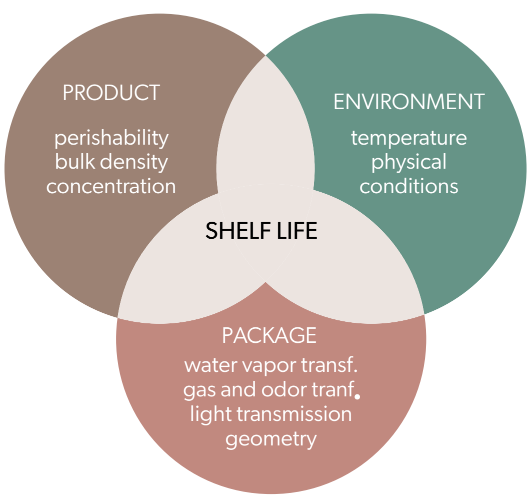
Environment variables
The effects of light, temperature, humidity and the concentration of oxygen in the environment are the most important variables external to the product can modify the expected times of food products shelf life, but never to the point of transforming a very stable food into a very perishable one. Certainly, the environmental variable factors can influence the critical nature of the food. For example, at lower temperatures the factor microbial charge, may be less critical and other factors become decisive. A change in environmental conditions, therefore, can make a variable of food negligible in different circumstances.

Packaging variables
Variable factors that directly affect the packaging can be identified in the gas and vapor barrier offered by the packaging, its transparency to light, its ability to resist mechanical and thermal stresses and in its inertia in contact with food. In other words, the packaging variables modulate the environmental variables, creating a micro-environment different from the external one (the macro-environment).

The package properties can have a significant effect on many of the extrinsic factors and, indirectly on the deteriorative reactions. Thus, the shelf life of a food can be altered by changing its composition and formulation, processing parameters, packaging system or environment to which it is exposed.

As it was previously described, the three categories cannot be considered independently because they interact by influencing each other. The food and beverage deterioration methods and storage times are consequently different and highly variable in between cases. Figure 12 describes the most important variables that determine each factor.

Figure 12

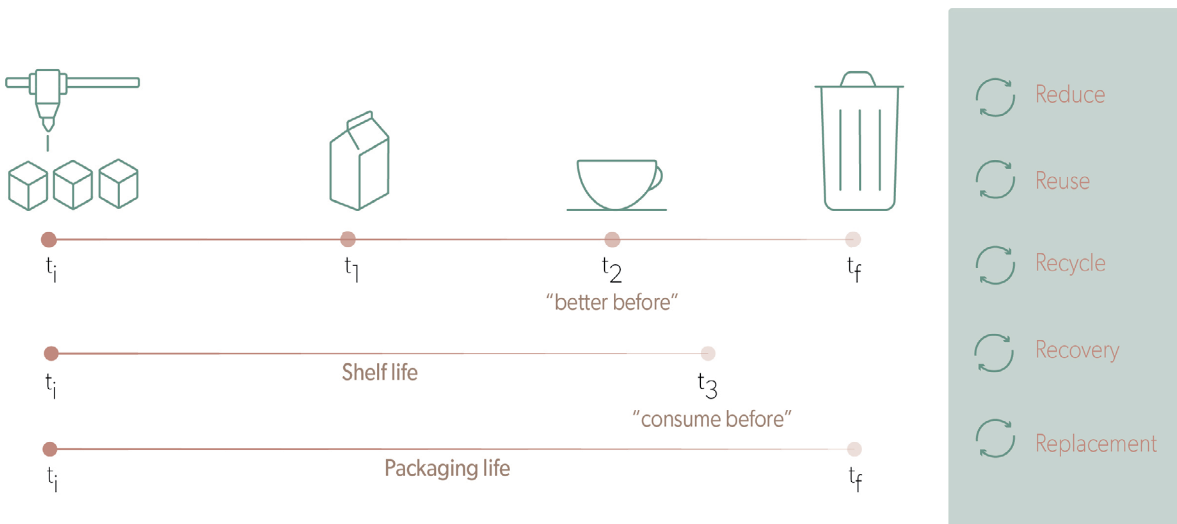


Determining Factors of Food Product Shelf Life
Note. Figure 12 shows a visual representation of three principal factors that determine the shelf life of a food product. Concept explained in common between authors.
Source. (Robertson, 2012, Chapter 12; Robertson, 2009, Chapter 1)

The barrier properties required to preserving the food content in its original state, and consequently, the type of material suitable for the packaging is based on the food type and environmental factors. Similarly, the material selected for the packaging will determine the micro-environment conditions, highly influencing the shelf life, the packaging lifetime, and the way it can be disposed after its service time.

From a sustainable approach, the lifetime of a food packaged product should consider the shelf life of the food product and the end of life of the packaging associated. Figure 13 shows five sustainable alternatives a food product can take to close the loop of its life cycle.

Figure 13



Sustainable Food Product Life Cycle
Source. (Robertson, 2012, Chapter 12; Robertson, 2009, Chapter 1)

1.3.8 Eco-design

Sustainability must be considered as a core package design concept. It should be included during the earliest packaging development stage of a product to minimize environmental impacts, maximize cost saving, and avoid excessive or deceptive packaging. In contrast, environmental considerations must be aligned to the manufacturer's overall development and management strategies (Han, 2013, Chapter 22).

To develop sustainable food packaging solutions, it is fundamental to begin from the design stage. Which must also change from the traditional paradigm to an eco-design one.

According to the European Union's Eco-design Directive (2009/125/EC)[13], eco-design means "the integration of the environmental aspects into product design to improve the environmental

[13] Directive 2009/125/EC of the European Parliament and of the Council of 21 October 2009 establishing a framework for the setting of eco-design requirements for energy-related products.
https://eur-lex.europa.eu/legal-content/EN/ALL/?uri=CELEX%3A32009L0125

aspects into a product throughout its whole life cycle". The "environmental aspects" represent an element or function of a product that can interact with the environment during its life cycle (European Commission, n.d). To make sustainable food products available in the various markets, the development of properly designed packaging is essential to reduce both costs and environmental impacts throughout the packaging's entire life cycle, while maintaining the food quality and safety.

The eco-design of a food packaging not only means the most suitable material selection. The design must consider a complete and holistic view from its initial industrial process to its final disposal. Table 6 lists attributes that should be addressed when designing sustainable food packaging.

Table 6. *Attributes involved in designing a sustainable food packaging*

Demand area	Time and place	Attributes
FUNCTION	Conception	Containment, protection, communication, convenience
ENVIRONMENT	Food production	Emissions, effluents, waste, energy consumption
	Packaging	Resources, package size, weight, reuse, recycle, disposal
	Distribution and marketing	Temperature, shelf life, transportation
	Consumer	Storage temperature, preparation, food waste
SOCIETY	Food production	Preference for local food
	Packaging	Hygienic safety
	Distribution and marketing	Fair-trade, government policy, shelf life control
	Consumer	Attitude on accepting recycled products, willingness to sacrifice convenience for environmentally friendly packaging
ECONOMICS	All phases	Cost effectiveness, financial resources

Source. (Yam & Sun Lee, 2012, Chapter 18, p. 366)

A holistic eco-design approach is based on the sustainability principle of the equilibrium between social, environmental, and economic demands. Concerning environmental demands, natural resources can be dealt in packaging manufacture and usage. Social demands, welfare and safety of human communities should be taken in the application process of food packaging. From an economic standpoint, sustainable packaging must be viable and must allow fair profits. Harmonizing these demands is essential for the successful eco-design of food packaging.

The eco-design of food packaging considers the prevention of food loss and waste as the primary factor and should be balanced with packaging itself adverse environmental impact.

Sustainable packaging could avoid or reduce the environmental damage caused by humans and can be achieved by using the four principles of effectiveness, efficiency, cycle, and safety, as defined by the Sustainable Packaging Alliance (SPA)14. Table 7 summarizes the major strategies for the eco-design of food packaging.

Table 7. *Packaging strategies for eco-designing of food packaging*

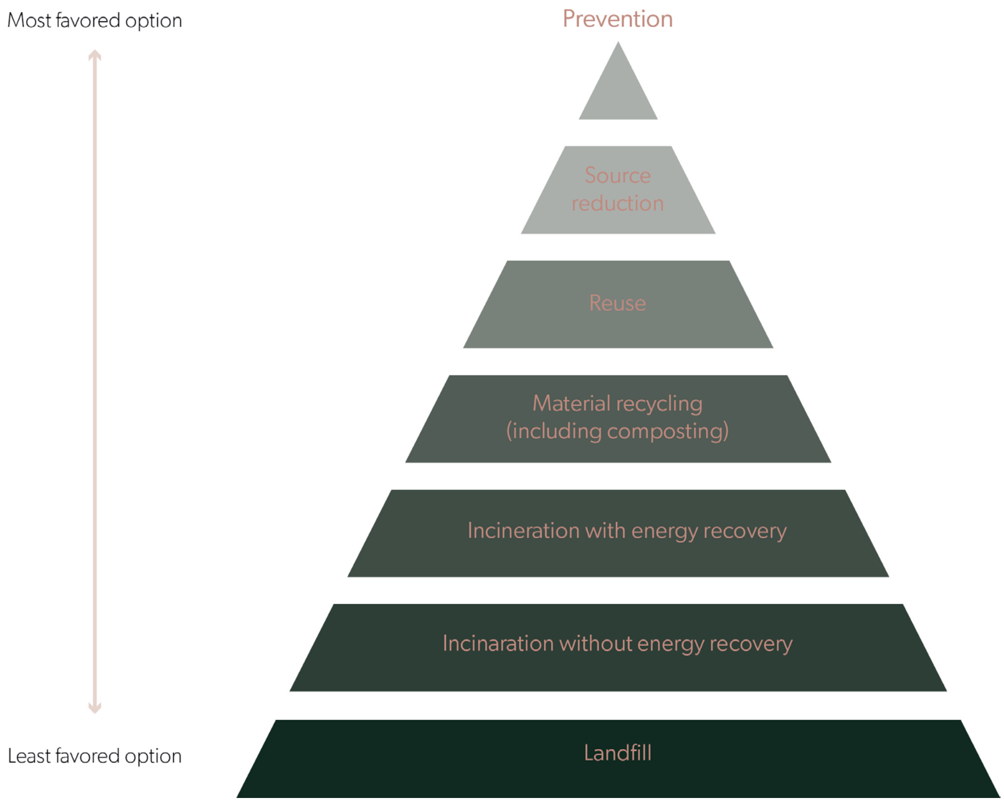
Principle	Strategies
EFFECTIVE	• Examine which packaging can best activate the function of containment, protection, communication, and convenience. • Minimize the total number of packaging layers or components through combined optimization of primary, secondary, and transportation packaging. • Design packaging system by reviewing information on the environment impact from whole life cycle analysis. • Minimize total cost in product supply chain. • Provide to consumers the information and advice on impact and disposal of the packaging.
EFFICIENT	• Minimize packaging volume (including void space), weight, and thickness in the extent not to sacrifice the product safety and packaging. • Find ways to improve transportation efficiency by using concentrated product, bulk packaging, and maximum space fitting. • Find ways to minimize the food waste and maximize the efficiency of energy and material used in the whole system. • Design the food packaging system in balanced harmony with shelf life, consumption behavior.
CYCLIC	• Check the available ways to collect and return the empty packages for reuse and recycling. • Use reusable packages as much as possible • Use single recyclable material for all packages components whenever possible. • Use symbols for recyclability. • Specify the identification of compostable and renewable materials where they are used. • Eliminate chances for recyclable plastics and compostable polymers to be mixed together in the recycle program.
SAFE	• Avoid toxic materials such as heavy metals and halogen compounds in manufacture of any package components. • Avoid, in package manufacture, the use of materials or additives that can migrate to food from contact packaging material.

Sources. (Selke,1990; Lewis et al., 2007; Maxwell & Van der Vorst, 2003; Verghese, 2008; Jedlicka, 2009, as cited in Yam & Sun Lee, 2012)

[14] Sustainable Packaging Alliance (SPA). The Sustainable Packaging Coalition is a trademark project of GreenBlue Org. GreenBlue is a 501(c)(3) nonprofit dedicated to the sustainable use of materials in society. https://sustainablepackaging.org/about-us/

Eco-design methodologies could be established on the 5R principle: reduce, reuse, recycle, recovery, and replacement using renewable or degradable resources. Which is based on the hierarchy of solid waste management (Figure 14).

Figure 14



Hierarchy of Solid Waste Management
Note. Figure 14 shows the waste management hierarchy, which has become accepted as dogma in many countries and among some policymakers, politicians, and environmentalists.
Source. (Andrady, 2015; Han, 2013; Robertson, 2012).

Production of eco-designed food packaging uses fewer resources and subsequently produces less waste and emissions. Prevention and source reduction are accepted as the most favored choices in the solid waste management hierarchy. Source reduction could be achieved by altering packaging design or manufacturing processes to reduce the number of materials used.

There are some debates about reduction versus recycling until now, because the best choice will depend on the packaging material type and the geographical circumstances. The recycling rate of packaging materials depends on the availability of a regional recycling management infrastructure and processing capacity (Han, 2013).

Recovery of packaging wastes can be summarized in three different treatment streams: organic, chemical recovery, and energy recovery. About the fifth "R", referred to the replacement of raw materials, it is the newest technology in terms of sustainable or green materials, and can be associated with biodegradability or recycling waste management.

Several measures to evaluate the environmental impacts of packaging have been proposed, including global warming, energy consumption, ozone depletion, land use, eutrophy, airborne emissions, water-borne emission, solid waste production, among others (Yam & Sun Lee, 2012).

Methodologies and assessments as LCA (life cycle assessment), MIPS (material input per system), CED (cumulative energy demand), MET (material use, energy use, and toxicity matrix) available in the market, aid to appraise the product development based on eco-design principles. However, the most widely used and comprehensive approach is LCA.

1.3.9 Emerging trends

Sustainability in food packaging is already a strong trend in the food industry fundamentally based on the environmental problems associated with food loss and packaging waste. Concern claimed by the consumer awareness.

Significant efforts drive the evolution of other innovative packaging technologies. Which can maintain and monitor food safety and quality, extend shelf-life, and reduce the environmental impact of packaged foods. Active, intelligent, and green packaging technologies can work synergistically to yield a multipurpose food-packaging system with no adverse interactions between components, and this aim can be seen as the ultimate future goal for food packaging technology. Therefore, following trends presented in this section are based in the new advances in technology associated to food packaging solutions (Han et al., 2018; Sharma & Ghoshal, 2018).

Intelligent packaging

Intelligent packaging refers to monitoring the internal and external environment of the package to predict the quality and remaining shelf life of the product. It provides valid and accurate information to the consumers about the safety, quality, and integrity. These packages detect unsafe foods and identify conditions that adversely affect the quality of the food.

Intelligent packaging technology comprises indicators, radio frequency identification (RFID), biosensors and barcodes. Indicators can be qualitative or semi-quantitative devices that provide immediate visual information through a change in color or deviation in color intensity. These indicators can be external, attached to the package's exterior, such as time-temperature indicators (TTIs). Also, internal indicators, present inside the package, such as the pathogen indicators and gas leak indicators. The third category of indicators are those which increase the information flow between package and consumer (e.g. barcodes).

RFID systems are used to capture, write, store, and read the data of the object to which they are attached through radio waves. Sensor-based RFID tags are more popular in food industries, as it is vital to maintain the quality of produce from farm to plate. Storage conditions such as temperature, humidity, gas composition, light exposure, pH, and pressure have considerable effect on the safety and quality of the food products. Thus, monitoring these conditions by incorporating indicators or sensors with RFID tags can detect the temperature profile, leakage, and pathogen.

Biosensors are compact analytical tools that can detect, record and/or transmit data or information about biochemical reaction. Generally, biosensors comprise two main components: bioreceptor and transducer. Bioreceptors are mainly biological materials such as enzymes, metabolites, hormones, antibodies or nucleic acids, whose main function is to recognize the target analyzed. In comparison, transducers perform the function of conversion of biochemical signals into an electrical response.

Active packaging

Active packaging refers to incorporating some active additives in the package to meet the consumer's expectations and to satisfy them. In contrast, the traditional package are considered passive packages. The dynamic approach of active packaging is based on the interaction of its components. The interaction between food, package, active additives, and environment prolong the timeframe of realistic usability and upgrades the product's safety. Furthermore, this active interaction keeps up the quality and organoleptic characteristics of the food product.

The different frameworks used as a part of the active packaging change the environment inside the package by addition or elimination of gases in headspace and may likewise interact with the product surface. There are two types of active components for food packaging solutions, emitters, and scavengers. Among them, are oxygen scavengers, carbon dioxide emitters/scavengers, antimicrobials, and ethylene absorbers.

Oxygen scavengers
The oxygen inside the package even at low levels triggers the deteriorative responses and limits the time span. The different reactions that occur within the sight of oxygen are generated by a lipid oxidation known as rancidity. Rancidity deteriorates the flavor, accelerate the growth of aerobic microorganisms and molds. Also trigger the oxidation of pigments present in food, which cause undesirable changes in the color. Furthermore, these reactions can degrade nutrients, for instance, the degradation of vitamin C. The oxygen scavenging compounds react with the package's oxygen present to reduce its concentration and make it unavailable for deteriorative reactions.

Carbon dioxide emitters/scavengers
Generating carbon dioxide in the packages is a complementary technique to oxygen scavengers as carbon dioxide is known to suppress microbial growth. In fact, the application of carbon dioxide to suppress the microbiological population is found in certain products such as meat, bakery products, cheese, and poultry. Therefore, Carbon dioxide emitters are comparatively potential candidates to be used in active packaging solutions.

Antimicrobials
Antimicrobials either inactivate the microbes or extend the lag phase in the growth cycle, reducing their growth rate. Antimicrobial can be chemical or natural. Class of chemical antimicrobials include organic acids, ethanol, metals and gaseous antimicrobials. Bacteriocins and natural plant extracts come under the category of natural antimicrobials.

Ethylene absorbers

Ethylene is a gas produced by fruits and vegetables during ripening, which acts as plant hormone. Rate of respiration is a critical factor correlated with perishability of fresh produce. And ethylene accelerates the rate of respiration, leading to aging and senescence of fruits and vegetables making it important to eliminate ethylene from packages. Furthermore, the accumulation of ethylene inside the package results in several other deteriorative reactions such as yellowing of green vegetables, bringing about loss of chlorophyll and bitter flavor.

Smart packaging

Smart packaging refers to the integration of active and intelligent functions in one system. These systems can detect and control the required parameters affecting the quality profile of the product. The various techniques being explored these days under the smart package are self-cleaning, self-heating/cooling and self-healing packages. Smart packaging also enables a better packaging communication function through smart labeling that allows customer understanding of the correct use and disposal process.

When modern packaging is focusing on delaying microbial and biochemical deterioration, a strong parallel emphasis is done on sustainable food packaging. The application of these innovations using bioactivity of functional components is expanding widely because of potential benefits to consumers and environment. These novel techniques can enable a sustainable treatment of food products, due to its promising facilities to maintain safety and food quality. This could also address food waste and packaging waste management through its communication facilities between product to consumer. Through dynamic labels consumers can understand when the product has arrived to the final of its shelf life or if the packaging material can be composted or eaten.

However, additional effort should be focused on overcoming the technical constraints and high costs associated with these technologies, which have been the main factors preventing wider commercial implementation. Finally, to increase the safety and effectiveness of new food packaging technologies and ensure its sustainability, continuous research and development should be performed based on collaboration between government regulatory agencies, industries, consumers, and multidisciplinary experts.

1.4 Sustainable plastics for food packaging

Plastics are not harmful because they are used as "plastics", plastics are highly useful and currently indispensable for food packaging applications. But the way in which it is generally made, and then disposed are not favorable for our planet, ecosystems, and human life. Clearly, the development of suitable plastics for food products need a sustainable approach since its design stage.

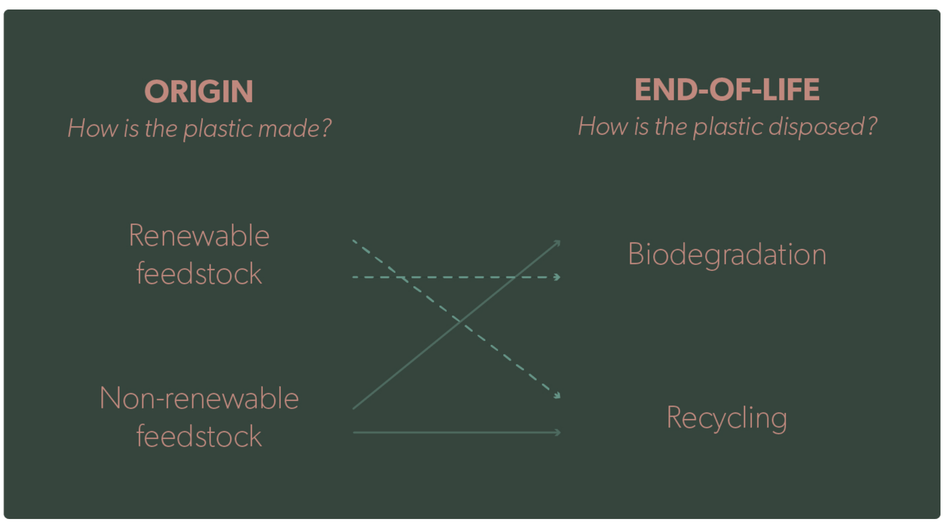
Change to sustainable plastics is not an easy scope from any angle. As all new technologies, many aspects need to be studied and proved throughout the product life cycle to achieve a successful performance. Fortunately, there are already many large companies, innovating, investing and

currently commercializing novel sustainable plastics solutions.
The development of a material is considered sustainable, in the way it decreases the environmental, social, and economic negative impacts during its whole life cycle. Therefore, a plastic meets the sustainable grounds through its production and final dispose strategies closes its life cycle, converting it in a circular cycle. Sustainable plastic in food packaging can be based in one or more of the following general objectives:

- Responsible utilization of renewable resources
- Avoid the utilization of non-renewable resources
- Reduce of GHG emissions
- Optimization of resources for manufacturing processes
- Minimization of food loss and packaging waste
- Apply: recyclability, biodegradability, and/or composability as final disposal treatment (Only in specific cases, incineration with energy recovery)

These general objectives are ground of the current sustainable alternatives presents in the market and in continue investigation. Already many variants can be found as sustainable plastics suitable for food packaging applications, which means they are safe and fit food packaging functions (section 1.3.1). Figure 15 summarizes the main branches plastics can take under a sustainable approach, showing the current alternatives for food packaging applications.

Figure 15



Sustainable routes for plastics in food packaging
Note. Due to the sustainable approach taken in the present study, other end-of-life treatments are not taken into account.

Sustainable plastics in food packaging can be addressed since the *"how is made"* phase, it means which type of feedstock employ in its production. It can also be defined from its end-of-life treatment, in other words, *"how is the plastic disposed".* Therefore, as it is represented in figure 15, each arrow represents a combination categorized as sustainable solution.

Considering the state of the art of these options and its intrinsic classifications, this study will initially organize them in two branches based on the origin of the raw material and its sustainable end-of-life possibility, as follow presented:

1. Virgin plastics: Considering raw materials used for the first time to produce plastics packages for food products. In this group will be included those bioplastics which can biodegrade. Although some of them could be recycled, they are considered in this group taken into account the biodegradation property as priority sustainable end-of-life option. Due to biodegrading property is not uniquely related to the origin of the feedstock, in this group will be considered plastics coming from renewable and non-renewable resources. Virgin plastics will be further developed in Chapter II.

2. Recyclable plastics: In this second group will be considered the bioplastics which cannot biodegrade due to its chemical characteristics, but they can be properly recyclable. Therefore, this group will take into account those plastics made from renewable origin and those ones which come from petrochemical resources. Recyclable plastics group will be further developed in the chapter III.

Furthermore, the bioplastics which are used as coatings in food packages will be presented and developed in the chapter IV. In each chapter, will be further discussed the sustainable solutions already available in the food packaging market, its research progress, and promising applications in the future.

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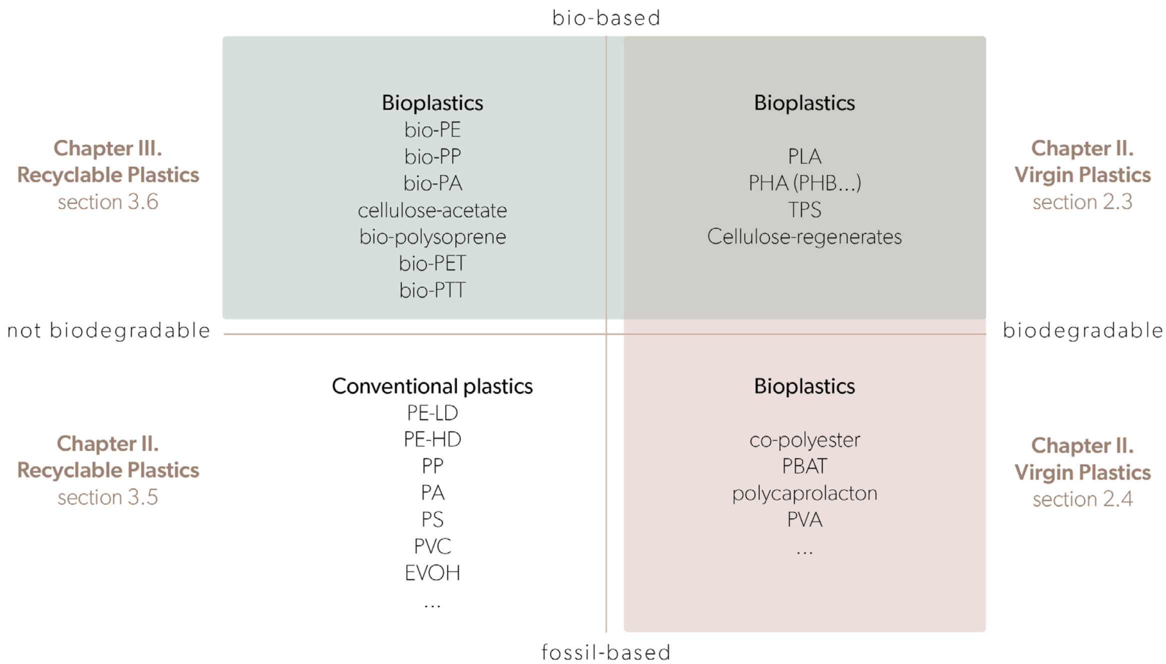
Chapter II.

VIRGIN PLASTICS

2.1 Introduction and classification

This standpoint of classification, as seen in the first chapter (section 1.4) contemplates two main groups of sustainable alternatives for food packaging applications: *Virgin plastics* and *Recyclable plastics*. Considering raw materials used for the first time to produce plastics packages for food products, *Virgin plastics* include those bioplastics which can biodegrade. Even though, some of them could be recycled, they are considered in this group taken into account the biodegradation property as priority sustainable end-of-life option. In this group will be considered plastics coming from renewable and non-renewable resources, due to biodegrading property is not uniquely related to the origin of the feedstock. Figure 16 shows a simple two-axis model that categorize bioplastics considering its origin and its sustainable end-of-life option.

Figure 16



Bioplastic's classification by origin and biodegradability
Source. (European Bioplastics, n.d)

1. Plastics that are not biodegradable and are made from petrochemical resources: This category encompasses what is known as classical, conventional, or traditional plastics. Undoubtedly, for the reasons explained before, this group is not considered in the present chapter but due to its possibility to be recycled, will be contemplated in the chapter III (section 3.5).

2. Biodegradable bioplastics from renewable resources: Bioplastics made from biomass feedstock material and show the property of biodegradation. This set of solutions are considered in this chapter as part of virgin plastics (section 2.3).

3. Biodegradable bioplastics from fossil resources: Biolastics that can biodegrade but are

produced from fossil resources. This group of bioplastics used virgin feedstock in their production and are also a sustainable solution in polymers market for food packaging, therefore, will be considered in the virgin plastics division (section 2.4).

4. Non-biodegradable bioplastics from renewable resources: Bioplastics produced from biomass but without the biodegradation property. This special group is considered as part of recycled plastics behind a sustainable perspective and therefore will be further detailed in chapter III.

In theory, all resources are renewable at a particular moment of their lifetime, the difference lies when in time this occurs. From a sustainable perspective, renewable resources are considered those ones that can renew in one year, which is possible because their quantity is not decreasing due to human use, but it is quickly restored through natural processes. These include wind, solar, geothermal, wave and tidal energy, and biomass. Evidently, fossil-fuels exceed this period because they entail millions of years to be made again by nature. Therefore, from a sustainable perspective, fossil fuels, oil, and natural gas are considerate non-renewable resources (Rujnić-Sokele & Pilipović, 2017).

Nevertheless, there are sustainable routes for polymers derived from petrochemical resources. Biodegradation and recycling are successful alternatives that can provide a circular life for these materials, and this is the aim reason to why it can also be considered behind a sustainable approach for specific cases as it will be discussed later.

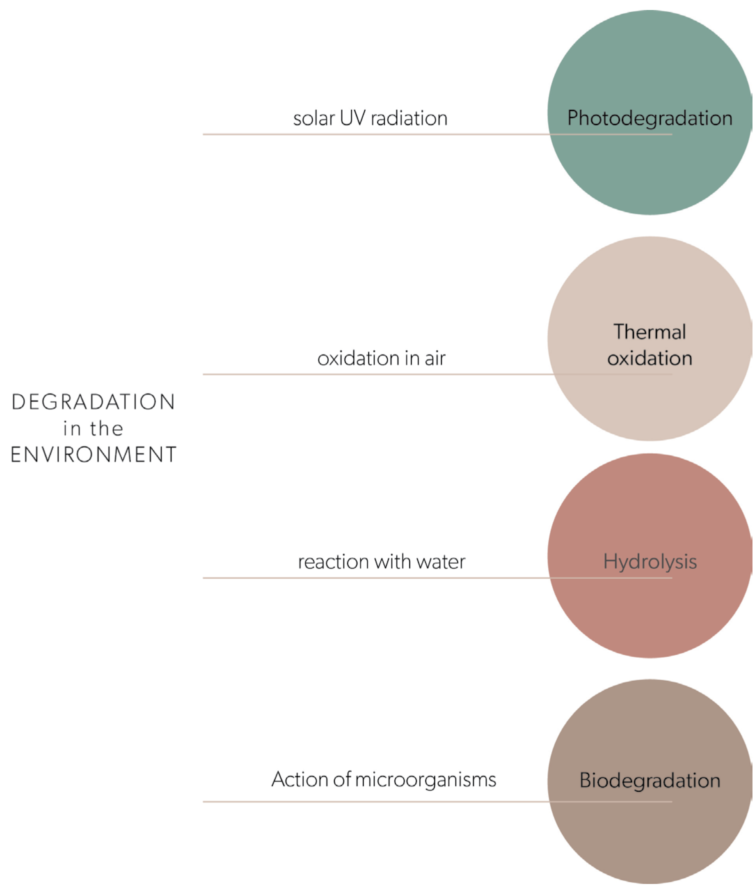
On the other hand, the term bioplastic was introduced by the European Bioplastics and includes biodegradable plastics, biobased plastics, or both (Rujnić-Sokele & Pilipović, 2017). Nevertheless, it is important to highlight that referring to biobased plastics and bioplastics is not exactly the same. As explained before, bioplastics can be bio-based or fossil-based, while bio-based plastics are always defined as bioplastics. Regarding ASTM D7075-04 standard, bio-based materials are defined as materials containing carbon-based compounds(s) in which the carbon comes from contemporary (non-fossil) biological sources.

Before entering to each Virgin plastics sub-group will be discussed the degradability property of materials and its implications in food packaging applications made of plastics.

2.2 Degradability

Degradation is the chemical change that alters the properties of a material due to useful properties such as high strength or high stiffness which are affected in this process. Degradation processes can be categorized according to the principal organisms that bring out this chemical change (Andrady, 2015). Figure 17 explains the different degradation mechanisms.

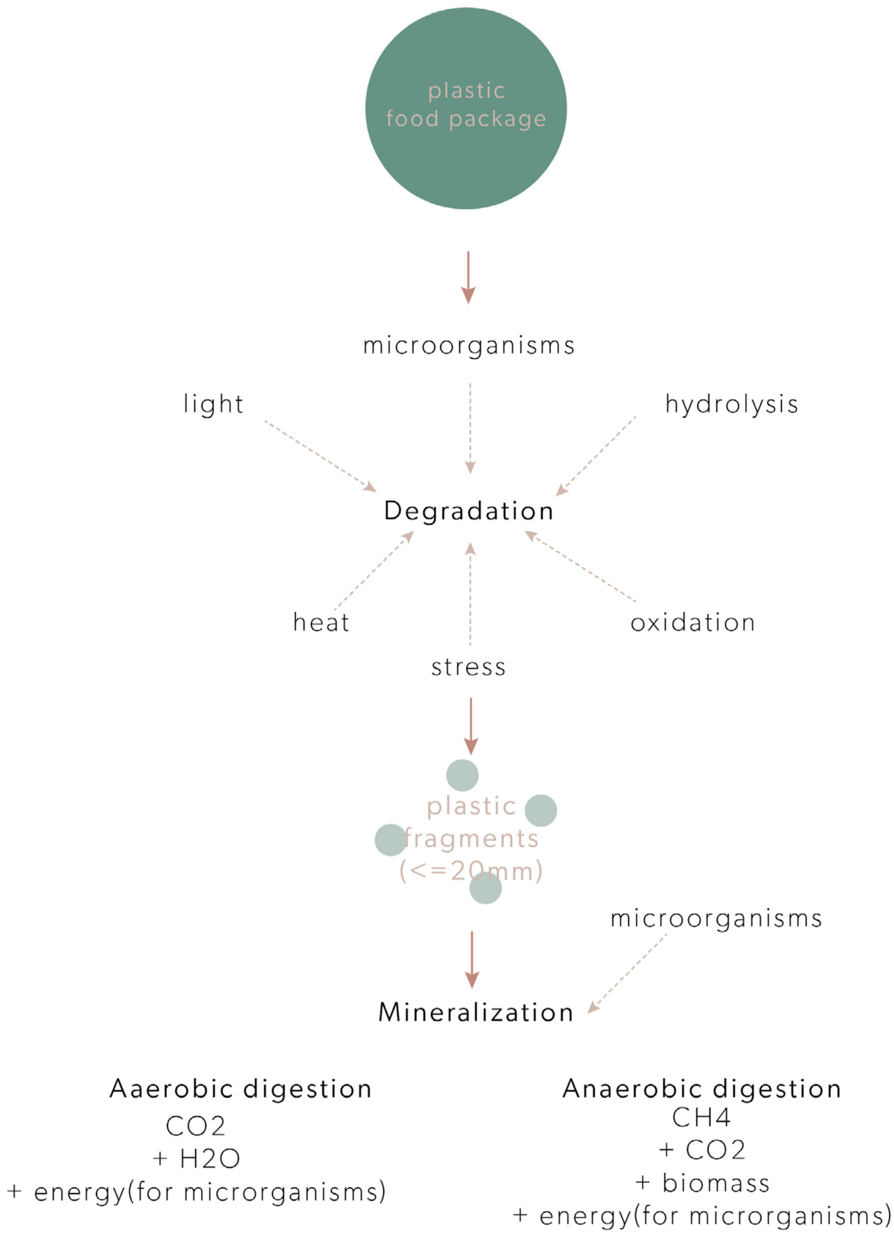
Figure 17



Principal agents of plastics degradation in the environment
Note. Figure 17 is an adaptation
Source. (Andrady, 2015).

Photo-oxidation is the most common process of degradation for plastics exposed outdoors or when they finish in landfills. At the same time, hydrolysis is a degradation mechanism available only to a few selected plastics. Common food packaging plastics such as PE, PP, PS and PVC do not hydrolyze appreciably under environmental conditions.
In a favorable situation, under solar irradiation, a food package made from plastic will lose strength and their mechanical properties until it finally will crack into small fragments. If they are exposed to a biotic medium, they will undergo biologically mediated degradation, converting the material into inorganic molecules such as CH_4, NH_3, and CO_2. As can be seen in figure 18, this last step where the plastic is converted into inorganic molecules is known as "mineralization".

Figure 18



Degradation process

Source. (Andrady, 2015).

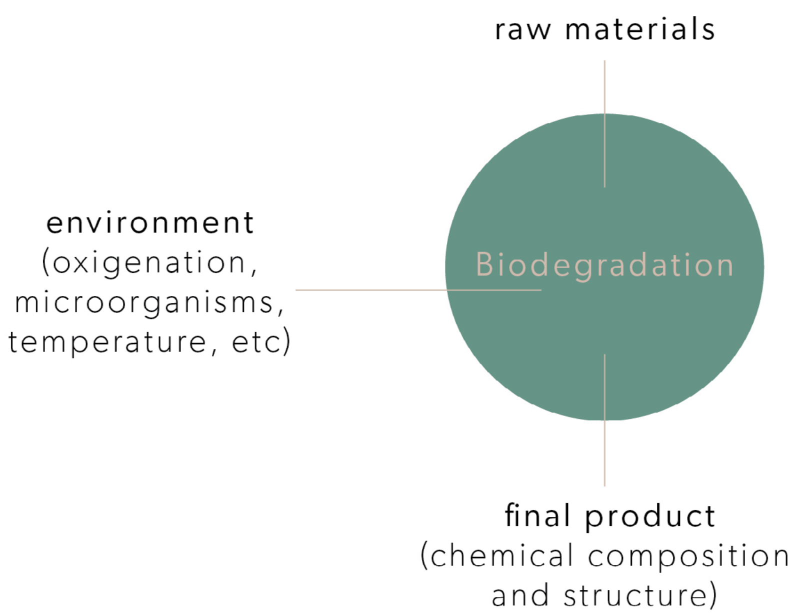
The problem of food packaging made with traditional plastics is that they can take decades or centuries to completely degrade. In theory all plastics invariably biodegrade, nevertheless , biodegradation term is commonly used to indicate that a biodegradable plastic degrades or breaks down at a much faster, and measurable rate (Andrady, 2015).

2.2.1 Biodegradability

A proper definition of a biodegradable bioplastic recognizes biodegradability as a material property where the bioplastic can degrade by the action of naturally occurring microorganisms such as prokaryotic (bacteria), eukaryotic (fungi and protozoa), and algae (Attaran et al., 2017) capable of excreting enzymes (depolymerases) that degrade the polymeric matrix (Rujnić-Sokele & Pilipović, 2017).

As figure 19 shows, the biodegradability of plastics is subject to the raw materials and the chemical composition and structure of the final product, and on the environment under which the product is expected to biodegrade (Rujnić-Sokele & Pilipović, 2017). Therefore, testing organizations to evaluate the degradation ability of specific polymers depending on the type of environment, exposure variables (availability of oxygen, temperature, humidity, etc), within the most important influencing factors. For instance, the ASTM D5338-931 as well as European standard EN 1343[2] (section2.2.3) requires a material to be at least 90% biodegraded in less than 6 months (Andrady, 2015).

Figure 19



Factors influencing the biodegradation process

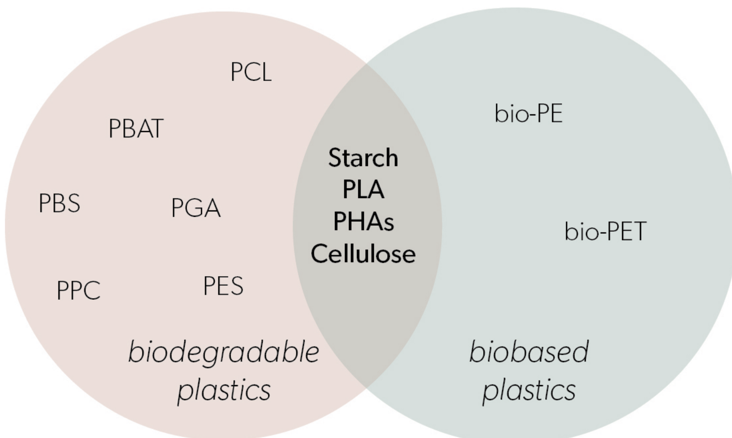
Source.(Andrady, 2015)

On the other hand, there is still a common and incorrect belief that a material derived from biomass is also biodegradable. However, the use of biofeedstocks does not necessarily mean that the finished product can biodegrade. Some biobased plastics are not always biodegradable and biodegradable plastics are not always biobased. Additionally, some polymers degrade in only a few weeks, while others take several months to degrade under the same environment (Rujnić-Sokele & Pilipović, 2017).

[1] ASTM D5338 – Standard Test Method for Determining Aerobic Biodegradation of Plastic Materials Under Controlled Composting Conditions, Incorporating Thermophilic Temperatures

Figure 20 illustrates the biodegradability differentiation property in plastics used for food packaging applications. Where can highlighted bioplastics such as PLA, PHAs, and bioplastics derived from cellulose and starch. These bioplastics represent the best sustainable alternative for food packages made of plastics due to it biodegradability property.

Figure 20



Classification of plastics for Food packaging applications
Note. Author representation based on the source
Source.(Andrady, 2015, pp.168)

Processing biobased and biodegradable bioplastics

Biobased biodegradable plastics have different aspects owing to their renewable origin that must be considered. These aspects include moisture, flow anomalies (wall slipping), thermal degradation and batch-to-batch variations. Biobased biodegradable plastics tend to be hygroscopic, so moisture can cause various problems, for example uncontrolled reduction of viscosity, undesired foaming and acceleration of thermal degradation or hydrolysis (Rujnić-Sokele & Pilipović, 2017).

Biobased biodegradable plastics are prone to thermal degradation, so special precautions must be made in processing. One of the problems during processing include the formation of adhesive pellets when drying, in which case an additional crystallization step may be needed. Because of their natural origin, biobased biodegradable plastics possess higher variability in processing. Nevertheless, nowadays, the industrial sector has obtained formulas which can be used in conventional plastic manufacturing processes. Injection moulding, cast and blown film extrusion, blow moulding, thermomoulding are principal examples (Rujnić-Sokele & Pilipović, 2017).

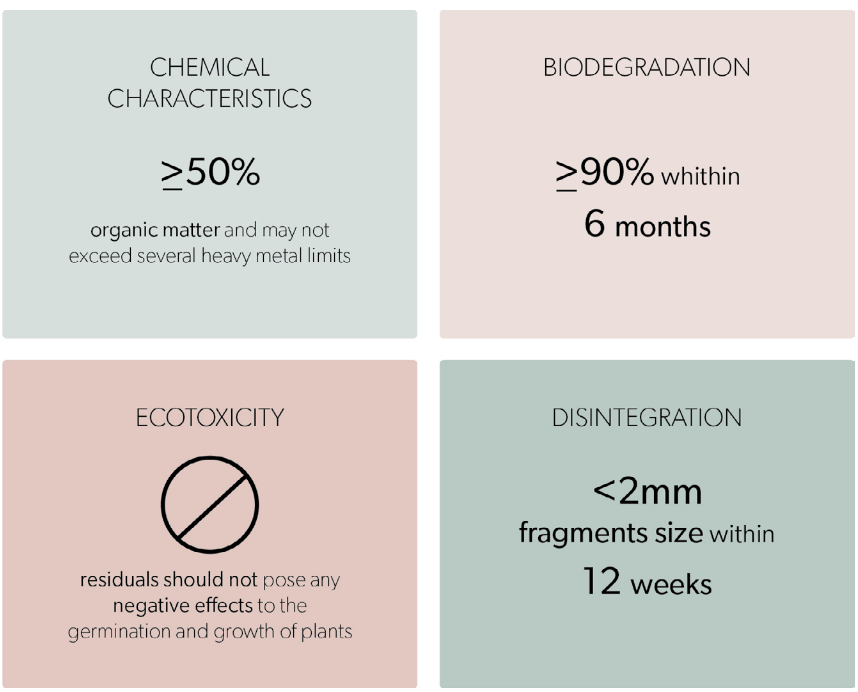
2.2.2 Compostability

Another term often referred to in bioplastic when determining its ability to degrade is compostability. Composting is the accelerated degradation of heterogeneous organic matter by a mixed microbial population in a moist, aerobic environment under controlled conditions. Aerobic waste management systems, such as composting facilities, generate carbon and nutrient-rich compost which makes it more beneficial when adding to soil (Rujnić-Sokele & Pilipović, 2017).

Compostable plastics are degradable owing to a biological process occurring during composting and are converted into carbon dioxide, water, mineral salts, and biomass. There are no toxic side effects, like toxic residue for water, soil, plants or living organisms. Products fully complying with the requirements of these standards are capable of undergoing a complete biological decomposition solely owing to the action of naturally occurring microorganisms under industrial composting conditions. It should be noted that not all biodegradable materials meet composting criteria. Materials that do not fulfil these criterias may still be biodegradable under specific environmental conditions (Plastics Europe, 2017; Rujnić-Sokele & Pilipović, 2017).

Figure 21 shows four criteria must be fulfilled for a plastic to be categorized as compostable. These criteria must be measurable under controlled composting conditions:

Figure 21



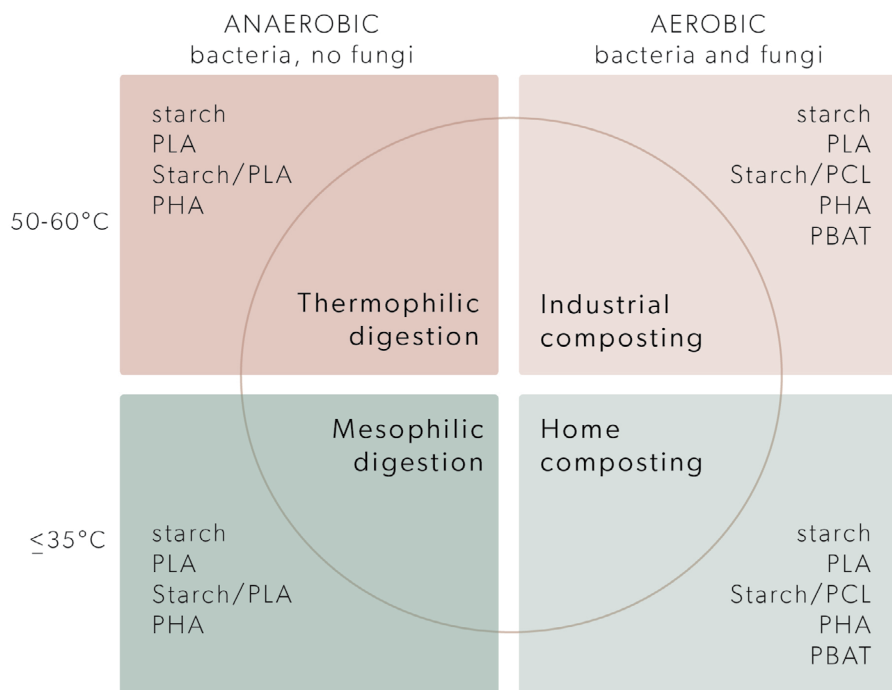
Classification Criteria for compostable plastics

Source. (Rujnić-Sokele & Pilipović, 2017)

Certification ensures that the product can be industrially composted and that not only the plastic but also all other components of the product are compostable, for example colors, labels, glues and – in case of packaging products – residues of the content (European Bioplastics, 2016; Rujnić-Sokele & Pilipović, 2017).

The most aggressive environment is compost, followed by soil, fresh water, marine water and landfill. There are two reasons for that, the first one is temperature, and the other is the presence of microorganisms that are fungi and bacteria. Figure 22 shows the types of biodegradation processes that can be considered for food packages. For instance, in industrial composting facilities the temperature is high (60 °C), which is important for certain biodegradable plastics.

Figure 22



Classification of biodegradation processes
Source. (Rujnić-Sokele & Pilipović, 2017)

The most favorable final disposition, from an environmental point of view, for biodegradable plastics is represented by the composting process, taking into account that the process conditions in terms of humidity, oxygen, temperature, presence of specific microorganisms, etc., must be strictly controlled to achieve noticeable results in terms of final products. However, within composting options, the most sustainable favorable is industrial composting, due to it brings out final enriched product for arable lands. Plastics suitable for composting should be collected through a separate collection scheme and brought to an industrial composting facility, neither of which is still present in many countries. Separating biodegradable and compostable plastics from conventional plastics using near infrared detection technology is possible but costly to put into operation.

On the other hand, some biodegradable plastics are suitable for anaerobic digesters whereby biowastes can be converted to methane, which can be used to drive generators for energy production (Mudgal et al., 2012; Song et al., 2009 as cited in Rujnić-Sokele & Pilipović, 2017).

2.2.3 Standards and labels

Due to not having harmonizing standards in the EU legislation specifically for environmental claims, the European Commission as well as national governments, ministries, and independent standardization institutes have issued a multitude of standards that can serve as a basis for evaluating claims for bioplastics and other biobased products.
The key standardization bodies creating standards are ISO, CEN[2] and ASTM. An accepted standard will be used for the certification of certain properties and the according label or logo will be awarded for easy identification. Below are details upon the most relevant standards and labels for biodegradable plastics (Rujnić-Sokele & Pilipović, 2017; European Bioplastics, 2019).

Standards for industrial composting and anaerobic digestion

- EN 13432 "Requirements for packaging recoverable through composting and biodegradation" requires at least 90% disintegration after twelve weeks, 90% biodegradation[5] (CO_2 evolvement) in six months, and includes tests on ecotoxicity and heavy metal content. It is the standard for biodegradable packaging designed for treatment in industrial composting facilities and anaerobic digestion.
- EN 14995 describes the same requirements and tests as EN 13432, while applying not only to packaging but plastics in general.

  Counterparts
  - ISO 18606 "Packaging and the environment – Organic Recycling" (worldwide)
  - ISO 17088 "Specifications for compostable plastics" (worldwide)
  - ASTM D6400 "Specification for Labelling of Plastics Designed to be Aerobically Composted in Municipal or Industrial Facilities" is the US standard with clear pass/fail criteria. The corresponding label is the BPI Compostable in Industrial Facilities.
  - AS 4736 "Biodegradable Plastics suitable for Composting and other microbial Treatment", additionally includes the so-called earthworm test. The Seedling Australia logo is certified according to this standard.

Labels for industrially compostable products are, for example, the Seedling logo, OK Compost, and DIN[3]-Gerprüft Industrial Compostable, as well as the compostable logo of Consorzio Italiano Compostatori (CIC)[4] .

[2] CEN. European Committee for Standardization. is an association that brings together the National Standardization Bodies of 34 European countries. https://www.cen.eu/Pages/default.aspx

[3] DIN. Deutsches Institut für Normung e.V. (German Institute for Standardization). https://www.din.de/en

[4] The logo of CIC is awarded foremost on national level in Italy.

Figure 23

Seedling logo

OK Compost logo

DIN-Gerprüft Industrial Compostable

Principal labels for products industrially compostable
Source. (European Bioplastics, 2019).

Standards for home composting

Home composting should only be considered as complementary to industrial composting. This is because Industrial composting generates secondary products and raw materials such as organic fertilizer or bio-waste as feedstock for industrial products. While there is currently no international standard specifying the conditions for home composting of biodegradable plastics, there are several national standards, such as (Rujnić-Sokele & Pilipović, 2017; European Bioplastics, 2019):

- Australian norm AS 5810 "Biodegradable plastics - biodegradable plastics suitable for home composting".
- Belgian certifier Vinçotte (now TÜV AUSTRIA Belgium) had developed the OK compost home certification scheme, requiring at least 90% degradation in 12 months at ambient temperature.
- French standard NF T 51-800 "Plastics - Specifications for plastics suitable for home composting" was developed, specifying the very same requirements for certification.

Labels proving home compostability are OK compost HOME and DIN-Geprüft home compostable.

Figure 24

OK compost logo

DIN-Geprüft home compostable

Principal labels for products home compostable
Source. (European Bioplastics, 2019).

Biodegradability in soil

• The certification scheme "Bio products – degradation in soil" developed by TÜV AUSTRIA Belgium (former Vinçotte) is based on EN13432/EN14995 (Standards for the industrial composting of packaging/plastics) and adapted for the degradation in soil. The test demands at least 90% biodegradation in two years at ambient temperatures.

Counterpart

- In the USA, the standard ASTM 5988 describes a test method for determining the aerobic biodegradation of plastic materials in soil, without giving pass/fail criteria.

The label OK biodegradable SOIL is certified by TÜV AUSTRIA Belgium in case a product meets the requirement of their certification scheme. DIN CERTCO awards DIN-Geprüft biodegradable in soil in accordance with EN17033.

Figure 25

OK biodegradable SOIL

DIN-Geprüft biodegradable in soil

Principal labels for products biodegradable in soil

Source. (European Bioplastics, 2019).

Biodegradability in marine environments

There is currently no standard providing clear pass/fail criteria for the degradation of plastics in sea water (Rujnić-Sokele & Pilipović, 2017). Nevertheless, there are some standard which certify the biodegradation in marine environments, such as (European Bioplastics, 2019; Helian Polymers, 2020):

• ASTM D6691 "Standard Test Method for Determining Aerobic Biodegradation of Plastic Materials in the Marine Environment by a Defined Microbial Consortium or Natural Sea Water Inoculum" (>70% degradation of reference material).

• ASTM D6692 "Standard Test method for Determining the Biodegradability of Radiolabelled Polymeric Plastic Materials in Seawater".

• ASTM D7473 "Standard Test Method for Weight Attrition of Plastic Materials in the Marine Environment by Open System Aquarium Incubations".

• OECD 306 "Biodegradability in sea water" and ISO 16221 "Water quality – Guidance for determination of biodegradability in the marine environment".

At ISO-level, standardization efforts for the requirements for biodegradation of plastics in marine environments are well underway. For example, ISO 18830[5] and ISO 19679[6] are two standards on the test methods for determining the aerobic biodegradation (greater than 60%) of non-floating plastic materials in a seawater/sediment interface, both of which have been published in 2016 and are also eligible on CEN-level (European Bioplastics, 2019).

Nonetheless, these standards are only guidelines and do not provide clear requirements for conditions and timeframes. Research and development are on-going to create harmonized standards for marine biodegradation, which are needed before relevant products can be introduced to the market.

With research underway, questions concerning the limitations for this technology need to be answered: In which context and for which products does this technology make sense and how can it complement a circular economy? Once these questions have been answered, communication and advertising rules need to be defined.

TÜV AUSTRIA Belgium (former Vinçotte) has developed a certification scheme based on ASTM D7081, which demands a biodegradation of at least 90% in 6 months. The corresponding label is OK biodegradable MARINE. However, the certification scheme makes a clear distinction between the certification of the claim and the authorization to communicate about it.

Figure 26

OK biodegradable MARINE

Principal labels for products biodegradable in marine environments

Source. (European Bioplastics, 2019).

2.3 Biobased and biodegradable plastics

2.3.1 Definition and classification

It could be expected that biobased plastics are a new discovery of last century, but surprisingly some were used during human's earliest times. For instance, the Mayas civilization used to play with latex balls. Throughout history, man has relied on biomass to meet his needs and to innovate. Biomass is the whole of living matter: Plant and animal. (Rujnić-Sokele & Pilipović, 2017).

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[5] ISO 18830: 2016. Plastics — Determination of aerobic biodegradation of non-floating plastic materials in a seawater/sandy sediment interface — Method by measuring the oxygen demand in closed respirometer. https://www.iso.org/standard/63515.html

[6] ISO 19679: 2016. Plastics — Determination of aerobic biodegradation of non-floating plastic materials in a seawater/sediment interface — Method by analysis of evolved carbon dioxide. https://www.iso.org/standard/66003.html

Over the past few decades, biodegradable polymers have attracted considerable attention due to the increasing concern about plastic's waste problem. Due to the biodegradability property of some bioplastics the environmental contamination of plastics in natural environments could decrease. Additionally, the consumer demand for highly-quality food products have led to increased interest in the development of biodegradable packaging materials using annually renewable biopolymers.

Furthermore, bioplastics made from biomass or bio-based bioplastics could reduce the dependence in fossil resources and therefore the reduction of GHG emissions which is the principal reason for global warming. Greenhouse effect that suffers plenty of ecosystems and natural life in all its extension (Attaran et al., 2017; Habel et al., 2018).

Due to the special properties and functions that a food packaging must ensure, the replacement of fossil-derived plastics for biomass materials is determined based on the following functional properties (Attaran et al., 2017):

1. Durability
2. Ability to act as a gas barrier
3. Heat resistance
4. Impact resistance
5. Flexibility

Moreover, biobased bioplastics packaging can be classified according to their origin and production method into three classes as illustrated in figure 27 (Robertson, 2012; Andrady, 2015; Attaran et al., 2017; Nakajima et al., 2017):

Class 1 - *Natural derived biomass polymers*

Polymers directly extracted from biomass including chemically modified ones such as cellulose, cellulose acetate, starches, and chitin. Also considered as bio-derived plastics.

Class 2 - *Bio-engineered polymers*

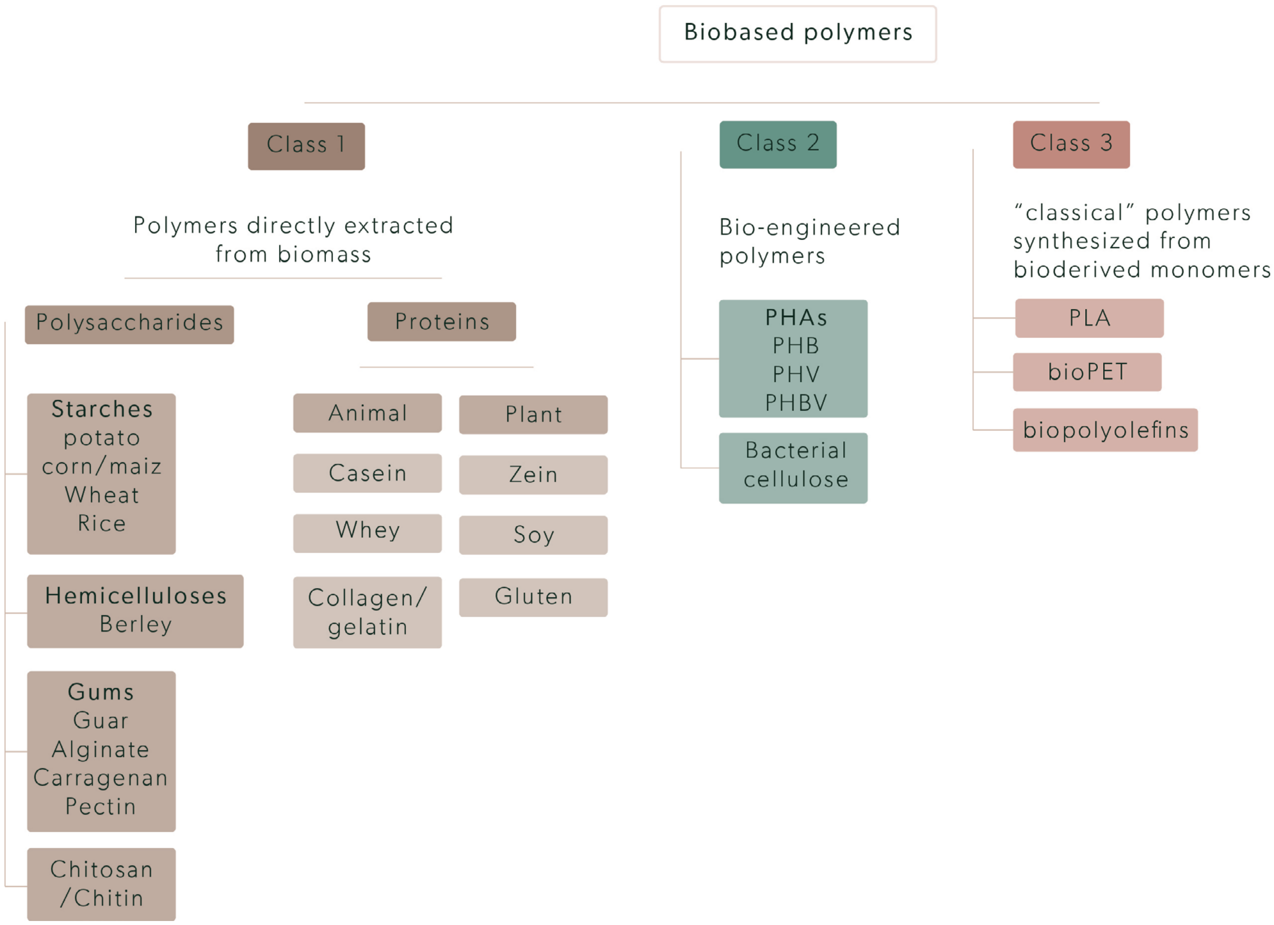
Bio-synthesized by using microorganisms and plants such as PHAs, and poly (glutamic acid).

Class 3 - *Classical polymers*

Polymers produced directly by classical chemical synthesis from biomass monomers such as PLA, PBS, bio-polyolefins, and bio-PET.

This classification considers all biobased plastics independently in its biodegradation property and therefore it is important to clarify that not all these materials are biodegradable. Nevertheless, this classification is considered also to arrange biobased bioplastics which can be properly recyclable, and which will be discussed in next chapters.

Figure 27



Plastics classification by origin and production method
Note. Figure 26 shows an adaptation of the classification of plastics found in the literature studied.
Source (Robertson, 2012; Andrady, 2015; Attaran et al., 2017; Nakajima et al., 2017)

Class 1

In the first class of biobased plastics except for starch derivates, polysaccharides and proteins are used on a wide scale as coatings in food packaging applications, especially as coatings for paper films (Robertson, 2012; Attaran et al., 2017). Therefore, these group will be further discussed individually in the chapter IV.

2.3.2 Starch

Of the annually renewable materials, those that came from starch are the most common. Starch is extracted from cereals (wheat, corn, rice) and tubers (potatoes, manioc). Starch granules are composed of two natural polymers: amylose, and amylopectin (Reichert et al., 2020). Native starch can be converted into a thermoplastic material called "thermoplastic starch" (TPS) in the presence of plasticizers at high temperature (90°C-180°C). This change allows its use as an extrusion, injection molding or blow molding material, similar to most conventional petroleum-based thermoplastic polymers (Robertson, 2012).

Although TPS can be processed in the same way as traditional plastics, its sensitivity to water vapor and poor mechanical properties makes it unsuitable for many rigid packaging applications. The blending of starch with aliphatic polyesters improves their processability and biodegradability, maiking it PCL and its copolymers particularly suitable.
The combination of starch with water soluble polymer such as PVOH has been widely studied since 1970 and is currently applied to produce starch-based loose fillers as a substitute for expanded PS, as well as sheet extrusion and thermoforming.

To decrease the water sensitivity of starch-based materials, another approach is the chemical modification of the starch chains creating commercial water-resistant, starch-based polymers and starch. These materials are made with gelatinized starch (60-86%) and hydrophilic (e.g., EVOH) or hydrophobic petroleum-based biodegradable polymers (e.g., PCL or PBAT known as Ecoflex®) and compatibility agents. The polyesters form the continuous phase leading to materials having relative water resistance and acceptable barrier and mechanical properties (Robertson, 2012).

The most important starch/polyesters-based materials on the market are those produced by Novamont as Mater-Bi®(starch/PCL), Sphere as Bioplast® (starch/PCL), Showa Highpolymer as Bionelle™-Starch (starch/PBSA) and Plantic® (starch/PVOH).

Starch is one of the most interesting material for food packaging among all biopolymers today due to its commercial availability, the appealing balance of properties and industrial scale producibility. Starch has a competitive price within the renewable source material market, it is biocompatible and biodegradable (Attaran et al., 2017).

Class 2

Class 2 polymers consist mainly of the microbial polyesters known generally as PHAs or poly (hydroxyalkanoates), a family formed by renewable, biodegradable, and biocompatible polyesters that were first identified in 1925 by the French microbiologist Maurice Lemoigne (Robertson, 2012).

Bacterial Cellulose (BC), categorized in this class 2, is mainly used for films and coatings applications in food packaging, therefore will be consider in chapter IV.

2.3.3 PHAs

Polyhydroxyalkanoates or PHAs are produced in the form of intracellular particles by many commonly found microorganisms that accumulate PHAs as a carbon and energy sink when grown under nutrient stress in the presence of excess carbon. Under controlled fermentation conditions, some species can accumulate up to 90% of their dry mass as polymer (Robertson, 2012).

PHAs are linear aliphatic polyesters consisting of homo or copolymers of βhydroxyalkanoic acids that can be produced from the fermentation of sugars. The polymers produced are biodegradable and can be decomposed by a PHA-depolymerase detected in several bacteria and fungi in soil, compost, or marine sediment. Due to their characteristics, are suitable to produce packaging materials.

Within PHAs family, Poly (hydroxybutyrate) (PHB) is the most common and from which can derive hydroxybutyrate-co-hydroxyvalerate or PHBV. Although PHB homopolymer is relatively stiff and brittle, the introduction of HV comonomers greatly improves its mechanical properties by reducing the level of crystallinity and melting point, resulting in an increase in toughness or impact resistance. Therefore, the PHA family of polyesters displays a wide range of properties, from hard crystalline plastics to elastic rubbers with melting temperatures of 50°C-180°C (Robertson, 2012).

By changing the ratio of HV to HB, the resulting copolymer can be similar to PP (low HV) or LDPE (high HV) with regard to flexibility, tensile strength and melting point. PHBV has good mechanical and biocompatibility of PHAs can be further improved by blending with other polymers, modifying the surface, or combining PHA with other inorganic materials, thus making them useful for a wider range of applications.
As many biobased materials, the gas barrier properties depend on the ambient humidity. In the case of PHA, its WVTR is similar comparable to LDPE. While PHB has better O_2 barrier properties than PET and PP, and adequate fat and odor barrier properties for applications with short shelf-life products.

PHAs were first developed industrially in the 1960s and commercialized by ICI in the late 1980s under the trade name Biopol®, the first commercial product being a biodegradable, injection blow-molded bottle. Biomer in Munich has produced PHB since 1994 using proprietary bacteria (Robertson, 2012).
Ningo Tianan Biologic Material Co. Ltd was established in 2000 near Shangai and is the world's leading producer of PHBV. Other PHA producers include Greenbio in Tianjin, China, and Biocycle in Brazil. Since 2010, Telles, a joint venture between Metabolix and Archer Daniels Midland Company (ADM), has used proprietary technology for large scale microbial fermentation to produce PHAs under the trade name Mirel® at a commercial-scale plant in Clinton, Iowa.

Class 3

Because bio-polyolefins, bio-PE and its derivates cannot biodegrade but instead can be recycled, they will be considered in Chapter III.

2.3.4 PLA

Of all the possible biopolyesters that have been produced from biobased materials, PLA has shown the highest commercial potential and is now produced on a comparatively large scale. PLA is a linear, aliphatic polyester synthesized from lactic acid monomers. Which can be produced cheaply by the fermentation of glucose obtained from lactose in whey or sucrose in molasses (Robertson, 2012).

Lactic acid (2-hydroxypropanoic acid) can be presented in the form of L-lactide (two L-lactic acid molecules), D-lactide (two D-lactic acid molecules) and meso-lactide (an L-lactic acid and D-lactic acid molecule). The most common commercial polymers of PLA are predominantly L-lactide, with small amounts of D- and meso-lactides.

PLA can be either amorphous or semicrystalline, depending on the stereochemistry and thermal history. Poly (L.lactide) (PLLA) and poly (D-lactide) (PDLA) are semicrystalline polymers, while the atactic polymer, poly (D,L-lactide) (PDLLA) is amorphous (Robertson, 2012). The appearance of PLA is also affected by the crystalline content. Amorphous PLA and low crystalline PLA are clear materials with high gloss while highly crystalline PLA is an opaque white material.

The mechanical properties of PLA are greatly affected by the MW of the polymer, the chain architectures, and the degree of crystallinity, which is determined by the relative proportions of L- and D-lactide in the polymer backbone (Andrady, 2015). Since PLA is rigid and brittle with a low ability to plastic deformation below its T_g (58°C), it is necessary to plasticize PLA to produce flexible films. Plasticizers such a as water, poly (ethylene glycol) (PEG), lactic acid, nontoxic citrates, glycerol and sorbitol have been reported as effective for PLA (Robertson, 2012; Reichert et al., 2020).

Other polyblends such as PLA/PCL blend has resulted in an improvement in mechanical properties and thermal stability without a significant decrease in barrier properties. Furthermore, these combinations permit PLA reduce costs (Van den Oever et al., 2017).

PLA resins can be tailor-made for different fabrication processes and made into films, coextructuded into laminates, thermoformed and injection stretch blow molded into bottles (Andrady, 2015). PLA films also have superior twist retention properties, making them suitable for twist wrap packaging (Robertson, 2012). Due to the specific benefits of PLA such as transparency, gloss, stiffness, printability, processability and excellent aroma barrier, this material can be found in food packaging applications such as coatings for paperboard beverage cartons (BOPLA films[7]), plastic film wraps, barrier films, foods, trays and cup, bottles, within the principal presentations (Van den Oever et al., 2017).

PLA biodegrade at temperatures above the T_g; standard PLA is not considered biodegradable according to ASTM standards but is compostable in industrial composters (Robertson, 2012).

2.3.5 Standards and labels

The following section introduces the most relevant standards and labels for biobased plastics.

Determination of the bio-based content

CEN has developed different standards for the measurement of the renewable content of bio-based materials, including bioplastics (European Bioplastics, 2019).

[7] Biaxially oriented PLA films (BOPLA) which can replace biaxially oriented PP films (BOPP)(Bio-based and biodegradable plastics, 2017).

• EN 16640 – "Bio-based products - Determination of the bio-based carbon content of products using the radiocarbon method", describes how to measure the carbon isotope 14C (radiocarbon method). Depending on the measured amount of biobased carbon, according to certifications can be carried out and the corresponding label(s) can be awarded.
• EN 16785-1 – "Biobased products – Bio-based content - Part 1: Determination of the biobased content using the radiocarbon analysis and elemental analysis", accounts for other biobased elements in a polymer through elemental analysis.
• Part two of this standard EN 16785-2 – "Biobased products - Bio-based content - Part 2: Determination of the bio-based content using the material balance method", describes a material balance method to determine the renewable content of a bio-based product.
• EN 17228 "Plastics - Biobased polymers, plastics, and plastic products - Terminology, characteristics and communication" adopting the horizontal standards of CEN/TC 411 for bio-based plastics and polymers. It includes all relevant topics regarding terminology, bio-based content, Life Cycle Assessment, sustainability, and communication. This standard was published in 2019.

Labels referring to the biobased content of plastics are for example DIN-Geprüft bio-based, OK bio-based (both offering different labels reflecting the product's share of biobased content), and the new logo by Nederlandse Norm (NEN), based on EN 16785-1.

Figure 28

DIN-Geprüft bio-based logo | OK bio-based logo | Nederlandse Norm (NEN) logo

Principal labels for products with biobased content
Source. (European Bioplastics, 2019).

Sustainability and Life Cycle Assessment (LCA)

• ISO 14040 "Environmental management - Life cycle assessment - Principles and framework" (Robertson, 2012; Attaran et al., 2017; Spierling et al., 2018; Hatti-Kaul et al., 2020).
• ISO 14044 "Environmental management - Life cycle assessment - Requirements and guidelines" describes the principles of life cycle assessment (Robertson, 2012; Attaran et al., 2017; Spierling et al., 2018; Hatti-Kaul et al., 2020).
• EN 16760 "Bio-based products - Life Cycle Assessment", which provides specific LCA requirements and guidance for bio-based products based on the ISO 14040 series.

• EN 16751 was developed to standardize sustainability criteria of biobased products. However, it does not include any thresholds or limits and is not suitable for making claims on the sustainability of products or operations.
• SO 14067 "Carbon Footprint of Products", providing detailed information on how to measure and report the carbon footprint of products.
• D7075 "Standard practice for evaluating and reporting environmental performance of biobased products", evaluates the environmental performance of biobased products (Robertson, 2012; Attaran et al., 2017).

There is a number of certification schemes to prove the sustainability of biomass used in a product, for example ISCC PLUS, RSB (Roundtable on Sustainable Biomaterials), or REDcert. However, these schemes are not based on a standard but on the provisions of the EU Directive 2009/28/EC (Renewable Energy Directive).

Figure 29

Roundtable on Sustainable Biomaterials logo

Roundtable on Sustainable Biomaterials logo

REDcert logo

Principal labels for sustainable products
Source. (European Bioplastics, 2019).

Bioplastics – Communication standards

• EN 16848 "Bio-based products - Requirements for Business-to-Business communication of characteristics using a Data Sheet", published in 2016
• EN 16935 "Bio-based products - Requirements for Business-to-Consumer communication and claims", published in 2017.
• ISO 14020 series on "Environmental labels and declaration" is the main international guideline for "green claims". Three different types of environmental labels and declarations are promoted in these standards. ISO 14021 covers self-declared environmental claims, ISO 14024 to environmental labelling. And ISO 14025 to environmental declaration.
• ISO 14063 on "Environmental management - Environmental communication", focusing on setting up communication procedures in companies and containing a general guidance on the basics of environmental communication.
• ISO 14067 also provides general guidelines on how to use carbon footprint claims correctly.

Regarding circular economy, these standards offer a basis for assessing bioplastics and providing sound communication on corresponding claims.

2.3.6 Current Limitations

Generally speaking, the major limitations of most biobased and biodegradable packaging bioplastics for food applications are their performance, processing, and cost (Robertson, 2012; Hatti-Kaul et al., 2020).

The relatively poor thermal and mechanical properties have limited their applications. In particular, their brittleness, low heat distortion temperature, poor resistance to protracted processing operations, and their low barrier to water vapor (Attaran et al., 2017; Habel et al., 2018).

Currently many biorefineries have taken these routes to enhance biobased polymers properties. For instance, now it is possible to find applications in flexible packaging such as flexible films, fruit and vegetable bags, snack packaging. Furthermore, alternatives include rigid packaging such as bottles, coffee cups, containers, trays, deli bowls and lids (See section 2.5) Another feature improved is the heat resistance, a proper example is the formulation of Luminy® which has a heat resistance up to 100°C. Now it is possible to use biobased and biodegradable polymers for food with short and medium shelf-life.

Nowadays, biobased polymers are being used for short shelf-life foods stored at chill temperatures, since the materials are biodegradable. Potential applications include fast food packaging of salads, egg cartons, fresh or minimally processed fruits and vegetables, dairy products such as yoghurt and organically grown foods. While the high CO_2:O_2 permeability ratio of certain biobased packaging materials suggests that they could find application in the packaging of respiring foods such as fruits and vegetables (Robertson, 2012).

In recent years, cost has declined and is expected to decline while production volume is increasing and process optimization achieving better efficiencies.

Limited availability is another concern around biobased plastics (Robertson, 2012). Therefore, utilization of agriculture waste flows as raw materials or integrating production in a biorefinery would provide a dramatic ecological advantage as well as reduce pressure on arable land (Hatti-Kaul et al., 2020).

2.3.7 New techniques and future trends

Nevertheless, the latest studies in nanotechnology and material science prove it is possible to improve the barrier and processing properties of biobased materials using various techniques including (Robertson, 2012; Attaran et al., 2017; Habel et al., 2018; Hatti-Kaul et al., 2020; Reichert et al., 2020):

• Metallization with aluminum
• Coating with thin inorganic or inorganic layers such as silicon oxide (SiOx), aluminum oxide (AI2O3) and diamond-like carbon (DLC)
• Copolymerization or biobased copolymers
• Addition of nanoclays
• Adding additives that enhance properties (mainly fossil-based)

In contrast to the development of novel polymeric materials and new polymerization routes, blending is a relatively cheap and fast method to tailor the properties of plastics. Most commercial blends consist of two polymers combined with small amounts of a third compatibilizing polymer, typically a block or a graft copolymer. Some biodegradable plastics available in the sustainable food packaging market or in study are often comprised of polymer blends that contain partly bio-based (renewable) carbon derived from biomass and partly petrochemical carbon.

Coating with SiOx is the most common commercial technology to increase barrier properties of polymers and PLA bottles have been shown to be compatible with coating processes. For instance, a SiOx coating on PLA reduces the WVTR by 60%. Copolymerization also known as "polyblends" refer to the blending of biobased plastics with other polymers and additives to achieve the desired properties.

Nanocomposite technology in biobased food packaging polymers

Recent advancement in nanocomposite science and technology, compounding of polymers with biopolymer/nanoclay is a technique that can complement the drawbacks of conventional polymers (Attaran et al., 2017). Future biobased food packaging polymers are likely to be blends of polymers and nanoclays (so-called bionanomposites) in order to achieve the desired barrier and mechanical properties demanded by the food industry (Robertson, 2012).

Bionanocomposites are a mixture of biopolymers with nanosized inorganic or organic fillers with special size, geometry, and surface chemistry properties (Chivrac et al., 2009 as cited in Robertson, 2012). Based on where they are found, clay minerals are divided into two main groups (Attaran et al., 2017): Residual clays are manufactured through the surface weathering of rock or shale. They could be produced by chemical decomposition of rocks (e.g granite containing silica and alumina), by solution of rocks (limestone); or by disintegration and shale solution.

1. Residual clay

   Residual clays are manufactured through the surface weathering of rock or shale. They could be produced by chemical decomposition of rocks (e.g granite containing silica and alumina), by solution of rocks (limestone); or by disintegration and shale solution.

2. Transported clay (or sedimentary clay) is removed from the original deposit through erosion and deposited at a distant place.

Clays are naturally occurring, inexpensive and eco-friendly substances and have been found to be useful in various applications.

Natural biopolymer blends are attractive candidates for green synthesis of polymer-based nanocomposites due to numerous advantages of these polymers including low cost, accessibility, biodegradability and flexible processability to improve and develop new sets of polymeric materials with desired properties. A uniform dispersion of nanofillers leads to a very large matrix/filler interfacial area that changes the molecular mobility and the consequent thermal and mechanical properties of the material, including heat distortion temperature and O_2 permeability (Robertson, 2012).

The most studied bionanocomposites are based on nanoclay and polysaccharides, namely, starch and its derivatives, cellulose, chitosan, and pectin. These nanocomposite films show improved mechanical properties. Starch has been the most studied polysaccharide in bionanocomposite systems, including in blends with PLA, PCL, PBS, PHBA and PVOH or with chemically modified (e.g., acetylated) starch matrices.

Furthermore, recent studies show a new and emerging class of clay-filled polymers known as polymer-clay nanocomposites (PCNs) or polymer–layered silicate nanocomposites (PLSNs) which could improve the performance of polymers for food packaging by adding nanoparticles (Attaran et al., 2017). PCNs represent one of the most promising classes of materials of the past few decades and have received much attention due to a significant increase in the mechanical and barrier properties in addition to the ease of preparation through simple processes for packaging applications.

The incorporation of nanoclays into packaging also offers a reduction in raw material use, less dependence on specialty products, elimination of secondary processes, less complex structures, and a reduction in machine cycle time. Other potential benefit of natural biopolymer-blended nanocomposite is the positive environmental impact due to the resulting materials can biodegrade. However, the lack of compatibility and interfacial adhesion between the nanoclay, natural biopolymer and matrix phase results in the essential need of compatibilizers. Which contracts its commercial widespread, thus, there is still studies and progress to achieve.

2.4 Fossil-based and biodegradable bioplastics

This group of bioplastics can be considered behind a sustainable approach due to their ability to biodegrade. Furthermore, fossil-based and biodegradable plastics are used in polyblends with other biobased biodegradable polymers to enrich their mechanical and barrier properties and achieve food packaging requirements. The best-known petrochemical-based biodegradable polymers are aliphatic polyesters or aliphatic-aromatic copolymers (Robertson, 2012).

2.4.1 PCL

PCL or poly (caprolactone) is a flexible, aliphatic, semicrystalline polyester that is miscible with many other polymers. It can biodegrade aerobically by a large number of microorganisms in various environments. However, the high cost and low performance of PCL has prevented its industrial widespread use in food packaging. PCL can be mixed with starch to lower its cost and increase its biodegradability, for example, Mater-bi® produced by Novamont (section 2.5). The PCL limits moisture sensitivity, boosts melt strength and helps plasticize the starch. With PCL contents of up to 20%, starch-PCL films have proved to be excellent O2 barriers. However, increasing the PCL content beyond this level impaired the O2 barrier properties while improving the water barrier properties (Robertson, 2012).

2.4.2 PGA

Poly (glycolic acid) (PGA) is a biodegradable, thermoplastic polymer, and the simplest linear, aliphatic polyester. PGA is a polyester resin that offers high gas barrier to both CO_2 and O_2, controllable hydrolysis and excellent mechanic strength, making PGA ideally for high performance packaging. The targeted application for PGA is multilayer PET bottles for carbonated soft drinks and beer. Since PGA offers a gas barrier 100 times higher than of PET, it is possible to reduce the amount of PET used in these bottles by more than 20%, while maintaining the equivalent barrier against CO_2 loss. PGAs unique hydrolytic properties make it highly compatible with widely practiced industrial PET recycling processes, ensuring that the material does not interfere with the purity and quality of recycled PET. In another packaging application, PGA multilayer designs have been shown to enhance the gas and moisture barrier of biobased polymers such as PLA (Robertson, 2012).

Production of PGA for packaging applications (marketed under the trade name Kuredux®) commenced in the United States, Europe and Japan and biodegrades into CO2 and water in compost within 1 moth at a similar rate to cellulose (Robertson, 2012).

2.4.3 PBAT

Poly (butylene adipate-co-terephthalate) or PBAT is an aliphatic-aromatic copolyester, synthesized from 1,4-BDO, adipic acid and TA. The polymer was developed by BASF especially for applications using compost as the disposal route and was designed to be a strong and flexible material with mechanical properties similar to polyethylene. As a consequence, it can be melt-processed on standard polyolefin equipment. Today, these types of polymers offer very good combinations of biodegradation and material properties and can be used for many applications (Eubeler et al., 2010). By adding special additives and optimizing the processing conditions, transparent cling films can be obtained using a blow film process. These films can be used for food packaging, including fresh meats, vegetables and fruits sold in supermarkets. It is sold under the name Ecoflex® and can be blended with starch as well as PLA (e.g., Ecovio® by BASF) to widen its properties while still retaining its biodegradable (Robertson, 2012).

2.4.4 PBS

Polybutylene succinate (PBS) and its copolymers are a family of biodegradable polymers with excellent biodegradability, thermoplastic processability, and balanced mechanical properties similar to those of polypropylene (PP) is usually synthesized via polycondensation of succinic acid (or dimethyl succinate) and BDO. The monomers can be obtained from petrochemical-based or renewable resources. At present, succinic acid is commercially manufactured via hydrogenization of maleic anhydride to succinic anhydride, followed by hydration to succinic acid. Succinic acid can also be obtained from fermentation of microorganisms on renewable feedstocks such as glucose, starch, and xylose. While conventional commercial processes for BDO synthesis use fossil-based feedstocks have been developed and are being commercialized (Xu & Guo, 2010 as cited in Robertson, 2012 & Reichert et al., 2020).

Biodegradable aliphatic polyesters, trademarked Bionolle™ and manufactured by Showa Highpolymer, include PBS and PBSA. They are produced through polycondensation reactions of glycols such as EG and 1,4-BDO, and aliphatic dicarboxylic acids such as succinic acid and adipic acid. Bionolle™ polymers are white, crystalline thermoplastics with melting points ranging from 90°C-120°C, T_g ranging from -45°C to -10°C and density about 1250 kgm-3. They have excellent processability, so they can be processed on conventional equipment commonly used in processing polyolefins at temperatures of 160°C-200°C into various molded products such as injected, extruded and blown ones (Robertson, 2012). PBS can also be used as an additive for plasticizing other bio-polymers such as PLA (Reichert et al., 2020).

2.4.5 PPC

Polypropylene carbonate (PPC) is a form of aliphatic polycarbonate a polymer synthesized from the catalyzed copolymerization between CO2 and propylene oxide (PO).
Since PPC is an amorphous polymer with a low Tg, it is common to make polyblends with crystalline biodegradable polymers that can improve its mechanical properties. Many biodegradable crystalline polymers such as PLA, PBS and Ecoflex have been used for such purposes. However, its poor thermal stability, mechanical strength, and dimensional stability have limited its applications and makes it unsuitable as a stand-alone packaging material (Flodberg et al., 2015). To enhance the thermal properties of PPC, last studies are based in nanocomposites (Robertson, 2012; Muthuraj & Mekonnen, 2018).

2.5 Case studies

The state-of-the-art biodegradable bioplastics is presented in this section. Initially is cited four study cases of this group and is continue with the collection of the last biodegradable bioplastics trademark's suitable for food packaging applications (Robertson, 2012; Andrady, 2015; Nakajima et al., 2017; Hatti-Kaul et al., 2020).

Mater-Bi® Cling Film

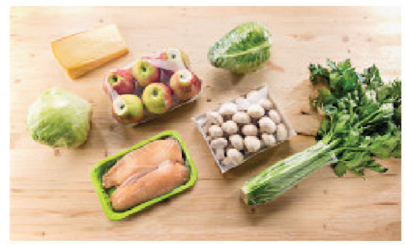
NOVAMONT

Mater-Bi® Cling Film is one of the last launches of Novamont bioplastics. This biobased cling film have been designed to provide long shelf life to fresh food. The film is intended to be used in the large-scale retail trade and large and medium-sized packaging centres.

Novamont
Italy, since 2020

Material Sustitution
PS, LDPE

References
https://www.novamont.com/eng/read-press-release/novamonts-new-mater-bi-cling-film-is-now-available/



SUSTAINABILITY

Origin
~100%
biobased

End-of-life
biodegradable
compostable

Certifications
Industrial Composting: European standard EN13432
Recycling: European standard EN 13430
Energy recovery: European standard EN 13431
Meets the Essential Requirements of the European Directive on Packaging and Packaging Waste (94/62/EC)

ADDITIONAL PROPERTIES

high transparency

anti-fog effect

ENGINEERING PROPERTIES

strong breathability

high levels of mechanical performance

PRESENTATIONS

flexible packaging

FOODS

perishable and semi-perishable foods

PROCESSING TECHNOLOGY

most packaging machines

Luminy®

TOTAL | Corbion

(L105, L130, L175, LX575, LX530, LX175, LX975, LX930, D120, D070)

Luminy®PLA grades can be used in a broad range of food packaging appications using various conversion technologies. Luminy®PLA grades can be used as a part of a compound in case of flrexible packaging.

Total & Corbion
France & Netherlands,
since 2016

Material Sustitution
Thermoplastics

References
https://www.total-corbion.com/luminy-pla-portfolio/

SUSTAINABILITY

Origin
100%
biobased

End-of-life
Biodegradable
compostable
Recyclable

Certifications

Industrial Composting: EN13432 and ASTM D6400.
Biobased content: 100% according to EN16785-1 and ASTM D6866.
Reduced carbon footprint: LCA study
Made from GMO-free crops: Bonsucro-certified PLA

ADDITIONAL PROPERTIES

- transparent excellent gloss
- printtable
- sealable

ENGINEERING PROPERTIES

- rigidity and good flavor and aroma barrier
- heat resistance up to 100°C.

PRESENTATIONS

flexible and rigid packaging

FOODS

perishable and semi-perishable foods

PROCESSING TECHNOLOGY

Film extrusion
Thermoforming

Mirel™

(F1005, F1006, F3002)

Mirel is made from a family of PHA bioplastics that are biobased, biodegradable, and compostable. Its performance makes it suitable for food packaging applications. Depending on the Mirel grade, it can be biodegradable in natural soil and water environments, home composting systems, and industrial composting facilities.

Metabolix & ADM:
Telles
USA, since 2010

Material Sustitution
PP, LDPE, LLDPE, HDPE

References
http://www.mirelplastics.com/environmental-values/glossary-of-terms/

SUSTAINABILITY

Origin
100%
biobased

End-of-life
biodegradable
compostable

Certifications

Industrial Composting:
EN13432 and ASTM D6400.

ADDITIONAL PROPERTIES

coulored

color laser printtable

sealable

8 - 125μm typical thicknesses

ENGINEERING PROPERTIES

Inherent heat and moisture resistant

Ranging from flexible to rigid

Withstands hot liquids, HDT > 120°C

PRESENTATIONS

flexible and rigid packaging

FOODS

perishable and semi-perishable foods

PROCESSING TECHNOLOGY

Thermoforming
Injection moulding

Ecovio®

F2223, F2224, F2331, F2332, F2341, F23B1, FS22C3, FS2312, FS2341

Ecovio® compound combine the benefits of ecoflex® but with higher content of renewable raw materials. Ecovio is a finished product, therefore, additional blending is not required. These specific grades are designed as drop-in solutions suitable for blown film processes.

BASF
Germany, since 2007

Material Sustitution
LDPE

References
https://plastics-rubber.basf.com/global/en/performance_polymers/products/ecovio.html

SUSTAINABILITY

Origin
partly biobased

End-of-life
biodegradable
compostable

Certifications

Industrial Composting:
European standards EN13432 and the American Standard ASTM D6400.

ADDITIONAL PROPERTIES

white translucent color

Printable in 8 colors by flexo printing

20 - 120µm typical thicknesses

ENGINEERING PROPERTIES

High strength, stiffness and failure energy (dart drop)

Good thermostability up to 230 °C

PRESENTATIONS

flexible packaging

FOODS

perishable foods

PROCESSING TECHNOLOGY

blown film extrusion

Trademark by Company, Country	Type of material	Feedstock	Substituted material	Circular-life

Class 1

BIOPLAST GS 2189 by Sphere, France ~2019	Starch/PCL	>69% Biobased	PE-LD PE-HD PP PS PUR	Compostable Recyclable
BIOPLAST 105 by Sphere, France 2018	Starch/PCL	67% Biobased	PE-LD PE-HD PP PS PUR	Biodegradable Recyclable
BIOPLAST 300 by Sphere, France 2016	Starch/PCL	>30% Biobased	PE-LD PE-HD PP PS PUR	Compostable Recyclable

General properties	Processing technologies	Applications	References
Coulored with masterbatches Printable by flexographic and offset printing without pretreatment Sealable (hot, RF, ultra sonic)	Injection moulding Thermoforming Blown film extrusion	Rigid packaging	http://www.bio-tec-group.de/BroschBioplastGS2189_EN_Web.pdf
Coulored with masterbatches Printable by flexographic and offset printing without pretreatment Sealable (hot, RF, ultra sonic)	Injection moulding Thermoforming Cast film extrusion Blown film extrusion	flexible packaging	http://mater-bi.com/en/wp-content/uploads/sites/2/2016/05/scheda-packaging_EN_TUV_LR-.pdf
Coulored with masterbatches Printable by flexographic and offset printing without pretreatment Sealable (hot, RF, ultra sonic) Soft natural texture	Blow film extrusion	flexible packaging	http://www.bio-tec-group.de/BroschBioplast300_EN_Web.pdf

Trademark by Company, Country	Type of material	Feedstock	Substituted material	Circular-life

Class 1

Trademark by Company, Country	Type of material	Feedstock	Substituted material	Circular-life
ALGX Compostable by Eranova, France 2020	Starch (green alge based)	~100% Biobased	PE-LD PP PS	Compostable Recyclable
BIOPLAST 900 by Sphere, France 2014	starch/PCL	69% Biobased	PE-LD PP PS	Compostable Recyclable
BIOPLAST 500 by Sphere, France 2013	Starch/PCL	>50% Biobased	PE-LD PE-HD PP PS	Compostable Recyclable

General properties	Processing technologies	Applications	References
Produce 20 times more biomass than current land-based plant crops bioplastic formulation improves by 15% the mechanical properties of current bioplastics	Injection moulding Thermoforming Blow film extrusion	Flexible packaging Rigid packaging	https://eranov-abioplastics.com/?lang=en
Coulored with masterbatches Printable by flexographic and offset printing, pretreatment is recommended Sealable (hot, RF, ultra sonic)	Injection moulding Thermoforming	Rigid packaging	http://www.bio-tec-group.de/BroschBioplast900_EN_Web.pdf
Coulored with masterbatches Printable by flexographic and offset printing without pretreatment Sealable (hot, RF, ultra sonic) Soft natural texture	Blow film extrusion	flexible packaging	http://www.bio-tec-group.de/BroschBioplast500_EN_Web.pdf

Trademark by Company, Country	Type of material	Feedstock	Substituted material	Circular-life
Class 2				
(PHA) bioplastics by Venvirotech, Spain 2018	PHA	**~100% Biobased**	PP PS LDPE LLDPE	Biodegradable compostable
Class 3				
Ecovio®PS1606 by BASF, Germany ~2013	Ecoflex /PLA/additives	**Partly Biobased**	PE LDPE	Biodegradable Recyclable
Ecovio® (IA1652, IS1335, T2308, TA1241) by BASF, Germany ~2013	Ecoflex /PLA/additives	**partly Biobased**	PE LDPE	Biodegradable Recyclable

General properties	Processing technologies	Applications	References
Quickly biodegradable: 6-12 months Do not cause toxic effects. Suitable for polyblends	Thermoforming Blown film extrusion Injection moulding	Rigid packaging	https://www.ven-virotech.com/en/
Barrier for fat, liquids, aromas and mineral oils Relatively high WVTR Good printability using alcohol and/or water based colors Weld strength comparable to LDPE at temperatures 20 to 30°C below LDPE level Good scratch resistance	Extrusion coating Thermoforming	Flexible packaging Rigid packaging	https://plas-tics-rub-ber.basf.com/-global/en/perfor-mance_poly-mers/products/ec ovio.html
Printable Sealable Nature opaque and easy to colour High strength and stiffness Good thermostability up to 205 °C in processing High, but controllable (WVTR) Medium (OTR)	Injection moulding Film extrusion Thermoforming	Rigid packaging	https://plas-tics-rub-ber.basf.com/-global/en/perfor-mance_poly-mers/products/ec ovio.html

Trademark by Company, Country	Type of material	Feedstock	Substituted material	Circular-life
Class 3				
BIOPLAST GF 106/02 by Sphere, France 2001	Starch/PCL	**Partly Biobased** (natural potato starch)	PE-LD PE-HD PP PS PUR	Compostable Recyclable
Bio-Flex® F 1100 by FKuR Kunststoff GmbH, Germany ~2018	PLA	**partly biobased**	LDPE	Biodegradable compostable
Bio-Flex® Home Compost (grades) by FKuR Kunststoff GmbH, Germany ~2017	PLA	**10-40% Biobased**	LDPE	compostable

General properties	Processing technologies	Applications	References
Coulored with masterbatches Printable by flexographic and offset printing without pretreatment Sealable (hot, RF, ultra sonic) Soft natural texture	Blown film extrusion Injection moulding	Flexible packaging Rigid packaging	http://www.bio-tec-group.de/BroschBioplast-GF10602_EN_Web.pdf
Translucent Density: 1.25 g/cm³ Melting temperature: > 155 °C	Thermoforming Blown film extrusion Injection moulding	Flexible packaging	https://fkur.com/wp-content/uploads/2019/03/FKuR-technical-data-sheet-bio-plastics-compound-blown-film-BIO-FLEX_F_1100_EN.pdf
Translucent to opaque grades Thickness: 8-28µm. Melting temperature ~155 °C	Thermoforming Blown film extrusion Injection moulding	Flexible packaging	https://fkur.com/en/processes/blown-film-extrusion/

Trademark by Company, Country	Type of material	Feedstock	Substituted material	Circular-life
Futerro® by Futerro, France & Belgium 2010	PLA	**Biobased** (sugar beet, sugar cane, wheat, maize and cellulose)	PP PS LDPE LLDPE HDPE	Biodegradable compostable Recyclable

FOSSIL-BASED AND BIODEGRADABLE BIOPLASTICS FOR FOOD PACKAGING APPLICATIONS

Trademark by Company, Country	Type of material	Feedstock	Substituted material	Circular-life
ecoflex® F Blend C1200 by BASF, Germany 2011	starch/PBAT	**Fossil-based**	PE-LD	Compostable Biodegradable

General properties	Processing technologies	Applications	References
Transparent nature and excellent gloss Printable Aroma and taste barriers Oil and fat resistance Shrink properties Very good mechanical properties	Thermoforming Blown film extrusion Injection moulding	Rigid packaging	http://www.futerro.com/endoflife-management.html
Transparent Printable Good thermostability High elongation at break and high failure energy High, but controllable water vapour transmission rate (WVTR)	Blown film extrusion Cast film extrusion	Flexible packaging	https://plastics-rubber.basf.com/global/en/performance_polymers/products/ecoflex.html

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Chapter III.

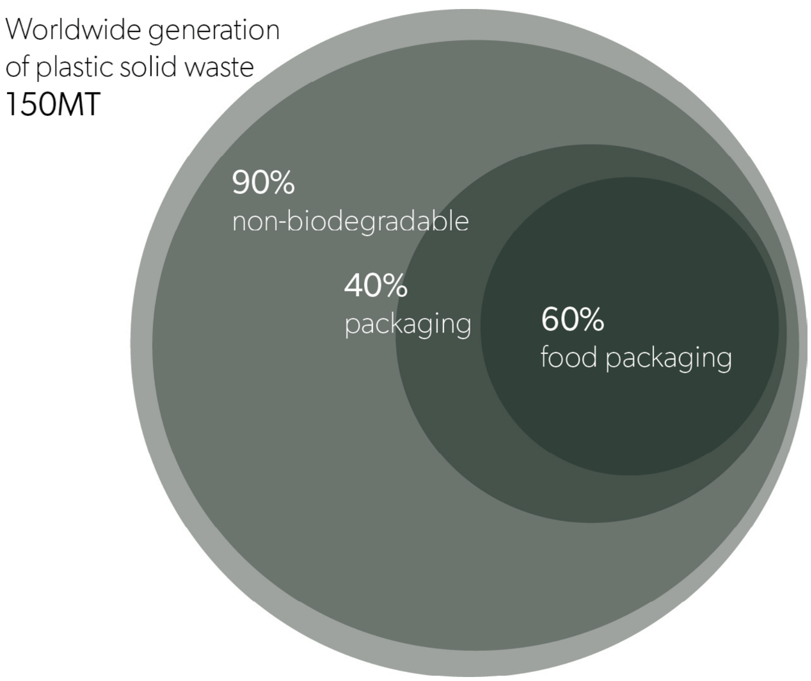
RECYCLABLE PLASTICS

3.1 Introduction and classification

Worldwide generation of plastic solid waste (PSW) increases daily and is currently around 150 million tonnes per annum (Rahimi & García, 2017). Now, around 90% (Narancic et al., 2020) of total plastics worldwide produced annually are not biodegradable, of which 40-44% (Rai et al., 2021) correspond to the packaging sector and where almost 60% is used for food and beverage packaging applications (Matthews et al., 2021). Packaging is the largest plastics sector in production and is the largest plastic discarded group. Most of the packaging plastics produced are discarded the same year they were produced. For example, in 2015, 42% of plastics produced (146 Mt) was used as packaging whereas plastic waste leaving use was 54% packaging (141 Mt) (Geyer et al., 2017; Narancic et al., 2020).

Unfortunately, globally, only 2% of the total plastics waste is recycled. In Europe approximately 27.1 million tons of postconsumer plastic waste were recycled in 2016, of which31.1% was recycled, 41.6% was incinerated and 27.3% went to landfill (Hatti-Kaul et al., 2020). That still leaves the large volume of plastic waste posing a tremendous environmental problem.

Figure 30



Average Annual Global Plastic's Production and Food and Beverage Packaging Participation

Note. Author representation based on the sources. The estimation of global plastic solid waste is based in 2015; the approximated percentage of non-biodegradable plastics correspond to study done in 2016; the approximated percentage of plastic packaging sector correspond to a study done in 2018; and the approximated participation of food packaging segment in plastic production is based in a study done in 2019

Source. (Rahimi & García, 2017; Narancic et al., 2020; Matthews et al., 2021; Rai et al., 2021).

Current statistics estimate that at least eight million tonnes of plastics leak into the ocean each year. According to current statistics released by The European Commission[1] it represents around 1.5–4% of the global plastic produced annually. Furthermore, approximately 79% of all plastic ever made has not been recycled, generating a large volume of plastic waste (Narancic et al., 2020).

At the moment, a low recycling rate of approximately 14% for single use plastics can be observed globally. In Europe, recycling rates vary widely across member countries. While for many countries landfilling is the first or second option for plastic waste treatment, countries such as Switzerland and Germany have implemented landfilling restrictions and have achieved landfilling rates of less than 10%. Fortunately, since 2016 the figure for recycling in Europe is more promiser with an average recycling rate of 31.3%. Under the 7th Environmental Action plan[2], has outlined that all member states must end incineration of recyclable materials and reach a recycling rate of 50% by the year 20203 (Narancic et al., 2020).

Not only plastics can damage the environment and trigger the death of species if they finish up on other environment not proper with its end of life. Several chemical additives, such as plasticizers, flame retardants, stabilizers, antioxidants, and pigments, included in plastic products to enhance their polymer properties and prolong their shelf life can be hazardous to the environments, humans, and other organisms (Hatti-Kaul et al., 2020).

This is the principal reason why recycling comprises a proper technology towards a more circular plastics economy for polymer's solutions. Plastics recovery and recycling should be part of any plan to tackle plastic waste specially for those plastics that are not able to biodegrade. Durable plastics must follow a circular approach to avoid finish in landfills per centuries, which is the less favored alternative behind a sustainable standpoint (Narancic et al., 2020).

Besides recycling permits to give more than one life cycle to plastics products, it is a fundamental technology to return the economic value of discarded food packages to the active economy. It is estimated that 95% of the value of plastic packaging material (70 million-105 billion euros) is lost after its first use cycle (Hatti-Kaul et al., 2020). These low recycling rates mean large losses of the material value to the economy. Thus, plastics are now at the top of the political agenda in Europe and across the world. Recycling implies that plastics, instead of being discarded as waste, should re-enter the economy as valuable commodities, hence retaining their economic value as well as conserving natural resources, reducing waste, and minimizing their carbon footprint.

Moreover, through recycling the use of petrochemical resources is avoided to produce more virgin plastics. Many studies show recycling is a method to decrease CO_2 and greenhouse gases emissions. Recent analysis also prove that recycling saves more heat and energy reinforcing the circular economy approach for food packages products. On average, each tonne of plastic recycled saves ~130 million kilojoules (123 million British thermal units (BTU)), a value equivalent to the energy liberated on combusting ~22 barrels of oil4 (Rahimi & García, 2017).

Furthermore, not all biobased polymers are biodegradable but can be successfully recycled in the same industrial process as commodities plastics. Thus, after the possibility of plastics to biodegrade in certain environments, recycling is promising sustainable alternative for this bioplastic's group.

[1]The European Commission. https://ec.europa.eu/info/index_en

[2]The 7th Environmental Action Programme (EAP). https://ec.europa.eu/environment/action-programme/

[3]Official updated data about recycling rates for EU in 2020 is not yet available.

This chapter will consider the bioplastics suitable for food packaging applications that cannot biodegrade but can be efficiently recycled. These non-biodegradable bioplastics solutions constitute those which come from fossil based (section 3.5), and those biobased polymers (section 3.6).

Figure 31

Average Annual Global Non-biodegradable Plastics Segregation by Raw Material Origen

Note. Author representation based on the sources. The approximated participation of fossil-based and bio-based plastics in non-biodegradable plastics is based on studies done from 2016 to 2020, where the variations in percentage are minimum.

Source. (Rahimi & García, 2017; Hatti-Kaul et al., 2020; Narancic et al., 2020; Rai et al., 2021; Matthews et al., 2021)

3.2 Current problematic

While biodegrading is a natural process conducted in managed environments (home composting, industrial composting, anaerobic degradation), recycling plastics is an industrial process that reuses the same or initial materials for more than one life cycle.

Recycling of plastics is not a simple process due to the huge variety of currently produced and rapidly discarded polymers. For many reasons such as consumer education, local municipal organization, and economic restraints of sorting waste; dozens of different polymers with multiple contaminants enter to the same recycling process. This heterogeneous mixture of plasticizers, stabilizers, dyes, and other additives makes difficult the production of high-purity monomers. The presence of additives and impurities complicates the recycling procedure, affecting the processing cost and properties of the recycled product (Rai et al., 2021). Therefore, correct post-consumer sorting and automated separation techniques are essential to reach higher recycling rates of plastic.

On the other side, the continued recycling of a plastic results in a deterioration in its desirable properties when compared to the virgin material. Although recycled materials may have physical properties similar to those of virgin plastics, the resulting monetary savings are limited, and the properties of most plastics can be significantly compromised after several processing cycles. Therefore, research efforts towards the development of efficient recycling methods for all PSW components will be critical in realizing the substantial economic and environmental benefits associated with recycling (Rahimi & García, 2017).

Biodegradable plastics in the recycling waste stream

Plastics pollution is a big problem that involves manufacturers, systems, government, and users. The heterogeneous blends of polymers and the diversity of their components may result in some complications in the treatment and quality of existing plastic recycling systems. For example, the addition of starch or natural fibers to traditional polymers can complicate recycling processes. Although it is feasible to mechanically recycle some bioplastic polymers, such as PLA, a few times without significant reduction in properties, the lack of a continuous and reliable supply of bioplastic polymer waste in a large scale presently makes recycling less economically attractive than for conventional plastics (Rujnic-Sokele & Pilipovic, 2017).

Stakeholders from the recycling industry have raised the concern that the proportion of reprocessed materials will contain biodegradable parts and thereby the technical characteristics (e. g. strength, durability, etc.) of the final product would be compromised (Rujnic-Sokele & Pilipovic, 2017). This being because these products tend to have inferior long-term properties even after proper stabilization, thereby limiting their market ability (Attaran et al., 2017).

Furthermore, some food packaging alternatives such as multilayer lamination of different biopolymers and materials used to improve the food shelf-life also compromised its recyclability (Rujnic-Sokele & Pilipovic, 2017).

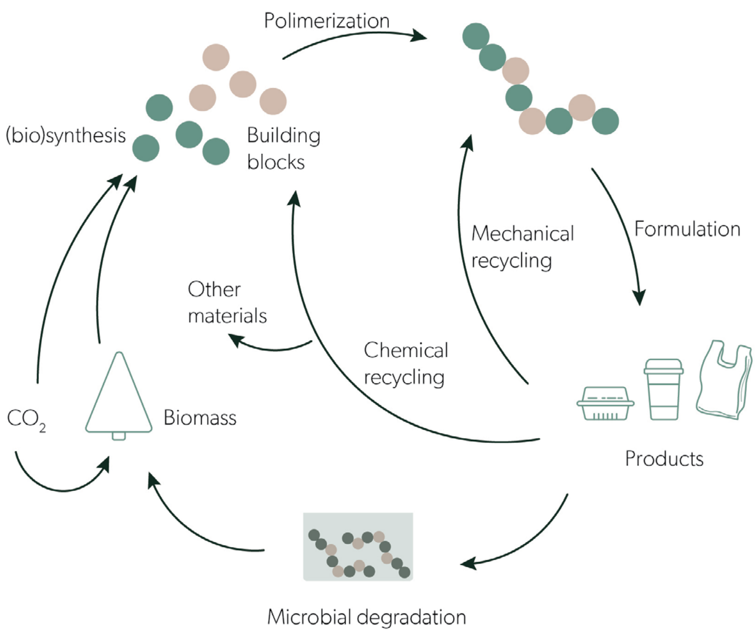
Thus, recycling requires prior collection and segregation steps, which have an important role in producing quality end-products. The issue is that many biodegradable and conventional plastics cannot be distinguished by the optical systems used for waste separation. In addition, both types of products have similar weights and densities, which prevent any easy mechanical separation, and increases the cost of processing and thus the price of recycled products (Attaran et al., 2017; Rujnic-Sokele & Pilipovic, 2017). Good practices in food packaging sorting are the responsibility of the food product brand, the customer or user, and local government waste administration.

On the other hand, the biodegradability of material generated from recycled plastics could be further characterized and investigated. Currently, very little information is available on the degradation of recycled plastics. As these plastics often consist of a blend of polymers and may have had stabilizers and other reagents added during the recycling process, the variety of interactions within the components of recycled plastic is considerably more complex than that of plastics generated from virgin material. Some information is available on the abiotic degradation of some of these mixed blend materials, suggesting that they are more stable against photo- and mechanical degradation than virgin plastic. This will slow down the rate of biodegradation. It is probable that the slower initial stage is due to anti-ageing additives and stabilizing reagents making different polymers compatible (Annemette et al., ca. 2019).

3.3 Recycling methodologies and technologies for plastics

Several approaches have been investigated to address plastics packaging waste problems, such as designing highly selective catalysts and unique chemistries to effectively depolymerize plastic materials into building blocks or monomers for polymer production or designing additives for more effectively recycling. In the short term, these approaches could be part of the solution to reduce plastic waste. Still, for longer term implementation, questions such as economic viability of the process must be addressed and appropriate recycling strategies for the upcycled plastics must also be considered (Sangroniz et al., 2019). Nevertheless, at the moment, chemical and mechanical recycling are the two approaches most extensively used for plastic based products (Figure 32).

Figure 32



Presentation of the Production of Biobased Plastics and their Recycling
Source. (Rahimi & García, 2017)

Mechanical recycling is the most common and economical method available for recycling postconsumer plastic waste, and involves collection, washing, sorting based on plastic type, and grinding of the material into smaller fragments for remolding. Given that the process results in varying degrees of polymer degradation, mechanical recycling is limited by the number of reprocessing cycles (Narancic et al., 2020; Hatti-Kaul et al., 2020).

Mechanical recycling is operated in two modes: primary and secondary recycling. Primary or closed-loop recycling implies reprocessing of the plastic back to the product used for the same purpose as the original plastic (Hatti-Kaul et al., 2020). Nevertheless, the most important drawback of this method is that can only use of almost unaltered waste, such as process scrap or post-consumer materials of known origin. The production of plastic bottles from blends of recycled PET (rPET) and virgin PET is a noteworthy example of primary recycling (Rahimi & García, 2017).

Secondary recycling affords materials for uses different from those for which the original material was initially manufactured. Most postconsumer plastics such as PET, HDPE, LDPE are recycled through this process. Also, other polymers lower in value, thus this process is often called "downgrading" or "downcycling" (Rahimi & García, 2017).

In mechanical recycling, sorting is the decisive process stage, due to the final product quality depend on it. The sorting of PSW is important to remove contaminants, such as metals, wood, and rubber, and to separate the individual polymer materials, each of which may respond very differently to reprocessing. As the manual separation of plastics is ineffective, challenging, and time-consuming, automated separation techniques have been developed to expedite the process. Such sorting techniques rely on measurable differences in material properties, such as density, electrostatics, wettability, or spectral signatures. For example, the densities of commercial polymers can differ greatly, with PVC (~1.10–1.45g/cm^3) and PET (~1.38–1.40g/cm^3) being denser than polyamide (~1.07–1.18g/cm^3), polystyrene (~1.04–1.11g/cm^3) and polyethylene (~0.91–0.97g/cm^3) (Rahimi & García, 2017).

The complex demands of polymer recycling have necessitated advancements in mechanical and optical technologies for materials identification and separation. State-of-the-art mechanical and optical separation technologies have shown promise to sort plastic components from mixed waste with particles as small as 2mm. These methods thus maximize the usefulness of processing PSW sources. Mechanical separation systems target the physical properties of materials, which can be further distinguished by using optical techniques in parallel. Some of the most used technologies for plastics separation are summarized in table 8 (Rahimi & García, 2017).

Table 8. *Technologies for plastic separation*

TECHNOLOGIES FOR PLASTICS

Magnetic density separation
Separates waste according to the density differences between plastics, despite an overlapping or narrow range of densities for different plastics. Magnetic density separation (MDS) uses a mangnetic mixture (nanometre-sized FeO particles suspended in water), the effective density of which varies vertically in a magnetic field, to sort plastic particles of varying density. MDS enables single-step, accurate and rapid material sorting for high-density polyethylene, low-density polyethylene and polypropylene from each and from other materials such as rubber and metals.

Triboelectric separation
Uses friction to charge polymer particle surfaces and separate them based on their anionicc or cationic nature.

Froth flotation methods
Target the critical surface tension (wettability) of plastics and separate them based on their hydrophobic or hydrophilic character.

Speed accelerator
Work to detarminate polymers from other materials at high speed for further separation.

Solvent-based recovery
Effective for polyvinyl chloride, which is soluble in organic solvents and can thus be separated form insoluble materials.

Hyperspectral imaging
Uses an in-line sensor technology that combines spatial and spectral analysis of defined solids. Cameras perform 3D imaging of the materials by interrogating them with light in the 400-1,700 nm spectral range (in the visivle and infrared). Hyperspectral imaging technology is exploited as a quality control technique to continuosly monitor processes in which polyolefind are separeted by mechanical methods such as MDS.

Laser-induced breakdown spectroscopy
Used to evaluate compositional elements of materials by comparing real-time atomic emission spectral data of plastics to references sources.

X-ray fluorescence and infrared spectroscopy
These are examples of other methods that are used for spectroscopic separation puroposes.

Ultrasound technology
Currently used for commercial medical imaging and has also shown promising results for monitoring the separation of plastics waste and assessing process quality information.

Source. (Rahimi & García, 2017).

An alternative approach to processing plastic solid waste is chemical recycling, which relies on the affordability of processes and the efficiency of catalysts. Chemical recycling uses a chemical process to degrade polymers into their petrochemical constituents, which may either be repolymerized to the original product or converted into other useful products, such as basic chemicals and/or polymers for new plastics or fuels. An example of chemical recycling is pyrolysis (thermolysis), in which plastics are subjected to high temperatures in the presence of a catalyst to give a mixture of smaller molecules that are difficult to separate. An alternative would be selective degradation of the polymer chains under controlled conditions to the precursor building blocks. Which are purified and subsequently repolymerized to form building blocks for new polymers (Hatti-Kaul et al., 2020).

Chemical recycling is not practiced to a large extent on an industrial scale because the present methods require a large energy input (Hatti-Kaul et al., 2020). However, if the pure monomer could be recovered through chemical recycling, the prices of polymers would be decoupled from oil prices. This has motivated the search for mild processes for the catalytic conversion of polymers directly to monomers or to new polymers (Rahimi & García, 2017).

The discovery of catalytic methods for the chemical recycling of polymers, operative under mild temperatures and high selectivity, could aid monomer recovery on an industrial scale. Processing methods should be simple and not energy-intensive, with monomers easily isolable and reversions highly efficient. Crucially, the materials must also possess mechanical and thermal properties necessary for the target applications while maintaining their environmental robustness to serve as drop-in alternatives to the plastics currently used (Rahimi & García, 2017).

Mechanical processing of plastics is a complex process but usually it is preferred due to its cost-effective method compared with chemical one (Rai et al., 2021). Nevertheless, the recover-and-recycle rates for plastics are extremely low due to the low efficiency of mechanical recycling. State-of-the-art sorting technologies based on the physical properties of polymers (for example, magnetic density separation and triboelectric separation) or on their optical characteristics (for example, hyperspectral imaging and laser-induced breakdown spectroscopy) are limited in their ability to differentiate between polymers. This is especially the case for complex materials, such as composites, or materials that have been partially degraded. Therefore, there is a need to develop plastics separation technologies that are unconstrained by the type of waste input, its original application, or the presence of non-plastic contaminants. Once a high-purity feedstock is available, new recycling methods for plastics, such as chemical depolymerization processes, will be easier to implement (Rahimi & García, 2017).

New approach

Despite the fact that for decades plastics have been designed centered in its durability, versatility and low cost, the present constraints of recycling methods have demand other solutions since the polymer design stage. A new trend in sustainable development is based on the *designing to enhance recyclability.* A novel example is seen in last advancements in chemical recycling. This shows it is possible to design copolymer structures fully chemically recyclable with excellent barrier and mechanical properties comparable to petroleum-based PET and superior to biobased PLLA. With specifically designed monomers, reaction conditions can be used to select the direction of the monomer-polymer equilibrium or the closed-loop chemical cycle.

Recently several chemically recyclable polymers have been developed, such as polyesters, polyurethanes, and polycarbonates. Among these materials those based on poly (γ-butyrolactone) (PγBL)[4] core is highly interesting since they are obtained using renewable sources and can be fully recycled back to their monomers. These copolymers represent a promising class of bioplastics that could be implemented in food packaging applications with a closed loop lifecycle through chemical recyclability, contributing toward the overarching goal of eliminating or diminishing plastic pollution (Sangroniz et al., 2019).

[4]Biobased poly(γ-butyrolactone) (PγBL) is a fully biodegradable and bioabsorbable biomaterial that has shown superior properties compared to those of other aliphatic polyesters.

A transition to a sustainable plastics system requires not only a shift to fossil-free feedstock and energy to produce the carbon-neutral building blocks for polymers used in plastics, but also a rational design of the polymers with both desired material properties for functionality and features facilitating their recyclability (Hatti-Kaul et al., 2020).

The current low recyclable polymers rates open a great room for improvement and highlight the potential of the plastic recycling industry to contribute significantly to the global economy. Many opportunities exist for the design of reusable polymeric materials, and studies on dynamic covalent materials, self-immolative polymers and vitrimer networks will inform the design of new recyclable constructs.

Because the recycling of plastics is largely motivated by economic factors, new inexpensive materials designed with end-of-life recycling in mind have proved attractive, so long as the depolymerization conditions includes catalysts that are easily separated after use. These products and processes must be further developed to make renewable technologies more competitive, considering the fluctuating nature of oil prices (Rahimi & García, 2017).

The development of new methods and materials that meet economic and sustainable requirements will result in global savings of billions of dollars, minimizing the dependence on non-renewable petrochemicals for plastics production.

3.4 Standards and regulations

Since the 1980s, the Plastics Industry Association (PLASTICS)[5] created some codes to identify the most used plastics known as Resin Identification Codes (RIC) to help develop consistency in manufacturing and recycling processes). RIC is also known as SPI[6] code (Matthews et al., 2021). Table 9 shows the plastics codes and their food packaging specific applications.

[5] Plastics Industry Association (PLASTICS). Is a purpose-driven organization that supports the entire plastics supply chain. https://www.plasticsindustry.org/

[6] Society of the Plastics Industry, Inc. (SPI), due to the first name of Plastics Industry Association (PLASTICS)

Table 9. *Plastics Identification Codes and its Principal Food Packaging Applications*

Source. (Matthews et al., 2021)

Nevertheless, the limit information given by these codes have raised concern in the principal sector parts and organizations. The Ellen MacArthur Foundation[7] express the need to develop a Global Plastics Protocol to substantially improve the collection, sorting, reprocessing yields, quality, and economics. The Global Plastics Protocol would investigate the possibilities and economic benefits "of harmonizing the labeling and chemical marking across plastic packaging and aligning these standards with after-use separation and sorting systems". A survey made by European plastic converters shows 76% of European plastic converters stated that improvement of collection and sorting of plastic waste was the most suitable way to increase the quality of Recycled Plastic Material (rPM). Respondents also stated that "to improve the quality of recycling, joint action of the industry is needed in addition to the development of quality standards and chemical recycling" (Matthews et al., 2021).

[7]Ellen MacArthur Foundation. Is a UK registered charity which promotes the circular economy. https://www.ellenmacarthurfoundation.org/

Two of the most important regulatory frameworks for recyclable plastics as materials for food packaging applications are release by the European Union and the United States of America (Briassoulis et al., 2019):

EU: Commission Regulation (EU) 2015/1906

Commission Regulation (EU) 2015/1906 of October of 22 2015 amending Regulation (EC) No 282/2008 on recycled plastics materials and articles intended to come into contact with foods.

US: FDA Guidance for Industry (FDA, 2006)

Use of Recycled Plastics in Food Packaging (Chemistry Considerations). Recycled plastics food-contact materials must meet the same regulatory requirements that FDA sets for virgin plastics materials.

Considering the standards available for mechanical and chemical recycling currently in order, can be cited next ones (Briassoulis et al., 2019)

- ISO 18604: 2013 Packaging and the environment-Material recycling
- EN 15343: 2007 Plastics. Recycled plastics. Plastics recycling traceability and assessment of conformity and recycled content.
- EN 15342: 2007 Plastics. Recycled plastics. Characterization of polystyrene (PS) recyclates.
- EN 15344: 2007 Plastics. Recycled plastics. Characterization of polyethylene (PE) recyclates.
- EN 15345: 2007 Plastics. Recycled plastics. Characterization of polypropylene (PP) recyclates.
- EN 15346: 2007 Plastics. Recycled plastics. Characterization of poly(vinylchloride)(PVC) recyclates.
- EN 15348: 2007 Plastics. Recycled plastics. Characterization of poly (ethylene terephthalate) (PET) recyclates.

Recycled plastics in food contact materials (FCM)

In 2017, more than nine years after Regulation 282/ 2008 was adopted, Plastics Recyclers Europe[8] raised concern about the lack of progress made in authorizing plastic recycling processes. Evira-the Finnish Food Authority[9] notes that it is not currently possible to use mechanically recycled plastic as a food contact material except behind a barrier. In 2018, about 140 applicants (most concerning PET recycling) were waiting for authorization from the European Commission.

[8]Plastics Recyclers Europe (PRE) is an organization representing the voice of the European plastics recyclers who reprocess plastic waste into high-quality material destined to produce new articles https://www.plasticsrecyclers.e

[9]Evira. Finnish Food Safety Authority is a centralized body operating under the Ministry of Agriculture and Forestry in Finland. https://www.ruokavirasto.fi/

However, the same year at the 6th meeting of the Scientific Network of the food ingredients and food packaging (FIP) Unit on (FCM[10]), the European Commission revealed that adoption and application of the recycling processes is planned for early 2019 (Matthews et al., 2021).

Despite EFSA[11] issuing over 140 favorable scientific opinions on the safety of plastic recycling processes, the European Commission has not authorized any of the processes. Safe Food Advocacy Europe[12] (SAFE) describes some of the issues associated with using recycled plastics as FCM. For recycled plastics, the levels of oligomers can migrate into food and non-identified contaminants are higher than virgin plastics. Many plastics absorb chemicals through cross contamination during waste management because there is no segregation of food contact plastic and non-food grade plastic. Therefore, SAFE explains the risk of exposure to toxic substances and possibly banned chemicals is much higher with recycled old plastics.

Nevertheless, to characterize the risks from these substances comprehensive information on all chemicals involved is needed. This is one of the principal reasons why recycling rates of post-consumer plastic packaging remains low despite plastic packaging being theoretically highly recyclable. Nonetheless, using recycled plastic for FCM may lead to greater levels of these possibly hazardous chemicals, which in turn can migrate into the food. It is imperative to adequately assess the safety of recycled packaging due to the association between the exposures of certain chemicals migrating from food packaging with chronic disease. Plastics Recyclers Europe PET working group contends that the years of delay in authorizing recycled plastic for FCM have led to uncertainty leaving businesses in legislative no-man's land, which reduces investment and more seriously a conceivable skepticism of legislation regarding FCM. However, recycled material could be utilized in food packaging behind a layer manufactured from virgin materials, increasing the use of recycled materials without exceeding chemical migration levels.

3.5 Fossil-based and recyclable plastics

Only seven types of fossil-based polymers cover approximately two-thirds of the total plastics demand in food packaging applications and these are: Low Density Polyethylene (LDPE), Low Linear Density Polyethylene (LLDPE), High Density Polyethylene (HDPE), Polypropylene (PP), Polystyrene (PS), Polyvinyl chloride (PVC) and polyethylene terephthalate (PET) (Siracusa & Blanco, 2020). As illustrated in table 10, nowadays, the largest groups in plastics production are PE (~ 36%), PP (~ 21%), and PVC (~ 12%), followed by PET, PUR, and PS (<10% each). Together, these seven groups account for 92% of all plastics ever made (Geyer et al., 2017; Sangroniz et al., 2019).

Recycled polymers could be significantly cheaper than virgin materials, with the monetary savings mostly arising from the energy savings, which typically fall in the ~40–90% range depending on the polymer type.

[10] 6th meeting of the Scientific Network of the food ingredients and food packaging (FIP) Unit on (FCM). https://www.efsa.europa.eu/sites/default/files/event/180710-m.pdf

[11] Scientific Network of the Food Contact Materials (FCM). FCM is a platform for cooperation on risk assessment activities and approaches of mutual interest to EFSA and Member States. The Network enhances collaboration between scientists involved in risk assessment of food contact materials to support and harmonize risk assessment practices in this area. https://www.efsa.europa.eu/en/food-ingredients-and-packaging/networks

[12] Safe Food Advocacy Europe (SAFE) is a non-profit independent organization based in Brussels whose goal is to ensure that consumers› health and concerns stay at the core of EU food legislation. https://www.safefoodadvocacy.eu/

Financial savings associated with recycling plastics can be substantial and depend on the grade and type of the recycled material and on the cost of the virgin material, which itself is dependent on oil prices that fluctuate and can often be fairly low (Rahimi & García, 2017).

Plastics that have higher recovery rates[13] are not necessarily cheaper than those less frequently recycled. According to a 2015 report from the United States Environmental Protection Agency (EPA[14]) (table 10), the plastics with the highest recovery rates are polyethylene terephthalate (PET, SPI code 1, 19.5%), high-density polyethylene (HDPE, SPI code 2, 10%) and low-density polyethylene (LDPE, SPI code 4, 5%). All other plastics, including polypropylene (PP, SPI code 5) and polystyrene (PS, SPI code 6), were recovered in less than 1%. The recovery rate for polyvinyl chloride (PVC, SPI code 3) was effectively zero. These values are averaged over the many products that end up in municipal solid waste (MSW), and the relative recovery rates for specific products may be higher; PET bottles, for example, are recovered at ~31% (Rahimi & García, 2017).

Table 10. *Principal Conventional Plastics, Demand and Recovery Rate*

		Demand	Currently recyclable?	Recovery Rate (%)
01	**PET** Polyester	7.4%	yes	19.5%
02	**HDPE** High-Density Polyethylene	12.3%	yes	10%
03	**PVC** Polyvinyl Chloride	10.2%	No	0%
04	**LDPE** Low-Density Polyethylene	17.5%	mostly no	5%
05	**PP** Polypropylene	19.3%	sometimes	1%
06	**PS** Polystyrene	6.6%	sometimes	1%
07	**other**	26.7%	No	varies

Source. (Rahimi & García, 2017; Matthews et al., 2021)

[13]The fraction of the total PSW on MSW is referred to as the recovery rate.

[14]United States Environmental Protection Agency (EPA) is an independent executive agency of the United States federal government tasked with environmental protection matters. https://www.epa.gov/

SPI code 1

Polyethylene terephthalate (PET) is a tough and moldable plastic used extensively in food packaging production, especially in beverage bottles, even though can be found in microwaveable trays. The ductility of PET during mechanical recycling drops from ~310 to ~218% after one cycle and is 2.9% by the third cycle. As a result, only a small portion of PET is recycled for its original application, with most (50–77%) being converted into fibers used to produce mixed materials such as carpeting (Rahimi & García, 2017).

The lower molecular weight recycled PET is used for fiber production. The comparatively high thermal stability of polyethylenes allows HDPE and LDPE to be processed through several melt and remold cycles (Hatti-Kaul et al., 2020).

SPI codes 2, 4 and 5

High-density polyethylene, low-density polyethylene, and polypropylene. The approaches for HDPE, LDPE, and PP are similar to each other. HDPE finds use in bottles and films, with LDPE being a main constituent of food packaging, and plastic wrap. The recycling of polyethylene is not adversely affected by the presence of other materials - indeed, additive or blend formulations can undergo recycling more cleanly than the parent material. The comparatively high thermal stability of polyethylene allows it to undergo multiple melt-and-remold cycles in mechanical recycling processes. For example, LDPE can be extruded up to 100 times at 240°C, although long-term performance suffers after 40 extrusions, with significant changes in processability and mechanical properties observed (Rahimi & García, 2017). HDPE polymers are often used as rigid containers, films or layers for dry food. While LDPE are usually used to manufacture films for wrapping films, carrier bags. Also, can be used to produce bottles.

SPI code 3

Polyvinyl chloride (PVC) is an inexpensive, high-performance, durable polymer that is apply in food packaging applications such as wrapping films, bottles, trays, and containers (Rahimi & García, 2017).

SPI code 6

Polystyrene (PS) is inexpensive, durable, and chemically inert, such that it sees use in many products including food services and packaging. The low recovery rate was partially a result of the difficulties associated with the separation of the waste. Approximately 10% of polystyrene is in the form of expanded polystyrene (EPS) foam, and only 50% of polystyrene produced is used in its pure form, with the remainder being blended with other materials or used as segments in copolymers. The diversity of polystyrene materials complicates their sorting. For example, the density of polystyrene products can vary from 1.04 g/cm^3 to much lower values in the range of ~0.016–0.64g/cm^3 for EPS in particular (Rahimi & García, 2017). Some PS applications in food packaging are usually found as disposable cups, plates and trays, egg trays; and in rigid containers such as yogurt cups. Whereas EPS is usually used for cool boxes and trays due to their insolation properties.

SPI code 7

SPI code 7 is the collection of "other" category that includes polyurethane, polyurea, polycarbonate, biopolymers, nylon, polymethyl methacrylate (PMMA), high-performance thermoplastics such as polyether sulfone (PES) and thermosets such as epoxies (Rahimi & García, 2017). In this subgroup can be found some food packaging applications of the six first groups due to its variety and polyblends.

Table 11 shows the most important mechanical and barrier properties of the principal fossil-based polymers used in the manufacturing of food packaging applications.

Table 11. *Physical Properties of Fossil-based Polymers Applied in Food Packaging*

	Thermal		Mechanical			Permeability		
Polymer	**Tg (°C)**	**Tm (°C)**	**T (MPa)**	**eb (%)**	**OP**	**H2O: WVTR ($g/m^2/day$)**	**O2 ($g/m^2/day$)**	**CO2 (ml μm m^{-2} day^{-1} atm^{-1})**
PET	70-87	243-268	48-72	20-300	++	15-20	100-150	300-600
HDPE	-125 to -90	135	22-31	100>=1000	+++	7-10	1600-2000	12,000-14,000
PVC	60-100	n.d	40-51	40-75	++	0.5-1-0	2-4	400-10,000
LDPE	-125 to -100	112-135	8-31	200-900	++	10-20	6500-8500	20,000-40,000
PP	-10	167-177	31-41	100-600	+	10-12	3500-4500	10,000-14,000
PS	100	n.d	35-51	1-4	++	-	4500-6000	14,000-30,000
PA	50-60	220	40-52	5-10	++	300-400	50-75	n.d
PVA	70-75	215-220	25-30	220-250	++	n.d	n.d	n.d
EVOH	60-65	180-150	45-110	180-250	+++	1000	0.5	n.d

Source. (Rahimi & García, 2017)

The design of next-generation recyclable polymers is motivated by the production of materials for specific applications. The market demands high performance and convenient network with the right conditions at the end of life (Rahimi & García, 2017).

3.6 Bio-based and recyclable bioplastics

This section comprises those polymers which have a renewable origin and are analogous to conventional petroleum-derived polymers. Even though these plastics are biobased and can reduce greenhouse emissions in bioplastic production, their bonding is identical to their petrochemical versions, and so they are not biodegradable (Narancic et al., 2020). These suitable bioplastics for food packaging are part of class 3 of biobased polymers (figure 26) as explained in section 2.4.1.

An increasing trend in recent years in polymers solutions for food packaging has been to replace partly or wholly the fossil-based building blocks of conventional plastics by identical molecules of renewable origin, the so called "drop-ins", such as bio-PE produced from bioethanol, bio-PET made using biobased ethylene glycol as the monomer, and bio-PTT with biobased 1,3-propaneidol (1,3-PDO) and bio-PP. Being chemically identical to their oil-based counterparts, these biobased "greener" alternatives can be processed using the existing infrastructure and made available to users already familiar with their performance and applications. Furthermore, these biobased equivalents of popular fossil-based plastics can be recycled in current recycling schemes even though the recycling rate is still very low (Hatti-Kaul et al., 2020; Narancic et al., 2020).

The production of plastics traditionally derived from petrochemical sources such as PET and PE, from natural resources, has garnered interest from plastic manufacturers, as the same processing equipment can be used in its manufacturing. Which helps in lowering total investment in infrastructure for their manufacture (Narancic et al., 2020).

3.6.1 Bio-PE

Around 1970's, due to climbing oil prices, bio ethanol attracted the fuel industry and the benefits of its derivates (Nakajima et al., 2017). Bio-PE is made from ethylene monomer derived from dehydration of ethanol, which can be made by fermentation of various animal and plant-based feedstocks including molasses, corn, sugarcane, sugar beet and wheat grain (Robertson, 2012; Andrady, 2015). For instance, a hectare of sugarcane land yields about three tons of bio-PE. Both ethylene and ethylene glycol (EG) can also be produced by cracking bionaphtha. Bio-PE has characteristics equivalent to those of conventional polyethylene and can be used in identical applications. From bioethylene it is possible to produce all the polyethylene types: HDPE, LDPE and LLDPE (Robertson, 2012).

The big advantage of bio-PE is the fact that its properties are close to fossil-based PE, which has a complete infrastructure for processing and recycling. However, it faces direct competition with fossil-based feedstock, the price of which heavily fluctuates (e.g., shale gas is cheap). The downside of biobased PE is that it is not biodegradable (Nakajima et al., 2017).

Associated with fossil fuel resource conservation, the use of bio-based feedstock also results in substantial reductions in carbon emissions. For instance, sugarcane-derived PP has a net negative carbon emission (or a sequestration) of 2.3 kg of CO2/kg per bio-PP produced. Reliance on agricultural biomass as a raw material is the main limiting factor for this technology. However, improving the process to use lignocellulosic waste materials can address this problem (Andrady, 2015).

The two largest bioethanol from sugarcane producers globally are based in Brazil. They have formed joint ventures to produce bio-PE: Braskem (Nakajima et al., 2017; Narancic et al., 2020) with Toyota Tsusho Corporation and Dow Chemical Company with Crystalsev. Given that PE is the most common polymer used for food packaging, it is expected that many companies will switch to bio-PE to lower their carbon footprints and to align to more sustainable production (Robertson, 2012).

Bio-PE-Based Blends

A limited number of papers discussed the blending of bio-based polyolefins, and only one example was found with bio-based PLA. As reported have been investigated the effect of ethylene-glycidyl methacrylate (E-GMA) and ethylene-methyl acrylate-glycidyl methacrylate (EMA-GMA) copolymers as compatibilizer agents in PLA/Bio-PE blend. The data underlined that the use of the E-GMA and EMA-GMA copolymers significantly enhanced the impact strength of the PLA/Bio-PE blend, thanks to the reaction between hydroxyl or carboxyl groups in PLA and the epoxy groups in the copolymer matrices (Luzi et al., 2019).

3.6.2 Bio-PET

Polyester is made by polycondensation of ethylene glycol (EG) with terephthalic acid (TPA). Bio-based EG is made from molasses, sugarcane, switchgrass, and bagasse via their fermentation into ethanol. Using this bio-EG with fossil fuel-based TPA, a PET that is partially bio-based is obtained (Andrady, 2015). Nonetheless, biobased terephthalic acid (bio-TPA) is being developed to further improve the sustainability of PET, as bio-TPA is produced from naturally derived sustainable biomass feedstock. Due to, theoretically, combining bio-EG and bio-TPA could achieve 100% natural biomass feedstock derived bio-PET (Nakajima et al., 2017).
PET plays an important role in the plastic market and food packaging applications. But its poor degradability makes recycling technology the most proper circular path toward a sustainable life approach (Andrady, 2015; Nakajima et al., 2017). Even though PET is the most widely recycled plastic (Tg∼74°C) undergoes a loss of molecular mass during recycling (Hatti-Kaul et al., 2020). In fact, polymer-to-polymer material recycling of PET has been launched in some fields, but it is always accompanied by non-negligible deterioration of the polymer's physical properties in the final recycled products. This occurs due to side-reactions and thermal degradation, hydrolysis, and thermo-oxidative degradation during recycling. Ways to chemically recycle PET are under development, but many technical difficulties, such as the high stability of PET under normal hydrolysis, alcoholysis, or breakdown processes, must be overcome.

Despite these shortcomings, some pioneering companies in the beverage industry have launched successful sustainable bio-PET solutions for its products. The Coca-Cola Company (TCCC), which has shared its *know how* about bio-PET soda bottles with Heinz (ketchup industrial producer), and PepsiCo are part of the most representatives.

The Coca-Cola Company (TCCC) has accelerated the production of bio-PET known as "PlantBottle". PlantBottle, which was launched in 2009, consists of 30% biobased materials, 100% biobased EG (bio-EG) and petroleum-derived terephthalic acid (TPA). These bio-PET bottles are recyclable and can be processed without a variance from regular PET performs the same in products. Moreover, these new bio-PET bottles reduce carbon emissions by 8-11% according to the manufacturer, compared to the fossil fuel-based PET. (Andrady, 2015; Nakajima et al., 2017; Narancic et al., 2020). Nevertheless, the bio-EG price is yet significantly superior respect to its traditional equivalent based one. Therefore, its market diffusion is still limited (Luzi et al., 2019).

A fully bio-based polyester beverage bottle was subsequently developed by PepsiCo, which use TPA bio-derived intermediates such as bio-xylene or from bio-derived 5-hydroxymethylfurfural or furandicarboxylic acid (FDCA) from fructose via alkoxymethyl fufural. Efforts to synthesize bio-TPA via the muconic acid route without going via p-xylene are also under developement (Andrady, 2015).

3.6.3 PEF

Newly developed biobased polymer

Another 100% biobased alternative polyester well suited for beverage bottles and likely to be economical compared to PET is poly (ethylene furanoate) (PEF).

The conversion of biomass feedstocks allows us access to not only carbon-neutral alternatives, but also a diversity of novel structures not easily obtained from fossil resources, such as furan-based monomers and isosorbide. Biomass-derived saccharides (such as sucrose) can be converted in alkoxymethyl furfural, which is oxidized into 2,5-Furan dicarboxylic acid (FDCA). The FDCA can be condensed with EG to yield PEF polymer, a new recyclable fully biobased product (Andrady, 2015; Hatti-Kaul et al., 2020).

But at the beginning, Bio-Poly (Ethylene Terephtalic Acid) (PEF) was not considered special; it was a downgraded PET because of its slow crystallization and low Tm. However, in 2008, some reliable information about PEF, including its currently known polymerizations and its widely known thermal properties (Tm around 210 °C and Tg around 80 °C) were reported. Other studies followed, increasing the scientific understanding of the physical properties of PEF. The thermal decomposition temperature of PEF is approximately 300 °C, which also results in β-hydrogen bonds. The brittleness and rigidity of PEF result in about 4% elongation at break (Nakajima et al., 2017).

Poly (ethylene 2,5-furandicarboxylate) (PEF) is currently considered as an attractive sustainable substitute of PET, since it has better barrier and interesting thermal characteristics (e.g., lower melting phenomenon and higher glass transition temperature) than PET. Furthermore, the reduced permeability of PEF to CO_2, O_2, and H_2O is a great benefit for food packaging applications (Andrady, 2015; Luzi et al., 2019, Hatti-Kaul et al., 2020).

The most important properties and functionalities of PEF are compared with those of PET in table 12. The remarkably high gas barrier properties of PEF should be emphasized; the high O_2 barrier is advantageous for packaging, leading to PEF's practical application in the food and beverage industry (Nakajima et al., 2017).

Table 12. *Comparison of the physical properties of PEF and PET*

	PEF	PET
Density (g/cm^3)	1.43	1.36
O_2 permeability	0.0107	0.114
CO_2 permeability	0.026	0.46
Tg (°C)	88	76
Tm (°C)	210-230	250-270
E-modulus (GPa)	3.1-3.3	2.1-2.2
Yield stress (MPa)	90-100	50-60
Quiescent crystallization time (min)	20-30	2-3

Source. (Nakajima et al., 2017).

The excellent thermal properties and superior barrier properties of PEF compared with PET (over six times higher for O_2, and twice as high for CO2 and water) makes it an ideal substituent for the polymer in food packages (Hatti-Kaul et al., 2020). In addition, due to the superior gas barrier properties of PEF, it is being used for bottles, films, and other packaging materials in the food and beverage industry (Nakajima et al., 2017).

The results of scientific studies have been successfully applied to pilot and upcoming industrial production of PEF. The most widely known example of industrial PEF production is that of Synvina, which is based on FDCA derived from fructose (Nakajima et al., 2017).

3.7 Case studies

The state-of-the-art bioplastics and conventional plastics able to be recyclable are listed in next case studies and summary charts. The cases are presented in a chronological order starting from 2021. There are some trademarks or projects still in R&D stage (Robertson, 2012; Andrady, 2015; Nakajima et al., 2017; COWI et al., 2019; Hatti-Kaul et al., 2020).

bio-polyolefins
R&D

INEOS

INEOS will use UPM BioVerno, a sustainable raw material from a renewable residue of wood pulp processing, to produce bio-attributed polyolefins.

INEOS Köln & UPM Biofuels
Germany, since 2019

Material Sustitution
PE, LDPE, LLDPE, PP

References
https://www.ineos.com/news/shared-news/ineos-olefins--polymers-europe-announces-range-of-bio-attributed-olefins-and-polyolefins/

SUSTAINABILITY

Origin
100%
biobased

End-of-life
recyclable
in existing streams per plastic type

Advances

Each step in the supply chain has been fully certified by the Roundtable on Sustainable Biomaterials (RSB) starting from UPM Biofuels converting the wood-based residue (crude tall oil) into hydrocarbons, through to the final polymer. RSB's assessment includes social and environmental impact, as well as demonstrable positive climate benefit.

ADDITIONAL PROPERTIES

coulored

printable

sealable

ENGINEERING PROPERTIES

Density
PP<LDPE<HDPE

Thermal resistance
LDPE<HDPE<PP

Flexibility
LLDPE<LDPE<HDPE<PP

PRESENTATIONS

flexible and rigid packaging

FOODS

perishable and semi-perishable foods

PROCESSABILITY

cast and blow film extrusion
Thermoforming
Injection moulding

Anellotech license

R&D

Suntory has partnered with Anellotech to advance the development and commercialization of cost-competitive aromatics including bio-paraxylene – the key component needed to make bio-based PET. Suntory currently uses 30% bio-based PET for its Suntory Tennensui brand of mineral water and is pursuing the development of a 100% bio-based bottle through this collaboration.

SUNTORY Anellotech

Anellotech & Suntory
USA & Japan, since 2017

Material Sustitution
PET

References
https://anellotech.com/chemicals

SUSTAINABILITY

Origin
100%
biobased

End-of-life
recyclable
in existing PET stream

01

Advances

An initial volume of high-purity bio-paraxylene test samples has been produced. These samples have completely met all of the ASTM International specifications for downstream derivatives in conversion to PET. As larger amounts of paraxylene are purified, Anellotech will begin to make renewable PET resin for prototype bottle manufacture and product trials.

ADDITIONAL PROPERTIES

translucent
color

ENGINEERING PROPERTIES

semi-rigid to rigid

waterproofing
barrier

lightweight

PRESENTATIONS

rigid packaging

FOODS

perishable and semi-perishable foods

PROCESSABILITY

injection molding
blow molding

Synvina

Avantium
Netherlands, since 2017

Material Sustitution
PET

References
https://www.avantium.com/lead-products/#pef

PEF is a 100% plant-based and recyclable. PEF has a powerful combination of environmental features and superior functionality. It shows improved barrier properties for carbon dioxide (CO2) and oxygen, leading to a longer shelf life of packaged food products. Furthermore, PEF has a 50-70% lower carbon footprint than a fossil-based

SUSTAINABILITY

Origin
100%
biobased

End-of-life
Recyclable
in existing PET stream
01

Certifications

PEF is plant-based instead of fossil-based and has a 50-70% lower carbon footprint (source: Copernicus Institute at the University of Utrecht (Eerhart et al. 2012)

DESIGN PROPERTIES

transparent
excellent gloss

printable

ENGINEERING PROPERTIES

PEF's oxygen barrier is 10 times better than PET
PEF's CO2 barrier is 6 to 10 times better than PET
PEF's water barrier is twice as good as PET's

Tg: 86°C compared to PET (74°C)
Tm: 235°C compared to PET (265°C)

PRESENTATIONS

rigid packaging

FOODS

perishable and semi-perishable foods

PROCESSABILITY

injection molding
blow molding

Terralene® LD 2509 CL

FKuR Kunststoff GmbH
Germany, since ~2020

Material Sustitution
PP, PE

References
https://fkur.com/wp-content/uploads/2020/07/Technical-Data-Sheet-Bioplastic-injection-moulding-Terralene-LD-2509-CL-EN.pdf

Terralene® is a tailor-made family of polyethylene compounds based on renewable raw materials (sugar cane). Terralene® LD 2509 CL can be processed on existing production equipment without any adjustments. After its use, Terralene® LD 2509 CL can be placed back into existing polyethylene recycling streams without any detriment to the recyclate.

SUSTAINABILITY

Origin
50%
biobased

End-of-life
recyclable
in existing LDPE stream

04

Certifications

Very good recyclability certified by Interseroh FCM according to EN 10/2011 and US FDA.

DESIGN PROPERTIES

high translucency

printable

sealable

ENGINEERING PROPERTIES

density: 0.909 g/cm³
High barrier properties

self ignition temperatur > 300 °C
melting range: 160 - 165 °C

PRESENTATIONS

flexible and rigid packaging

FOODS

perishable and semi-perishable foods

PROCESSABILITY

Thermoforming
Injection moulding
Film extrusion

FOSSIL-BASED AND RECYCLABLE PLASTICS FOR FOOD PACKAGING APPLICATIONS

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life
Styrenics Circular Solutions (SCS) R&D by INEOS Styrolution Germany 2021	rPS	fossil-based	PS	06 recyclable
PETValue R&D by The Coca-Cola Company & Indorama Ventures Philippines 2020	100% recycled plastic	fossil-based	PET	01 recyclable
Future-PET R1-20 Future-PET N1-100 by Indorama Ventures Thailand-Indonesia ~2018	100% rPET	fossil-based	PET	01 recyclable

General properties	Processing technologies	Applications	References
high purity levels needed by converters, brand-owners and retailers for direct food contact outstanding environmental footprint (LCA results) while maintaining polystyrene's much valued application and processing benefits	blown film extrusion thermoforming injection molding	flexible packaging rigid packaging	http://styrenics-circular-solutions.com/wp-content/uploads/2021/02/20210209_SCS_ChallengeTestSuccess_MR_FoodContact_PS.pdf
lightweight good clarity good barrier against carbon dioxide (CO2) and oxygen (O2)	blow molding	rigid packaging	https://www.indoramaventures.com/en/updates/other-release/1393/coca-cola-and-indorama-ventures-ink-partnership-to-build-phs-largest-bottle-to-bottle-recycling-facility
good clarity melting point: ~250°C dimensional stability that reduces shrinkage and deformation Produced by mechanical recycling	blow molding	rigid packaging	https://www.indoramaventures.com/storage/downloads/product/-pet/bottle-sheet/FuTuRe-PET-R1-%2020.pdf

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life
RM 7004 POST-CONSUMER RPET by Indorama Ventures Netherlands ~2017	100% rPET	fossil-based	PET	01 recyclable

BIO-BASED AND RECYCLABLE BIOPLASTICS FOR FOOD PACKAGING APPLICATIONS

Class 3

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life
Terralene® HD 2508 by FKuR Kunststoff GmbH Germany ~2019	bio-HDPE	75% biobased	HDPE	02 recyclable
Terralene®PP 3509 by FKuR Kunststoff GmbH Germany 2018	PP copolymer/ biobased HDPE	33% bio-based	PP HDPE	02 recyclable

General properties	Processing technologies	Applications	References
Clear and dark blue flakes Density 325 +/- 50 kg/m3 Produced by mechanical recycling	blow molding	rigid packaging	https://www.indoramaventures.com/storage/downloads/product/recycled-product/Wellman-Recycling-product-specifications-RM7004-version-1.pdf
natural color almost odourless self ignition temperatur > 300 °C density: 0.93 g/cm³ Modulus of elasticity: 600 Mpa Tensile strength: 16.5 Mpa	injection moulding	rigid packaging caps and closures	https://fkur.com/wp-content/uploads/2019/03/TD_TERRALENE_HD_2508_EN.pdf
natural color almost odourless self ignition temperatur > 300 °C melting range: 160 - 165 °C density: 0.916 g/cm³ Modulus of elasticity: 1,100 Mpa Tensile strength: 24MPa	injection moulding	rigid packaging caps and closures	https://fkur.com/wp-content/uploads/2019/03/TD_TERRALENE_PP_3509_EN.pdf

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life

Class 3

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life
Terralene® HD 4527 by IFKuR Kunststoff GmbH Germany ~2017	bio-HDPE	95% bio-based	HDPE	02 recyclable
Terralene® PP 3402 by FKuR Kunststoff GmbH Germany 2017	PP copolymer/ biobased HDPE	33% bio-based	PP HDPE	02 recyclable
Terralene® PP 3505 by FKuR Kunststoff GmbH Germany 2017	PP copolymer/ biobased HDPE	33% bio-based	PP HDPE	02 recyclable

General properties	Processing technologies	Applications	References
natural grey almost odourless density: 1.07 g/cm^3 self ignition temperatur > 300 °C melting range: 130 - 145 °C high rigidity (Tensile strength: 17.5MPa)	injection moulding	rigid packaging	https://fkur.com/wp-content/uploads/2018/05/TD_-TERRALENE_HD_4527_EN.pdf
natural color almost odourless density: 0.922 g/cm^3 self ignition temperatur > 300 °C melting range: 160 - 165 °C	blow moulding	rigid packaging	https://fkur.com/wp-content/uploads/2019/03/TD_-TERRALENE_PP_3402_EN.pdf
natural color almost odourless density: 0.912 g/cm^3 self ignition temperatur > 300 °C melting range: 160 - 165 °C Modulus of elasticity: 1,075 Mpa	injection moulding	rigid packaging	https://fkur.com/wp-content/uploads/2019/03/TD_-TERRALENE_PP_3505_EN.pdf

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life

Class 3

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life
Terralene® PP 4732 by IFKuR Kunststoff GmbH Germany ~2017	PP copolymer/ biobased HDPE/mineral fillers	33% bio-based	PP HDPE	02 recyclable
Bio-based bottles (in progress-scaling up) by NaturALL Bottle Alliance Danone, Nestlé Waters, Origin Materials, Pepsico USA 2017	bio-PET	100% bio-based	PET	01 recyclable
BEVERAGE BOTTLES by Suntory, Japan 2016	bio-PET	30% bio-based	PET	01 recyclable

General properties	Processing technologies	Applications	References
white color almost odourless density: 1.100 g/cm^3 self ignition temperatur > 300 °C melting range: 160 - 165 °C Modulus of elasticity: 1,850 Mpa	thermoforming injection moulding	rigid packaging	https://fkur.com/wp-content/uploads/2019/03/TD_-TERRALENE_P-P_4732_EN.pdf
lightweight good clarity Good barrier against carbon dioxide (CO_2) and oxygen (O_2) Modulus of elasticity: 1,075 Mpa	blow molding	rigid packaging	https://www.nestle.com/media/news/naturall-bottle-alliance-welcomes-pepsico
lightweight good clarity Good barrier against carbon dioxide (CO_2) and oxygen (O_2)	blow molding	rigid packaging mineral water bottles 500 - 600 ml	https://www.suntory.com/softdrink/company/sustainability/environment/packagehistory/index.html

Class 3

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life
Terralene® HD3400 by FKuR Kunststoff GmbH Germany 2016	bio-HDPE	>95% bio-based	HDPE	02 recyclable
Eastlon PET CB-602AB by IFKuR Kunststoff GmbH Germany ~2016	bio-PET	20% bio-based	PET	01 recyclable
I'M GREEN™ PE GREEN HDPE SHE 150 by Braskem, Brazil 2010	bio-HDPE	94% bio-based	HDPE	02 recyclable

General properties	Processing technologies	Applications	References
natural color almost odourless density: 0.956 g/cm³ self ignition temperatur > 300 °C	blow moulding	rigid packaging	https://fkur.com/wp-content/uploads/2019/03/MSDS_TERRALENE_HD_3400_EN.pdf
high transparency almost odourless density: 1.3-1.4 g/cm³ Melting range: 240 - 260 °C Good barrier against carbon dioxide (CO2) and oxygen (O2) comparable to PET	blown film extrusion blow molding	flexible packaging rigid packaging	https://fkur.com/wp-content/uploads/2017/01/TD_BIO-PET-Eastlon-CB-602AB_en.pdf
white almost odourless density: 0.948 g/cm³ self ignition temperatur 350 °C	blown film extrusion fibre extrusion	flexible packaging rigid packaging	https://fkur.com/wp-content/uploads/2016/10/Technical-Data-Sheet-Green-HDPE-SHE150-ASTM.pdf

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life

Class 3

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life
I'M GREEN™ PE GREEN LDPE SPB 208 GREEN LDPE SPB 608 by Braskem, Brazil 2010	bio-HDPE	87-95% bio-based	HDPE	02 recyclable
I'm green™ PE Green HDPE SGM 9450 F Green LDPE SBF 0323 HC Green LLDPE SLH 0820/30 AF Green LLDPE SLH 118 Green LLDPE SLL 118 by Braskem, Brazil 2010	bio-HDPE bio-LDPE bio-LLDPE	bio-based	HDPE LDPE LLDPE	02 04 recyclable
I'm green™ PE Green HDPE SGE 7252 Green HDPE SHA 7260 Green HDPE SHC 7260 by Braskem, Brazil 2010	bio-HDPE	94-96% bio-based	HDPE	02 recyclable

General properties	Processing technologies	Applications	References
white almost odourless density: 0.948 g/cm³ self ignition temperatur 350 °C	injection moulding	rigid packaging caps for non-carbonated or low carbonated so drinks	https://fkur.com/en/applications/-caps-closres/
high transparency elegant surface gloss excellently colored For printing, solvent-containing or water-based printing inks can be used. flexible and highly stretchable (wall thickness down to 8 µm)	blown film extrusion fibre extrusion	flexible packaging	https://fkur.com/en/applications/bags/
high transparency almost odourless density: 1.3-1.4 g/cm³ Melting range: 240 - 260 °C Good barrier against H_2O, CO_2, O_2 comparable to PET	injection moulding	rigid packaging caps for non-carbonated or low carbonated so drinks	https://fkur.com/en/applications/-caps-closres/

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life

Class 3

I'm green™ PE Green LDPE STN 7006 Green LLDPE SLH 0820/30 AF by Braskem, Brazil 2010	bio-LDPE bio/LLDPE	84-95% bio-based	LDPE LLDPE	04 recyclable
BioFoam® by Synbra Netherlands 2010	bio-Expanded PS (EPS)	bio-based	EPS	06 Recyclable Biodegradable reusable

General properties	Processing technologies	Applications	References
density: 0.910–0.940 g/cm3 tensile strength: LDPE<LLDPE higher impact: LDPE<LLDPE flexibility: LDPE<LLDPE Good barrier against H_2O, CO_2, O_2	blown film extrusion blow molding extrusion coating	flexible packaging	https://fkur.com/en/applications/waste-bags/
lightweight Retains its shape High shock absorption Excellent insulator Insensitive to moisture Complete three-dimensional freedom of design	thermoforming	rigid packaging	https://www.synprodo.com/what-is-bio-foam/

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Chapter IV.

BIOPLASTICS FOR PAPER COATINGS

4.1 Definition and classification

The increasing environmental concerns, the fluctuation of the finite petrochemical resources, and the consumer demand for greener food packaging play an important role in the food industry. These primary reasons have triggered the research and innovation of sustainable alternatives for packaging types and presentations including coatings (Attaran et al., 2017; Nechita & Roman, 2020).

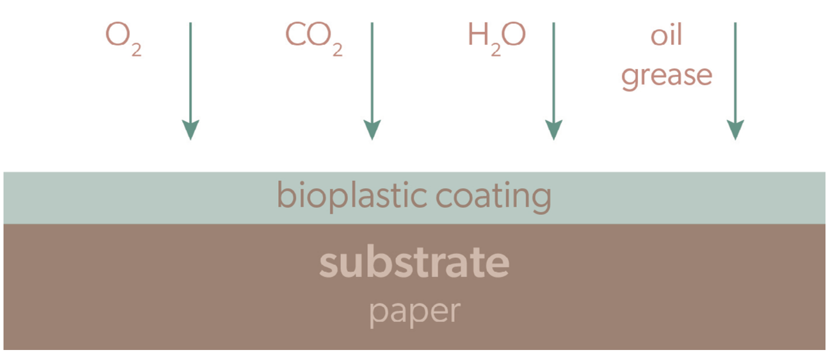
Bioplastic's coating represent a promising solution for the current use of synthetic coatings. Biobased coatings, for instance, can not only avoid the use of petrochemical resources; with a lower carbon footprint in its production but can also provide new combinations of properties.

Moreover, latest studies (Fotie et al., 2020) also demonstrate that bioplastics used as coatings are a sustainable alternative for multilayer food packaging solutions due to its option of compostability or recyclability. Common multilayer flexible packaging is generally not recyclable due to its difficult separation, which also demand additional costs.

Coating is a efficient method and application to ensure a suitable shelf life for food products. They are used to fulfill various functions to preserve a proper food quality state and the package properties.

Even though films and coatings are applications for encapsulating different foods to preserve their properties and fulfill the same role, they represent different concepts. Films are prefabricated by various methods and are used to embed food, while coatings can be applied on other packaging materials or the food surface using a viscous liquid (Avramescu et al., 2020). Therefore, they have different thicknesses, films used to be thicker than coatings. Sometimes, films and coatings are considered as synonyms or replaceable terms, but to avoid misunderstandings, in this study, coatings will be considered the secondary material applied as a thin coverture on the surface of the primary packaging material as it is illustrated by figure 33. The primary material is usually called substrate (Fotie et al., 2020).

Figure 33



Representation of coating on food packages

Source. (Fotie et al., 2020)

This secondary material can function as packaging materials and are currently employed in food packaging. Still, due to the attention on environmentally friendly alternatives for reliable and sustainable coating products, paper appears as the ideal packaging material.

Paper is an eco-friendly material that has the advantages of high recyclability, biodegradability and compostability from renewable raw material, compared with petroleum-based packaging (Nechita & Roman, 2020; Avramescu et al., 2020).

Paper was often used for the packaging of fluids and greasy foods since the 80s. But during the 1970s–1980s, when plastics were introduced into food packaging, paper-based materials lost their importance and ended up being replaced in many of its uses (Nechita & Roman, 2020).

Paper and paperboard show many advantages as packaging materials. And yet, current technologies employed synthetic polymers coating (e.g., ethylene vinyl alcohol (EVOH), polyvinylidene chloride (PVDC), polyethylene (PE), acrylic, latex, or fluorocarbon) to obtain adequate barrier properties. Inorganic layers are also sometimes laminated such as aluminum foil or deposited by metallization. Also, although combinations of these materials are often employed, such coated and laminated, paper solutions are usually not recyclable anymore (Nechita & Roman, 2020; Coltelli et al., 2016).

Nevertheless, due to its porous structure and the hydrophilic character of cellulose fibers, paper has inherently inferior barrier properties (i.e., low water and grease resistance, high permeability to gases and water vapors), and is sensitive to microbial attack. Therefore, to fulfill food protection requirements, packaging paper should enhance its barrier properties (i.e., against oxygen, carbon dioxide, moisture, water, micro-organisms, grease, or aroma) (Nechita & Roman, 2020).

Favorably, latest studies and scientific advances show that bioplastics can provide new combinations in composites for coatings formulas to meet barrier properties (low oxygen and water vapor permeability) and specific functionalities for a fully protective food packaging (Nechita & Roman, 2020).

Due to all these reasons, this section will specialize in bioplastic's coating on paper packaging. Moreover, using bioplastic's coating for paper packaging means a reduction of solid waste amount because of the biodegradability properties of both components (Avramescu et al., 2020).

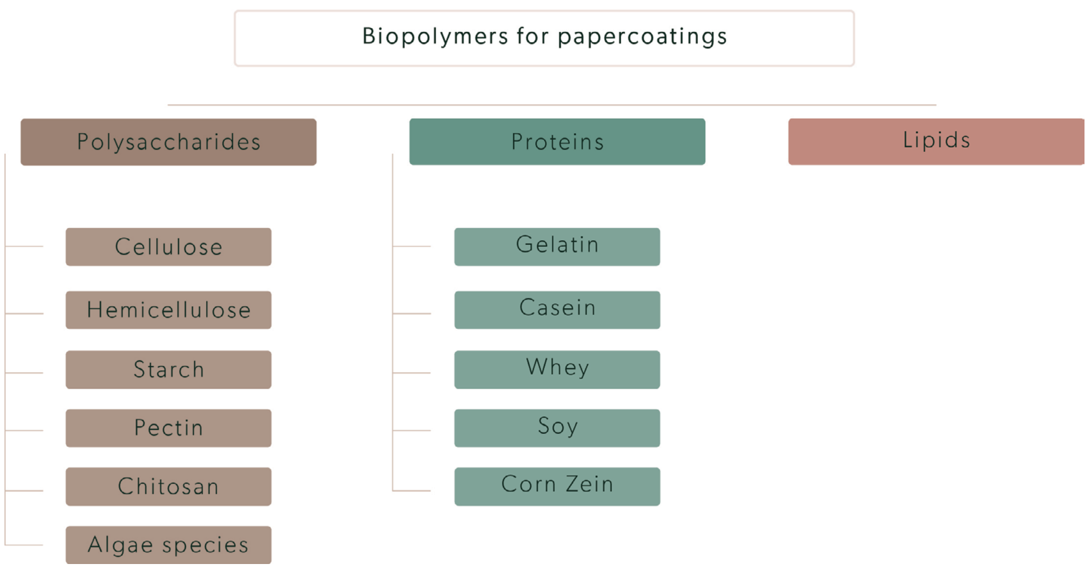
As has been seen in chapter II (figure 26), biolastics can be subdivided into 3 classes. Among them, class 1 bioplastics group is the most suitable for coatings applications, including paper coatings.

• Class 1, known as *natural derived biomass polymers* consider polysaccharides and those from protein groups. Most of the commonly available class 1 bioplastics are extracted from marine and agricultural products. They can be used alone or in blends with other biodegradable plastics such as PCL or biobased and biodegradable polyesters such as PLA. The high crystallinity and strong intermolecular interactions of class 1 bioplastics lead to their thermal degradation before achieving melt flow. Therefore, a combination of heat, mechanical shear and suitable plasticizers is necessary (Robertson, 2012).

• Class 2, called *bio-engineered* polymers in polymer science includes biobased polymers derived from Bacterial Cellulose (BC). BC is another source of nanocellulose secreted by specific bacteria (strains of Gluconacetobacter, Komagataeibacter, tea fungus) and have a width of about 3.5 nm and promising physical and mechanical properties such as high elastic modulus, high specific surface area, and high purity (Robertson, 2012; Nechita & Roman, 2020). Despite the suitable properties of BC for food packaging, most studies were focused on its properties for medicine and further studies are needed to evaluate its performance as barrier coating or film for food packages (Cazón & Vázquez, 2021). Additionally, its high production cost is usually considered as a limiting factor for food packaging applications (Azeredo et al., 2019). Based on these reasons there will not be any more details on BC.

Lipids are considered also a natural resource for bioplastics-based packaging materials, and due its properties are also contemplated in this section (Khwaldia et al., 2010). Figure 34 illustrate bioplastic's classification for paper and paper board food packaging applications.

Figure 34



Classification of Bioplastics suitable for Paper Coatings in Food Packaging
Note. Author graphic representation based on the source investigated.
Source. (Khwaldia et al., 2010; Avramescu et al., 202).

The choice of materials for a coating is mainly dependent on its desired function. Plasticizers, regular paper pigments, antioxidants, or antimicrobial agents can be added to paper coating solutions to improve its performance properties (Khwaldia et al., 2010; Avramescu et al., 2020).

Biobased polymers can be applied to paper or paperboard through different coating techniques, such as surface sizing, solution coating, compression molding, and curtain coating depending on the appropriate coating material and type of paper used (Khwaldia et al., 2010).

Surface sizing is one of the most frequently used processes for applying an aqueous coating to a paper substrate. In surface sizing, the coating's solid content is limited and is typically lower than 10% to 15%. A low solid content does not yield a fully continuous coating and increases the amount of drying needed. Meanwhile, a higher coating weight and better gas-barrier properties can be obtained by using curtain-coating technique in which the paper industry has begun to show considerable interest. A thick and continuous coating, necessary in several cases to obtain coverage of the paper, cannot be obtained by solution coating. However, this coating technique results in interesting mechanical properties (Khwaldia et al., 2010).

The compression-molding technique is suitable for applications where complete coverage and thick coatings were necessary, and which, therefore, involved significantly more coating material compared to solution coating.

As seen in the next figure 35, paper coatings are influenced by many factors, which in sum build its functionality in food packaging.

Figure 35

Factors affecting the properties of different types of paper coatings
Source. (Li et al., 2020)

This chapter will contemplate the most suitable bioplastics and their latest updates to be used as paper coatings considering its ability to act as a functional barrier to remain safe food content and ensure packaging functions. Furthermore, is presented novel cases in R&D stage or currently in commerce.

4.2 Polysaccharides-based coatings

Polysaccharides are carbohydrate polymers consisting of repeating units of monosaccharides (hundreds and thousands) linked by glycosidic bonds and formed by the condensation of monosaccharide residues through hemiacetal or hemiketal linkages. The polysaccharides can be originated from higher photosynthetic plants, marine biomass, bacteria, or fungi (Nechita & Roman, 2020). In food packaging, polysaccharides can be used as coating formulations for paper, edible coatings, and films or to obtain bioactive and sensor materials in active and intelligent packaging.

These compounds are highly available in nature and therefore biocompatible with natural substances (Avramescu et al., 2020). According to the literature, polysaccharides are the main candidates to substitute oil-based polymers in food paper coating due to their particular properties.

Polysaccharides are biodegradable and non-toxic, have film forming ability and good affinity for paper substrate. They can also provide a very good barrier to gases, aromas, and lipids, and serve as a bioplastic matrix for the incorporation of active agents for paper functionalization. Furthermore, polysaccharides biobased polymers have positive effects on mechanical strength (Nechita & Roman, 2020; Li et al., 2020).

Nonetheless, the polysaccharide's main disadvantage is their sensitivity to moisture that limits their large-scale utilization in barrier coatings for paper. The presented section shows that there has been intensive research regarding the chemical modification of polysaccharides. To introduce hydrophobic groups in their structure to improve water resistance and rheological properties[1] when used in barrier coatings for packaging paper applications (Nechita & Roman, 2020).

Therefore, finding an appropriate route for chemical modification and the right combination of polysaccharides with other bioplastics (e.g nanofibers or nanofillers) will generate interest in developing and applying these bioplastics in composite coatings for food packaging papers (Nechita & Roman, 2020).

Additionally, paper's raw material is mainly plant fibers, which contain mostly polysaccharides, i.e., cellulose. The hydroxyls of polysaccharide can link with the hydroxyls of paper fibers through hydrogen bonding, resulting in high affinity of polysaccharides to paper surface.

The long chain structure enables their molecules to easily connect forming a mechanically reinforced network. Also, some polysaccharides have amphiphilic chemical structures which contribute to good dispersing ability to inorganic fillers, thus the fillers can be dispersed and adhered to the paper surface more uniformly, improving the coating performance.

However, when used in coatings or films for food packaging, polysaccharides present some drawbacks. Polysaccharides do not behave well as a moisture barrier because of their natural hydrophilicity and crystalline structure (i.e., low water resistance, low barrier to water vapors, and properties dependent on the environment humidity). The insufficient mechanical property of pure polysaccharide coating layers also limits their application in pigment and functional coatings, but the properties of polysaccharides can be improved via physical or chemical methods to meet the requirements of applications in paper coatings. The large active hydroxyl groups of polysaccharides make immediate chemical modifications (Li et al., 2020).

Moreover, other bioplastics and environmentally friendly nanomaterials are combined with polysaccharides to prepare novel formulations with desired properties for food packaging, including coatings for paper/board packaging. Sometimes it is necessary to combine of more than two packaging materials to provide the best packaging solution for certain food products (Nechita & Roman, 2020).

[1]Rheology is the study of the flow of matter, primarily in a liquid or gas state, but also as "soft solids" or solids under conditions in which they respond with plastic flow rather than deforming elastically in response to an applied force

The most tested and applied polysaccharides as coatings on paper for foods packaging, are presented below.

Polysaccharides from Wood and Lignocellulosic Plants

4.2.1 Cellulose

Cellulose is a low price, renewable and biodegradable resource, being the most abundant bioplastics in nature; that acts as a consolidation component of plants and bacteria. It is a linear polysaccharide consisting of repeated cellboise units, a combination of two anhydroglucose rings linked via a β-1,4 glycosidic bond (Fahmy et al., 2020; Nechita & Roman, 2020).

Cellulose shows regular arrangement and structure of hydroxyl groups, characterized by its tendency to organize crystalline microfibrils with strong hydrogen bonds. This makes it a special biomaterial with a mix of useful properties such as high mechanical strength, low density, high durability, no presence of toxic elements, interesting and easy chemical modification, and stability (Luzi et al., 2019).

Cellulose is generally synthesized in plants, but it is also available in some bacteria (Acetobacter xylinum). Algae and higher plants such as cotton linter, corn husk, wheat, rice, maize, and barley stalk are principal examples. Additionally, cellulose content varies according to the botanic species, e.g., the cotton has about 90% cellulose, wood 40-50%, or bast fibers such as flax, hemp, or ramie about 70-80% cellulose (Fotie et al., 2020; Fahmy et al., 2020; Rai et al., 2021). Even though cellulose can be extracted from these animal and vegetable species, the agro-waste provides an economically attractive source for valorization to cellulose at the industrial scale (Fotie et al., 2020; Fahmy et al., 2020; Rai et al., 2021).

On the other hand, due to its hydrophilic character, water insolubility, poor film-forming ability and high crystallinity, the cellulose cannot be used in its native form. Nevertheless, scientific studies in bioplastic science allow to overcome these drawbacks (Nechita & Roman, 2020). Different chemical modifications, such as etherification and esterification, are frequently considered to improve the thermal processability cellulosic materials (Luzi et al., 2019). Cellulose derivatives such as carboxymethyl cellulose (CMC), methyl cellulose (MC), ethyl cellulose (EC), hydroxypropyl and hydroxyethyl cellulose (HPC and HEC) and hydroxypropyl methyl cellulose (HPMC) are examples of the already commercial formulas. These products can be used for both the wet-end and surface finishing of papers to improve barrier properties (Nechita & Roman, 2020). Numerous modified celluloses are commercially available, the main ones are cellulose esters (for melt compounding), cellulose acetate, and regenerated cellulose for fibers (Luzi et al., 2019).

Regenerated cellulose like cellophane has been used as a thin and transparent sheet for its high barrier to gases, oils, greases, and bacteria. Such properties also have contributed to the cellophane expansion in complex multilayer packaging capable of preventing the meat from oxidation and discoloration and from spoilage of fresh and dried oxygen-sensitive foods (Fotie et al., 2020; Rai et al., 2021).

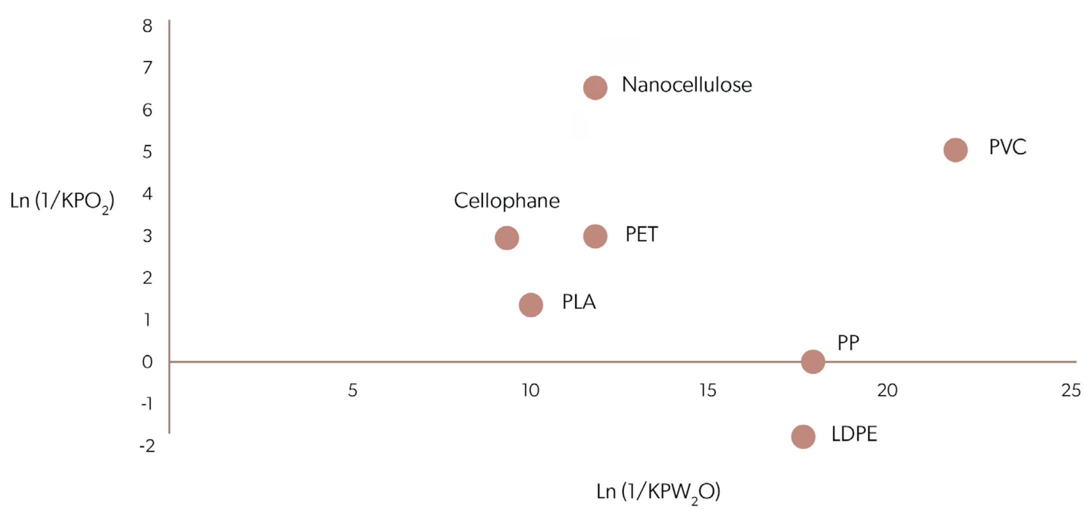
In the paper industry, cellulosic fibers are the primary raw material for paper and board production, but cellulose and its derivatives can also be used as coating on paper packaging (Nechita &

Roman, 2020). In food packaging, cellulose derivatives were used since the early 90s. Cellulose and its derivatives have the capability to mechanically reinforce and enhance the barrier properties of polymer materials. Besides, cellulose derivatives are more resistant to microbial attacks and enzymatic cleavage than native cellulose (Nechita & Roman, 2020).

Furthermore, the application and processability of plasticizers and polymeric blends are also evaluated considering that the chemical and mechanical characteristics are influenced by the blend composition (Luzi et al., 2019).

The current focus and future updates on cellulose research are based on nanocellulose (NC), including cellulose nanofibers and nano-crystalline cellulose. Plant-based nanocellulose (NC) is a biodegradable and non-toxic material whose mechanical, rheological, and gas barrier properties are competitive compared to those of oil-based plastics. For instance, a comparison illustrated by figure 36 with ethyl vinyl alcohol (EVOH) and polyvinylidene chloride (PVDC) - two traditional plastics used for food shelf-life extension by blocking CO_2, O_2, water vapor and aroma - demonstrates that nanocellulose displays the highest oxygen barrier properties (Fotie et al., 2020).

Figure 36



Oxygen Barrier Property Comparison

Note. Oxygen and water vapor resistance coefficients of the nanocellulose in comparison with synthetic, bioplastics, and bio-based structures with KPO_2 and KPW_20 expressed in cm^3 μmPa^{-1} day^{-1} m^{-2} and cm3 day^{-1} m^{-1} Pa^{-1}, respectively.Source. (Fotie et al., 2020).

Source. (Fotie et al., 2020).

However, the sensitivity of NC in humid ambient and lack of thermo seal ability has proven to be a major obstacle that can difficult its breakthrough in food packaging. Despite the advances, it can be observed that packaging manufacturers have not yet shown a particular interest in the nanocellulose processability due to the lack of guidelines and guarantee on the implementation success. Nonetheless, the nanotechnology advances have opened new opportunities to create cellulose-based materials with higher gas barrier performance (Fotie et al., 2020).

Cellulose Ethers

Cellulose ethers are compounds with a high molecular weight produced by substituting the hydrogen atoms of hydroxyl groups in the anhydroglucose units with alkyl groups. These cellulose derivatives include their offer solubility, viscosity in solution, surface activity, thermoplastic film characteristics and stability against biodegradation, heat (Nechita & Roman, 2020).

The mostly used cellulose ethers are: MC, EC, HEC, HPC, HPMC and CMC. CMC is frequently used as a co-binder and/or thickener in pigment coating color, but these derivatives have some limitations when used as a unique binder, so thus the most common applications are as rheology modifiers or water holding agents. MC, HPMC and HPC are biodegradable thermoplastic polymers that are soluble in cold water (Nechita & Roman, 2020).

HPMC is an edible material with good film-forming properties, which is odorless, flavorless, transparent, stable, oil-resistant, and nontoxic. HPMC has a good miscibility with a wide range of organic and inorganic materials, therefore can be used as film-forming material and to control the barrier and mechanical properties in paper coatings. Furthermore, the barrier properties and smoothness of coated papers with HPMC can be improved by the addition of beeswax.

Cellulose Esters

These polymers are water insoluble with good film forming properties and are often used in combination with the cellulose ethers to obtain the microporous membranes.

Two groups of cellulose esters are used in different applications: organic and inorganic groups. Organic cellulose esters (e.g., cellulose acetate, cellulose acetate phthalate, cellulose acetate butyrate, hydroxipropylmethyl cellulose phthalate) have been commercialized. While inorganic cellulose esters (cellulose nitrate and cellulose sulphate) are transparent compounds with good film forming abilities but are rarely applied alone due to their very low solubility and high flammability (Nechita & Roman, 2020).

Among organic cellulose esters, cellulose acetate (CA) has been receiving attention for use in food contact applications due to its non-toxicity, edibility, and biocompatibility. Furthermore, CA fibers are recyclable, they incinerate easily without leaving residue and decompose effectively in the soil as well as water. The most common applications of CA are as films or fibers.

In paper packaging applications, cellulose esters are part of the paper lamination process. For example, cellulose acetate foils can be attached to the surface of paper or paperboard stock by adhesives or hot laminating either alone or combined with aluminum foils to obtain foods packaging. Using the electrospinning process cellulose acetate nanofibers have been obtained, with potential applications in packaging materials for fresh fruits and vegetables.

Cellulose micro (or nano) fibrillated structures

The obtainment of micro(nano)-scale cellulose fibers has gained increasing attention due to their unique properties: high strength and stiffness, low weight, biodegradability, and renewability

(Robertson, 2012; Nakajima et al., 2017; Nechita & Roman, 2020). In plant tissue, the cellulose molecules are brought together into structural units known as elementary fibrils or microfibrils. These structural elements are packed as microfibrillated cellulose (MFC). The diameter of elementary fibrils is about 5 nm and microfibrillated cellulose, also called nanofibrillated cellulose (NFC), has diameters from 20 to 50 nm. This explains the existence of two classes of nanocellulose (Nakajima et al., 2017):

1. cellulose nanocrystals
2. cellulose microfibrils

Nanocellulose has been around since the early 1980s and can be extracted from natural sources such as cellulose from wood, lignocellulosic waste, and agricultural and foods waste. The extraction use chemical, enzymatic and mechanical processes, which include grinding and refining treatments (Robertson, 2012; Nechita & Roman, 2020).

In paper food packaging, NFC can be used as barrier material (against oxygen, water vapor, grease/oil) in surface paper treatments by sizing and coating (Nechita & Roman, 2020).

When associated with the other cellulosic materials or when used in nanocomposite applications, cellulose micro(nano) fibrillated structures show good barrier characteristics. Based on the compact structure formed by the cellulosic microfibrils as well as their ability to form intra- and inter-fibrillar hydrogen bonds, these cellulose derivatives can be promising for applications as transparent and biodegradable packaging films with high barrier properties (Robertson, 2012; Nechita & Roman, 2020). However, many studies have shown a high interest in applying MFC and NFC to improve the mechanical and barrier properties of food packaging and in the printing processes (Nechita & Roman, 2020).

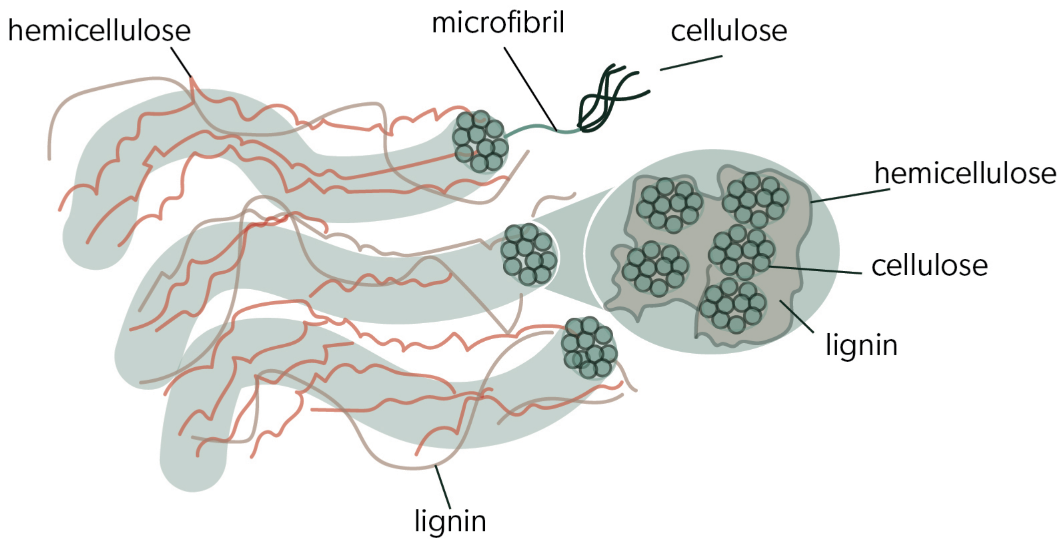
Furthermore, the water vapor and moisture resistance of cellulose nanomaterial-based packaging films can be improved using different technologies such as: layer-by-layer assembly, electrospinning, composite extrusion, casting evaporation, coating, and all-cellulose composites.

In addition, there are a large variety of active-antimicrobial materials which can be combined with NFC, but only a limited number of these have been investigated for antimicrobial paper. These include inorganic materials (e.g., metal oxides: titanium dioxide (TiO_2), zinc oxide (ZnO) and magnesium oxide (MgO), silver, gold, or copper nanoparticles) and organic compounds (polysaccharides based on hemicelluloses, chitosan, and chitosan derivatives), as well as biomolecules (nisin).

4.2.2 Hemicellulose

The hemicelluloses (HCs) are the second most abundant plant polysaccharides after cellulose in the cell walls of lignocellulosic biomass, representing 20-35% of this, as a function of biomass source (figure 38). Unlike cellulose, in which the monomer units are chemically homogenous, hemicelluloses are a series of complex, branched, and heterogeneous polymers. Hemicelluloses of cereal straws have a backbone of (l→4)-linked β-d-xylpyranosyl units. Their relative content and structural composition also vary within a species depending on the location in the plant or cellular tissue origin (Nechita & Roman, 2020).

Figure 37



Arrangement of hemicellulose in the plant cell walls
Source. (Nechita & Roman, 2020).

Hemicelluloses are abundant in agricultural by-products and woody materials. Cereal straws comprise the major portion of plant materials, and HCs amount is according to the particular plant species, such as: maize steins (28.0%), barley straw (34.9%), wheat straw (38.8%), rice straw (35.8%), and rye straw (36.9%) (Nechita & Roman, 2020).

In the paper industry, native or modified hemicelluloses have been used as additive to improve paper strength, retention aid, as binder in paper coatings or as wet reinforcement for cellulose nanocomposites (Nechita & Roman, 2020).

A commercial product-based xylan HCs has been developed in Sweden (Skalax), which can be applied as a thin film to a surface of paper-based materials by dispersion coating to provide barrier properties against oxygen, aroma, grease, and mineral oil. Nonetheless, some studies show that when used as food packaging, the water vapor permeability of xylan-based films are influenced by the low stretchability and other components. The most common applications of this material are oxygen barrier coatings on food or packaging. However, even though the raw material for xylan hemicellulose is abundant, the presence of a commercial product on the market and its large-scale extraction are still limited (Nechita & Roman, 2020).

In recent studies, functional HCs composite films with flexibility, thermoplasticity and UV-shielding ability were obtained using esterified HCs with vinyl benzoate reinforced with poly (vinyl alcohol) (PVA) and zinc oxide nanoparticles (ZnO). The films showed good flexibility and moderate water and oxygen barrier abilities. Chemical modifications improve HCs solubility and processability, and the addition of PVA and ZnO enhances the functional properties of the films, making them suitable and sustainable materials for application in the food packaging field.

Xylan HC was also used as an additive in NFC-based nanocomposite films to improve the water absorption capacity. The results demonstrate the feasibility of hemicellulose, which acts as a plasticizer in NFC films and their potential application to prepare bioinspired nanocomposite films for food packaging. Moreover, Glucomannan hemicellulose exists in high quantities in wood from coniferous trees (spruce) and exhibits interesting film-forming properties. It can be extracted from process streams of newsprint paper mills or fiberboard manufacturing. Glucomannan films have demonstrated good gas barrier properties, and therefore this HC has potential for use as components in barrier coatings for cardboard or paper for food packaging. Cross-linked glucomannan is soluble in water and organic solvents, has good water absorption properties, and could be a potential alternative as a *green* absorption material (gels, etc.).

4.2.3 Starch

As has been described in chapter II (section 2.3.2), starch is an abundant natural compound that is present in all plant components (stem, seeds, fruits, roots, etc.). Starch is the plant "battery" since it represents energy storage in chemical form, but also the main energy source for animals and humans (60-70%) of the human caloric intake comes from starch (Attaran et al.,2017; Avramescu et al., 2020).

There are several advantages to using starch, such as its low cost, large availability, non-toxic nature, and versatility in processing (as flexible as polyethylene and rigid as polystyrene), but it also has disadvantages. For example, raw starch is brittle, too hygroscopic, and has low mechanical properties (Attaran et al.,2017; Avramescu et al., 2020).

Starch is the most frequently present bio-additive in the paper industry, being used for both wet-end and surface/coatings applications. In their native form, starch can be used as a sizing agent (in the size press of paper machine) while after chemical physical, and/or enzymatic modifications it can be used as a coating additive due to its excellent film forming ability (Nechita & Roman, 2020).

Due to the brittleness of the native material with semi-crystalline nature, starch films do not have adequate flexibility and mechanical properties when are used in packaging. These properties can be improved by adding plasticizer with other materials, chemical or physical modification, enzymatic treatments, or composite combinations. Glycerol, sorbitol, or xylitol are typically plasticizers used for reducing the brittleness of starch films. However, due to the solution viscosity, film formation, and resistance to retrogradation (liquid to gel formation), thermoplastic starch is less used in paper coatings (Nechita & Roman, 2020).

However, for paper coatings, the most common practice is chemical oxidation of starch by reducing chain length and molecular weight of oxidized starch. The result will be a reduced viscosity of the coating solution.

The acetylation reaction is one of the most interesting ways to decrease starch hygroscopicity. Thus, high efficiency in paper coatings had the starch derivatives such as: acetylated starch, cationic starch and hydroxypropylated. Coatings with acetylated starch have been applied on kraft paper, with results in significant reductions of the water absorptivity and WVP (water vapor permeability) and improvement of barrier properties against gases and aroma compounds, maintaining the quality of the foods during storage.

Starch nanocomposites

Starch nanoparticles have many advantages in comparison with traditional dispersions of cationic or anionic starch[2]. These exhibit higher bonding strength and lower suspension viscosity at relatively high solid contents (up to 30 wt.%) (Nechita & Roman, 2020).

Another possibility of improving the starch hydrophobicity when used in paper coatings consists of mineral pigments (hyper platy nano clays) with high aspect ratios (highly plate particles). The coating color is then applied in thin layers on the paper and board surface with good results in water, oil, and air barrier properties, and maintaining the freshness of packed products (Nechita & Roman, 2020).

The potential of starch in paper coatings is based on its ability to be a suitable carrier for compounds with barriers or active antimicrobial properties, such as mineral nanoparticles (ZnO, MgO, metallic ions, nanoclays) or nanofibrillated cellulose to obtain composite coatings with good barriers or active antimicrobial properties for food packaging. By using carboxymethyl starch and ZnO nanoparticles composite coatings, the optical properties (e.g., whiteness and brightness) of coated paper are improved. Furthermore, these coated papers showed excellent antifungal and UV-protecting properties, compared to bulk ZnO-coated paper (Nechita & Roman, 2020).

Additionally, a nanocomposite film of silver nanoparticles (AgNPs) with starch was prepared showing promising mechanical, gas barrier, and antibacterial properties. Besides it, this nanocomposite film proves its safety due to migration of silver from the nano-film was discovered to be within the permissible limit (Fahmy et al., 2020).

Moreover, a recent work has demonstrated that water vapor and oxygen permeability decreased by 54% and 26%, respectively, by adding talc (3% w/w) to thermoplastic starch. Talc is a common silicate mineral that is distinguished from almost all other by its extreme softness (Britannica, n.d.). The same study also shows that 3% w/w of talc increased the stiffness by 15%, while 5% w/w of talc increased the Young's modulus up to 68% and 81%, respectively. These results show talc nanoparticles can act as an obstacle in preventing water vapor permeability as coating on paper (Fahmy et al., 2020).

4.2.4 Pectin

Pectin is a chain of α - (1,4)-linked D-galacturonic acid subunits that have an "accordion-like" conformation which result in molecules with extensible characteristics. Pectin is a crucial polysaccharide considering the structural role in plant tissues. Its biological function reticulates the cellulose and hemicellulose fibers, producing a more resistant structure (Avramescu et al., 2020).

It is found in almost all plants in different concentrations mainly in the middle lamella layer between cells. Pectin is a generic name for a family of polymers that differ in molecular weight, chemical configuration, and abundance of monosaccharide subunits. Rhamnogalacturonan I, rhamnogalacturonan II, and homogalacturonan are some examples (Avramescu et al., 2020).

[2] Cationic polymerization reactions are sensitive to temperature. Both the reaction rate and molecular weight rapidly decrease with increasing temperature. Anionic polymerization reactions typically yield more regular polymers with less branching, more controlled tacticity and narrow molecular weight distribution. https://polymerdatabase.com/

Natural sources of pectin can be obtained by different agricultural wastes: apple pomace (4.2-19.8%), citrus peel (13.4-37.52%), sugar beet pulp (23-24.87%), tomato waste (7.55-32.6%), mango peel (17.15%), watermelon rinds (19-21%), etc (Avramescu et al., 2020).

Polysaccharides from marine biomass

4.2.5 Chitosan

Chitosan is a linear polysaccharide of β-(1-4)-linked D-glucosamine and N-acetyl-d-glucosamine. Chitosan is produced by deacetylation of chitin behind controlled parameters such as temperature, native origin of chitin, and alkali (Luzi et ak., 2019). Chitin is a natural polymeric material that composes the exoskeleton of arthropods, but it is also present in the cell walls of yeasts and fungi. Chitin is generally present on the market after chemical treatment of crab's and shrimp's wastes (Attaran et al., 2017; Luzi et ak., 2019).

Chitosan is biodegradable, biocompatible, and non-toxic (Nechita & Roman, 2020). The membrane of chitosan is characterized by modest water permeation and low oxygen permeability, crucial in the conservation and preservation of some food that is sensitive to the presence of oxygen in the packaging. This natural bioplastic derived does not show affinity to water, in fact it is insoluble in water, but it becomes soluble in acidic solutions (acetic acid, formic acid, etc.) (Luzi et ak., 2019). Furthermore, chitosan exhibits film forming ability and antimicrobial properties (Fahmy et al., 2020; Avramescu et al., 2020).

On the other hand, its interesting film-forming characteristics allow the realization of coatings and membranes capable of being utilized for food conservation (Luzi et ak., 2019). This bioplastic can be easily used with other bioactive agents; as environment-friendly food packaging materials with various promising properties (Fahmy et al., 2020).

The development of polymeric blends composed by chitosan and other polymeric materials, such as PLA, starch, and proteins with nano-fibrillated cellulose or inorganic nanoparticles (nanoclays, metallic oxides, silver nanoparticles) represents a valid approach to guarantee an enhancement of mechanical performances, active-antimicrobial and barrier properties (Habel et al., 2018; Nechita & Roman, 2020). Several other approaches, like coating, dipping, casting, Layer-by-Layer (LbL) assembly, and extrusion, have been considered to realize chitosan systems with several characteristics. The promising food sector of chitosan-based coatings as antibacterial active compounds and sensing and barrier systems has an enormous progresses and promising future (Luzi et ak., 2019).

In paper coatings, chitosan and their derivatives were considered for active-antimicrobial features proved in many research studies, and for their barrier properties against water vapor, oxygen, oils, and grease. In these applications, chitosan can be used as an emulsion, using different coating methods or size press to be applied on paper surface (Nechita & Roman, 2020).

Furthermore, due to its appropriate film-forming and solubility properties in dilute aqueous acid solutions it can be used to cast free-standing films or coating onto paper/board or plastic films (Attaran et al., 2017).

The water-soluble chitosan derivatives (alkyl chitosan-ACh, quaternary chitosan-QCh and carboxymethyl chitosan-CCh) and microfibrillated cellulose were used as barrier and mechanical strength additives in coating formulas for packaging paper grades. The results have shown that the water and water vapor barriers are improved by applying ACh alone or in combination with MFC. The tensile strength properties (15-20%) and water vapor transmission rate (WVTR) (~30%) are enhanced by using CCh, while QCh has moderate effects on the water barrier and strength properties (Nechita & Roman, 2020).

Additionally, there have been recently reports for the first time, about water barrier blends of chitosan with EVOH copolymers. Optimal properties in terms of microstructure, optical characteristics, biocide, and water barrier activity could theoretically support the development of new bio-coatings based on chitosan salts and EVOH. The chitosan/copolymer blend can preserve transparency and dimensional stability, even in the presence of humidity. It also shows improved water barrier and exceptional biocide characteristics compared to chitosan neat (Luzi et ak., 2019).

4.2.6 Algae species

Algal plastics have become a recent trend as they possess high photosynthetic efficiency and growth rate. One unique characteristic of this natural-derived polymer is that it does not compete with the food source, making it a sustainable source for bioplastic extraction (Rai et al., 2021).

Nowadays, two approaches are commonly used to obtain bioplastic from algal sources. One is based on a composite preparation using algal biomass with plasticizers and additives, following mechanical/physical extrusion. The other approach involves the extraction of bioplastics (Starch, PHBs) cultivated within the intracellular spaces in the algal cells (Rai et al., 2021).

Microalgae species, Chlorella and Spirulina have been used for their bioplastic production potential. Chlorella has mostly been used for biomass-plastic blends. Blending here is necessary to obtain commercial grade bioplastics of high quality that significantly degrades over 170°C. The tensile strength of the algal based bioplastic has been improved by applying plasticizers like glycerol, PVA and compatibilizer like maleic anhydride (MAH). At the same time, compatibilizers are used to chemically modify plasticizers to facilitate the blending of microalgae biomass to PE and PP (Rai et al., 2021).

PVA blended spirulina plastic with glycerol as plasticizer and MAH as compatibilizer was prepared and the resultant bioplastic had tensile strength and elongation higher than that of a conventional polymer. An addition of MAH enhanced the surface smoothness. Another thermoplastic preparation using Spirulina biomass, is with wheat gluten and bio-based plasticizers, which had high tensile strength, enhanced thermal stability and low water absorption (Rai et al., 2021).

Aside from these two species of microalgae, Phaeodactylum tricornutum, Calothrix scytonemicola, Scenedesmus almeriensi, Chlamydomonas reinhardtii and their consortiums have been used to produce intracellular bioplastic like starch or PHA. However, the use of additives and plasticizers of chemical nature to obtain microalgae products limits their use in applications such as packaging of food and beverages (Rai et al., 2021).

Alginate

Alginate is a polysaccharide usually available as salts of sodium and calcium of alginic acid and naturally present in brown algae. Advantageous properties such as film-forming ability, non-toxicity, biodegradability, and biocompatibility are features that make alginate as one of the most promising and intensively studied bioplastics (Nechita & Roman, 2020) especially for bioplastic-based coatings.

Due to the large number of alginates and its derivatives already used as additives in the food industry, these bioplastics are safe for their use as functional barriers for food-contact materials. Moreover, different water-soluble alginate formulations are available on the market, which can be applied with conventional coating methods used in the paper packaging industry. However, only a few reports in the literature concerning the potential of alginate coatings to improve water barrier properties of coated paper, either used as sole polymer or in combination with other bioplastics (i.e., chitosan). Results show that this bioplastic cannot reduce the water resistance of paper but has some synergistic effect when used in combination with chitosan for instance (Nechita & Roman, 2020).

The water resistance of paperboard was improved by using the water-soluble sodium alginate (SA) combined with calcium chloride. By applying sodium alginate as the first layer on paper surface, followed by a post-treatment of dip-coating calcium chloride. The final coated paper with sodium alginate exhibits excellent oil resistance via the application of only 5 g/m2 of coating weight (Nechita & Roman, 2020).

In recent research studies composite coatings based on SA/sodium carboxymethyl cellulose and SA/propylene glycol alginate were used to obtain fluoro-free greaseproof papers. The results showed an exceptional kit of values that meet requirements in food package applications. The improvement of water resistance by the decrease of surface energy was obtained by a combining of propylene glycol alginate with sodium alginate. The reduction of surface energy can effectively improve the resistance for oil and water of paper (Nechita & Roman, 2020).

Table 13 shows the most representative properties of polysaccharides bioplastics and its principal food packaging applications.

Table 13. *Characteristics and food applications of polysaccharides*

Polysaccharide	Barrier Properties	Additional properties	Main food applications
Starch	• Barrier against gases and aroma • Low vapor permeability • Antifungal • UV-resistant	Biodegradable Transparent Odorless and tasteless High elongation High tensile strength	Extruded bags Net bags Containers Films and coatings
Cellulose	• High barrier against gases, oils, greases and bacteria • Ressistant to microbial attacks • Highly sensitive to water	Biodegradable Transparent Good mechanical properties Net to perform modifications, use plasticizer, or polymer blend	Cellophane membranes
Chitin	• Antibacterial and fungistatic properties	Biodegradable Highly transparent Biocompatible and non/toxic	Extruded bags Coffee capsules Films and coatings
Chitosan	• Modest water permeation • Low oxigen permeability • Insoluble in water • Soluble in acid solutions • High barrier against oils and grease • Antibacterial and fungistatic properties • Barrier to gases	Biodegradable Brittle-need to use plasticezers Biocompatible and non/toxic Good mechanical properties	Films,coatings and edible coatings

Source. (Luzi et al., 2019; Nechita & Roman, 2020; Fahmy et al., 2020; Avramescu et al., 2020 Fotie et al., 2020; Rai et al., 2021).

4.3 Protein-based coatings

Proteins represent, besides polysaccharides, another important pillar of the biological kingdom. There is a huge amount of academic work (Gontard & Guilbert, ca. 1993; Khwaldia et al., 2010; Tappi & Pima, 2011; Mekonnen et al., 2013; Coltelli et al., 2015; Khwaldia et al., 2017; Attaran et al., 2017; Avramescu et al., 2020) on protein-based coatings which could fill the need for high-performance renewable materials in the food packaging industry including paper coatings applications.

Proteins have a unique structure that confers a wide range of functional properties, especially a high intermolecular binding potential often allowing protein-based films to exceed polysaccharide and lipid-based film's mechanical properties. They have been studied in combination with different elements (plastic, paper, leather, etc.) or as an edible coating for food (Coltelli et al., 2015).

Proteins are known as natural polymers composed of amino acids in different proportions that vary from source to source. As raw materials are extracted from milk (casein, whey), corn (zein), wheat (gluten), cereal grain (8% - 15%), sorghum, peas (22% - 28%), sunflower seeds (28% - 42%), rice bran, cottonseed, peanut, keratin, etc. soybeans (38% - 44%); and from other animal sources such as collagen, gelatin, eggs (white egg). Some of these can also be recovered from under-used industrial by-products (Coltelli et al., 2015; Avramescu et al., 2020).

The intrinsic hydrophilicity of protein films results in good adhesion to polar surfaces such as paper and a good barrier to oxygen and carbon dioxide, but scarce to water vapor. In addition, some high molecular weight proteins are insoluble or only weakly soluble in water and may be used to obtain water resistant films. Applied as coatings on substrates used in the packaging sector, proteins are aimed to improve a wide range of properties such as gas and water vapor barrier and seal-ability. For instance, whey protein coatings have been shown as good barriers when applied on paper by increasing oil resistance and reducing water vapor permeability. Also, they exhibited excellent visual and good mechanical properties on their substrates. Zein-based coated paper has an oxygen barrier highly over performing those of PE that may be usable on paper boxes as an alternative to paraffin (Coltelli et al., 2015).

Besides, the main object of the present section dealing with coatings is the use of proteins as adhesive can also be relevant for the paper industry. Various proteins have been used for adhesive production during both ancient and modern ages, including blood protein, casein, and soybean protein. More recently also whey protein, coming from cheese industry waste, blended with sucrose, was used for formulating adhesives to be employed in the paper industries. Furthermore, protein-based adhesives ensure its recyclability because they can be washed away from the fibers during the recycling process and successive pulping. Traditional glues mostly stick onto the fibers making them less suitable for being employed in new paper-based products (Coltelli et al., 2015).

Protein films formation occurs through the denaturation of the initial material by heat, solvent utilization, and PH. The control of these parameters will highly influence the final quality film. Bioplastics are not always thermoplastic and to be dissolved need the addition of some plasticizers. The absence of plasticizers leads to thermal degradation, while adding plasticizers, the bioplastic becomes soft, elastic, and configurable in any shape (Avramescu et al., 2020).

An overview of proteins with film-forming properties that were tested for food packages, stand-alone films and edible coatings is illustrated in table 14 (Coltelli et al., 2015).

Table14. *Overview of proteins with film-forming properties tested for food packages, stand-alone films, or edible coatings*

Origin	Plant Protein	Animal Protein	Fungi
Globular proteins	wheat gluten rirce (bran) protein corn zein soy protein pea protein phaseolin peanut protein pistachios protein lupin protein sunflower protein cottonseed protein amaranth protein sorghum kafirin potato protein	wheat gluten egg albumin	hydrophobins
Fibrillar proteins		casein gelatin/collagen keratin fish myofibrillar protein	

Source. (Coltelli et al., 2015).

Protein characterization and functional properties

Proteins can be characterized and grouped according to their amino acid composition, geometrical conformation, solubility in various solvents, molecular weight, sedimentation behavior, their surface polarity and distribution or whether they maintain their native molecular configuration. Their potential application in food packaging coating depends on its techno-functional properties such as solubility, viscosity, and network formation (Coltelli et al., 2015).

Protein solubility - at least partial - and protein swelling is a prerequisite for a stable protein dispersion to be applied as coating layer to induce viscosity and network-formation (Coltelli et al., 2015).

Most proteins are soluble in water or other polar solvents, whereas few proteins become soluble in moderate polar solvents like alcohols. Protein solubility is highly dependent on pH of the solution and the ionic strength (Coltelli et al., 2015).

Only a few of the proteins presented in table 14 are widely commercially available as dried and concentrated or isolated protein fractions with protein content between 65% and 95% by weight. The poor commercial availability of protein ingredients from many raw materials, high costs and unfavorable technical properties have limited the interest in packaging applications. Nevertheless, extensive research in applications such as coatings on paper for food packaging was carried out (Coltelli et al., 2015). Below is presented some of the protein-based coatings for papers that have industrial potential to replace synthetic coated paper.

4.3.1 Gelatin

Gelatin refers to the purified and modified collagen protein. Collagen is the main protein component in animal connective tissues such as skins, cartilages, and bones (Coltelli et al., 2015). Gelatin is obtained through the hydrolysis of collagen and other insoluble proteins, is an important candidate for coatings production. And it is formed from proline, hydroxyproline, and glycine (Avramescu et al., 2020).

Gelatin has excellent film forming properties, but its films are brittle, and plasticization or toughening is needed for most practical applications. To modulate mechanical properties, the application of different plasticizers for gelatin films (sucrose, oleic acid, citric acid, tartaric acid, malic acid) were studied. The results show a better ductility with malic acid (Coltelli et al., 2015).

Additionally, mechanical properties of gelatin from different sources were compared. Tensile strength, elongation percentage at break and puncture deformation declined in the following order: gelatin from mammals, from warm, and from cold-water fish (Coltelli et al., 2015).

Gelatin is used in different mixtures for preserving meat food and even for improving the recipes. Besides, gelatin is often used as an additive in several water-based formulations for imparting anti-microbial or adhesive properties to paper (Coltelli et al., 2015).

4.3.2 Caseins

Dairy protein powders are the dominant protein ingredient and are used ubiquitously in the food industry. Thereby casein is the principal protein fraction in cow milk that accounts for 80% of its total proteins. It appears as self-assembled casein micelles with 50-300 nm diameter in milk (Coltelli et al., 2015). Casein represents a family of phosphoproteins (αS1, αS2, β, κ) found in different proportions in human or animal milk (Avramescu et al., 2020).

In general casein-based films have a hydrophilic character. For this reason, the films are sensitive to water and show high water vapor permeability. However, the introduction of nanoparticles can enhance barrier and mechanical properties to be suitable for industrial applications. For instance, it has been reported that Sodium caseinate (NaCas)-based formulations reinforced with halloysite nanotubes (HNTs) were applied as coating materials on paper sheets. The results showed that when increasing coating weight and HNT content, the coated paper reduced within the range of 47-79% the water vapor permeability (WVP) (Khwaldia et al., 2017).

Furthermore, evidence proves that the casein-based paper coatings thickness significantly influenced the barrier properties. For instance, an increase in the coating weight resulted in water vapor permeability decrease (Coltelli et al., 2015).

On the other hand, the effect of cross-linking in casein proteins related with its performance as coating was studied by many authors, but the results were not all in agreement. Nevertheless, it can be said that water vapor permeability can be influenced by molecular weight and crystallinity grade. Hence, the different chemical structures can influence barrier properties significantly (Coltelli et al., 2015).

Another treatment investigated to improve the casein film's properties is the enzymatic cross-linking of proteins with itself or with other proteins (for example zein hydrolysates) by transglutaminases. The resulting films showed higher mechanical strength and flexibility (Coltelli et al., 2015).

4.3.3 Whey

Only from the mid-eighties, the interest in whey has increased due to its content of nutritious and highly functional proteins (Coltelli et al., 2015). Whey is the soluble constituent of milk and represents about 20% of the proteins in cow milk. Being the by-product of cheese and casein manufacturing, it is highly available with an average global production capacity of 21.6 million tons/year (Avramescu et al., 2020). Nonetheless, it is still underused due to its high-water content and respective high transportation and processing costs (Coltelli et al., 2015).

Due to cross-linking and the high content of intermolecular hydrogen bonds, whey protein films provide aroma, oil, and excellent oxygen barrier properties. At low humidity conditions, oxygen permeability is comparable with EVOH polymer's performance and can therefore be used for coatings to improve food packaging's oxygen barrier property. On the other hand, due to their hydrophilic character, whey protein films display poor water vapor barrier (Coltelli et al., 2015).

However, water vapor barrier properties can be enhanced by the incorporation of lipids. Being used for paper coatings, whey proteins improved the packaging material properties by increasing oil resistance, reducing water vapor permeability as well as oxygen permeability compared to uncoated paper. Furthermore, the type of plasticizer used for film formation highly influences the resulting barrier properties of whey protein coatings. Whey protein isolate coated paperboard showed a good grease barrier using glycerol as a plasticizer. However, the high migration potential of glycerol caused cracking of the coating resulting in minor grease resistance (Coltelli et al., 2015).

Incorporating the used plasticizers increased elongation and decreased tensile strength in the following order: glycerol, polyethylene glycol (200), sucrose and polyethylene glycol (400). The most efficient plasticizers for desirable mechanical properties were glycerol and PEG 200 (Coltelli et al., 2015).

Similar to casein, whey has to be molded along with several other adjuvants such as chitosan and alginate to reach food packaging requirements. Below is presented some of the last blends analyzed:

- Whey protein concentrate/wheat cross-linked starch composite film
- Lactis BB-12–whey protein isolate–alginate
- Whey–glycerol (30–60% w/w of whey)
- Maltodextrin–Arabic gum–whey
- Green tea/rosemary extract–whey
- Whey–glycerol (5–15% w/w)—rosemary and thyme extracts

Studies show that the produced films act as a barrier for different microorganisms and present improved physicochemical properties (Avramescu et al., 2020).

4.3.4 Soy

Soy protein isolate (SPI) is usually a by-product of the soybean oil industry. Like other globular proteins, SPI shows good film-forming properties and is therefore suitable for edible films and coatings. By modification of SPI formulations, mechanical properties, as well as water resistance, can be improved. Soy proteins are used for cast film production out of aqueous solutions (Coltelli et al., 2015).

To avoid brittleness, a plasticizer must be added for film formation. For soy protein-based films, glycerol is the plasticizer most often used. Film qualities can be improved by alkali treatment, different types of plasticizers like glycerol or sorbitol, via chemical modification or addition of hydrophobic substances such as waxes or lipids. Using plasticizers and chemical modification (e.g., by sodium sulfite), dry processes like extrusion processing of soy protein films are also possible.

4.3.5 Corn Zein

Zein is a class of prolamine protein found in corn, which was first discovered by Gorham in 1821 in the product zea, known as *Indian corn* (Attaran et al., 2017). As the other natural protein types, zein protein composition varies widely depending on corn variety (Coltelli et al., 2015).
Within the studies about the effect of plasticizers on barrier properties, some authors investigated the oxygen permeability of zein films containing a mixture of sorbitol/glycerol/mannitol. The conclusion was that glycerol and sorbitol decreased oxygen permeation (sorbitol more significantly). In contrast, mannitol increased oxygen permeability. It also was found that glycerol favors the achievement of smoother surfaces with lower roughness than sorbitol and mannitol (Coltelli et al., 2015).

At the present time, much of the zein from corn gluten meal is applied for food and pharmaceutical coatings. Being mostly nonpolar in nature, zein films have been explored for coatings in numerous food applications. However, due to gradual water absorption the utilization of these materials for packaging decreases (Attaran et al., 2017).

In table 15, the barrier and mechanical properties of main protein's-based coatings are compared considering low-density polyethylene (LDPE). This comparison demonstrates several proteins significantly improve the oxygen barrier properties based on the LDPE. However, it is necessary to contemplate that poly (ethylene terephthalate) (PET) shows better oxygen barrier properties than LDPE and is the main market competitor of proteins as paper coating even though PE has mechanical properties completely different from them. Additionally, a chlorinated polymer such as poly (vinylidene chloride) (PVDC) shows very good oxygen barrier properties. However, the latter is banned in many countries because it can release chloride acid in the operations occurring at its end life (recycling or thermal treatments). Poly (Ethylene-co-vinyl alcohol) (EVOH) is considered up to now the best oxygen barrier polymer (Coltelli et al., 2015).

In all cases, the barrier to the humidity of proteins is low and can be improve by using hydrophobic additives or designing a multilayer system in which a biodegradable polymer is employed.

Table 15. *Comparative table for protein films properties*

Protein	Barrier Properties		Mechanical Properties		Adhesion
	Oxygen	Warer Vapor	Tensile Strengh	Elongation at Break	
casein/caseinates	3	1	1	2	3
whey	3	1	1	2	-
gelatin	1	1	2	1	3
wheat	2	2	1	1	2
soy	3	1	1	2	2
corn zein	3	1	1	1	2
feathers	-	2	2	1	-
egg albumin	-	-	2	2	-
fish myofibrills	-	2	2	1	-
hake	-	2	1	2	-
mussel	-	-	-	-	3
potato	-	-	2	1	2
rice	-	1	2	0	-
amaranth	3	2	0	2	-
pea	-	1	1	1	-
phaseolin	3	1	1	1	-
chickpea	-	-	-	-	-
faba bean	-	1	1	2	-
lupine	-	-	-	-	-
pistachio	-	0	2	1	-
peanut	0	0	1	2	-
sunflower	-	1	0	1	-
cotton seed	-	-	-	-	-
sorghum	-	1	1	2	-
hydrophobins	-	-	-	-	-

Note. 0, very low; 1, moderate: 2, high (comparable with petrochemical option-LDPE-); 3, very high (better than petrochemical option -LDPE-).
Source. (Coltelli et al., 2015).

4.4 Lipid and composite coatings

Lipid compounds, such as long-chain fatty acids and waxes, can be incorporated into the film or coating matrix because of their hydro repellency. Waxes are the most efficient substances to reduce moisture permeability. Their high hydrophobicity is a consequence of a high content in esters of long-chain fatty alcohols and acids, as well as long-chain alkanes (Khwaldia et al., 2010).

The incorporation of lipids, waxes, or long chain saturated fatty acids in protein films as a coating reduces the WVP. This barrier efficiency strongly depends on the component's polarity and the uniform distribution of hydrophobic substances (Coltelli et al., 2015). Paper and paperboard are frequently wax-coated to improve their water-resistance and increase the shelf life of food packaged products. Paraffin wax applied in a molten form was commonly used to produce a water vapor barrier (Khwaldia et al., 2010).

Lipid coatings provide a good moisture barrier, but they have certain disadvantages such as brittleness, lack of homogeneity, presence of pinholes and cracks in the surface of the coating. Although lipids indeed provide very good WVTR, when used as standalone films, they have a very poor integrity and thermomechanical properties, and poorer oxygen barrier than protein-based films. Therefore, a composite or bi-layer approach is necessary (Coltelli et al., 2015).

Composite coatings or multilayer coatings, applied either in the form of an emulsion or in successive layers (multilayer coating), have been prepared to combine the good structural and gas-barrier properties of hydrocolloid coatings with the good moisture-barrier characteristics of lipids. It is essential to consider that the method of application affects the barrier properties of the final coatings obtained (Coltelli et al., 2015).

Moreover, it has been measured the water barrier and grease permeation properties of Kraft paper coated with a combination of zein and paraffin wax, concluding that the zein layer of the bilayer coating contributes grease-proofing while wax layer improves the water resistance (Coltelli et al., 2015).

Finally, after having been presented state-of-the-art bioplastics for paper coating, next table 16 shows some applications of bioplastic' coatings on paper by type of food and barrier requirements.

Table 16. *Barrier properties by food type with classic and bio-based packaging alternatives*

Type of food	Barrier requirements	Classical packaging solution	Bio-based packaging solution
Meat/fish	high barrier against oxygen and gas (aroma)	trays (PS,PP,PVC with EVOH + LDPE or PV as coating) + foil (PVC) or lid bags for short term storage bags for short term storage-waxed paper (wrapping) paperboard external packaging transparent films(PP, PE)	multilayer packaging materials, functional bio-based coating (modified starches) + antimicrobial and anti-fogging system
fresh cheese	high barrier properties grease, O2, CO2, and N2, aroma , and light	transparent films/foils wrapping films (PE, laminated) trays bags (LDPE,EVA,PVdC) plastic cups (HDPE,PP,PS)+high barrier lid (PA/LDPE)	eco-paper for short term storage (wrapping) PHA/modified PLA films
dairy products / liquids	high barrier properties water vapor (scavenging moisture), O2, light moderate for grease and aroma	waxed paper, LDPE, PVC, or aluminium-coated/laminated paper or paperboard plastic films (BOPP), metal cans	paper/paperboard coated with bio-based materials
salad (flexible packaging)	high oxygen barrier water resistant	transparent laminated PP films	PLA films (perforated) coated paper with bio-based films+transparents window
take-away food	grease, thermal insulation	polystyrene foam trays	paperboard with internal-grease barrier coating

Source. (Luzi et al., 2019)

4.5 Regulations

Food safety

When it comes to the regulating of protein, polyssacharides or lipids films and coatings, either applied directly to food or over a substrate, they can be classified as food contact materials, food additives, ingredients or even food products. Nevertheless, there is no specific legislation on bioplastic coatings yet, so conventional regulations must be applied in each case (Coltelli et al., 2015).

In the case of the bioplastic coatings applied on paper or board-based materials, food contact materials within the European Union, need to comply with Framework Regulation 1935/200[4] (EC)[3], among other relevant regulations. According to this regulation, food contact materials need to be manufactured in compliance with good manufacturing practice so that, under normal and foreseeable conditions of use. Bioplastic's coatings shall not transfer their components into the food in quantities that could endanger human health or bring about an unacceptable change in the composition of the food or a deterioration in its organoleptic properties. All terms considered, safety regulatory requirements of bioplastic's coatings remain an issue that must still be addressed to scale-up their use to the industry despite the promising results reported, especially because of their absence from the main list of materials and additives approved for food contact (EC 10_2011 in case of plastics) despite their food nature and inherent safety (Coltelli et al., 2015).

More information about bioplastic's-based coatings about the current legislations for bioplastics respect food safety has been contemplated in chapter I (section 1.3.6).

End-of-life

Due to the bioplastic's natural composition presented in this section, they do not make difficult paper recycling and separation in layers is not necessary. Nevertheless, more investigation on biodegradability, compostability, and recyclability is necessary (Khwaldia et al., 2010). For instance, the thickness and the composition of the coating significantly influence paper fibers recycling by pulping (Coltelli et al., 2015). In terms of disposal management, biobased bioplastics coatings must align to the Packaging Directive 94/62/EC4 about the minimum recovery and recycling targets needed.

More information concerned bioplastic's-based coatings about the current legislations, standards, and labels for bioplastics respect biodegradability, compostability and recyclability has been contemplated in chapters II (section 2.2.3) and III (section 3.4).

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[3] Regulation (EC) No 1935/2004 of the European Parliament and of the Council of 27 October 2004 on materials and articles intended to come into contact with food and repealing Directives 80/590/EEC and 89/109/EEC. https://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2004:338:0004:0017:en:PDF

[4] European Parliament and Council Directive 94/62/EC of 20 December 1994 on packaging and packaging waste. In force, current consolidated version: 04/07/2018. https://eur-lex.europa.eu/legal-content/EN/ALL/?uri=celex%3A31994L0062

4.6 Case studies

The state-of-the-art bioplastics for paper coating as food packaging applications are presented in next case studies and summary charts. The cases are presented in a chronological order starting from 2021. There are some trademarks or projects still in R&D stage (Coltelli et al., 2015; Avramescu et al., 2020)

EcoShield®

Cortec
USA, since 2020

Material Sustitution
Polycoated/waxed paper

References
https://www.cortecv-ci.com/whats_new/announcements/EcoShield-Barrier-Coating-PR.pdf

EcoShield® Barrier Coating is a water-borne barrier coating that is recyclable and 100% repulpable designed as an environmentally friendly alternative to polyethylene and wax paper coatings for the manufacture of moisture resistant paper and corrugated boxes.

SUSTAINABILITY

Origin
partly biobased

End-of-life
paper recyclable

Certifications

Barrier properties: ASTM F 1249
Oil and grease resistance: TAPPI T-559 KIT Test
Repulable: TAPPI T 240 om-12 Repulpability Testing
Recyclable: System of Evaluation Aticelca 501:2019
Food Contact Material: FDA

ADDITIONAL PROPERTIES

Milky White to Off-White

Viscous Liquid

ENGINEERING PROPERTIES

water vapor barrier properties compared to a polyethylene coated paper and better than commercial waxed paper.

excellent oil and grease resistance

PRESENTATIONS

flexible and rigid packaging

FOODS

perishable and semi-perishable foods

PROCESSING TECHNOLOGY

extrusion coating
film lamination

Michem® Coat 2000

MICHELMAN

Michem® Coat water-based coating is selected primarily for grease and oil resistance. Other benefits include water resistance, release, coldset gluability, hot melt gluability, recyclability, and compliance with specific food contact regulations.

Michelman
USA, since 2020

Material Sustitution
EVOH, PVDC, LDPE, PE

References
https://www.michelman.com/solutions/michem-coat/michem-r-coat-2000/?utm_source=bpi-certified-coatings-2020-pr&utm_medium=press-release&utm_campaign=bpi-certified-coatings&utm_tags=|printing-and-packaging|sustainability|

SUSTAINABILITY

Origin
partly
biobased

End-of-life
paper recyclable
compostable

Certifications

Compostable: Biodegradable Products Institute (BPI)

ADDITIONAL PROPERTIES

Pale yellow
liquid

cold-set and hot-melt
gluable

Shelf Life
6 Months

ENGINEERING PROPERTIES

designed for maximum oil and
grease performance
water resistance

PFAS-free
PH: 5.0 – 7.0

PRESENTATIONS

flexible and rigid packaging

FOODS

perishable and semi-perishable foods

PROCESSING TECHNOLOGY

extrusion coating
film lamination

BiOrigin™

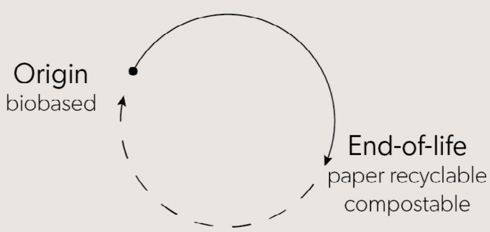
BiOrigin™ is the next generation of sustainable coated paper based in natural fibers and water using bio-based methods to enhance its functionality. BiOrigin™ encompass two presentations commercialy available: Water-based and oil/grease barrier coated paper, both readily printable.

Dunn Paper
USA, since 2020

Material Sustitution
EVOH, PVDC, LDPE, PE

References
https://dunnpaper.com/biorigin/

SUSTAINABILITY



Certifications

Industrial Compostable: EN13432 and BPI
Home compostable: OK comoppost
Biobased content: USDA Certified Biobased Product
Sustainable forest management: Programme for the Endorsement of Forest Certification (PEFC), Forest Stewardship Council(FSC), and Sustainable Forestry Initiative

ADDITIONAL PROPERTIES

White liquid

two presentations: water-based and oil/grease barrier coated paper

ENGINEERING PROPERTIES

ph:10-11

PFAS free by 2023

PRESENTATIONS

flexible and rigid packaging

FOODS

perishable and semi-perishable foods

PROCESSING TECHNOLOGY

extrusion coating
film lamination

Bio-Mi

Bio-mi Ltd. is a small and medium-sized research and development biocompany, dedicated to the production of final and semifinal thermoplastic materials using agro-based, forestry-based, aquatic feedstock, their residues, and side streams. Bio-Mi's advanced materials are to be the suppliers of bioplastics with a wide range of alternative solutions, offering a variety of unprocessed bioplastic materials and standard and customised bioplastic compounds.

Bio-Mi company
Croatia, since 2017

Material Sustitution
fossil-based thermoplastics

References
https://bio-mi.eu/index.php/en/products

SUSTAINABILITY

Origin
50-100% biobased

End-of-life
paper recyclable
biodegradable
compostable

Certifications
Food contact approved
Under the process of certification: OK compost INDUSTRIAL (EN 13432) and OK compost HOME

ADDITIONAL PROPERTIES

printtable
sealable
plasticizer free

ENGINEERING PROPERTIES

water and aroma barrier
excellent oil and grease resistance

PRESENTATIONS

flexible packaging

FOODS

perishable and semi-perishable foods

PROCESSING TECHNOLOGY

both blown
cast extrusion
extrusion coating

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life
Exploration Lignin R&D by Biome Bioplastics & Warwick University UK currently on work	lignin-based bioplastics	bio-based	fossil-based bioplastics	paper recyclable compostable
Michem® Coat 525 by Michelman USA 2020	functional coating	bio-based % NS	EVOH PVDC LDPE PE	paper recyclable compostable
Hydraban® 8000 by Michelman USA 2020	functional coating	bio-based % NS	EVOH PVDC LDPE PE	paper recyclable compostable

<table>
<tr><th>General properties</th><th>Processing technologies</th><th>Applications</th><th>References</th></tr>
<tr><td>Biome Bioplastics is now working with the Warwick team to develop methods to control the lignin breakdown process to determine whether these chemicals can be extracted in significant quantities.</td><td>blown film extrusion
thermoforming
injection molding</td><td>flexible packaging
rigid packaging</td><td>https://biomebioplastics.com/downloads/An_Exploration_of_Lignin.pdf</td></tr>
<tr><td>designed for maximum oil and grease performance
cold set gluable and approved for direct food contact
PH:4.5-6.5
Shelf Life: 6 months</td><td>extrusion coating
film lamination techniques</td><td>flexible packaging
rigid packaging</td><td>https://www.michelman.com/solutions/michem-coat/michem-r-coat-525/?utm_source=bpi-certified-coatings-2020-pr&utm_medium=press-release&utm_campaign=bpi-certified-coatings&utm_tags=|printing-and-packaging|sustainability|</td></tr>
<tr><td>excellent gluability
water repellency
printability
wax impregnated look to the finished sheet without sacrificing recyclability</td><td>extrusion coating
film lamination techniques</td><td>flexible packaging
rigid packaging</td><td>https://www.michelman.com/solutions/hydraban/hydraban-r-8000/?utm_source=bpi-certified-coatings-2020-pr&utm_medium=press-release&utm_campaign=bpi-certified-coatings&utm_tags=|printing-and-packaging|sustainability|</td></tr>
</table>

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life
NatureFlex™ 30NVO by Futamura UK 2019	wood-base cellulose	**bio-based** % NS	EVOH PVDC LDPE PE	paper recyclable compostable
Ingeo Propylester© by NatureWorks & Metalvuoto USA & Italy 2016	PLA	**bio-based** % NS	EVOH PVDC LDPE PE	paper recyclable biodegradable
Tetra Brik® Aseptic 1000 Edge with plant-based LightCap™ 30 by TetraPack Sweden 2016	sugarcane-based bioplastics	**>80% bio-based**	EVOH PVDC LDPE PE	recyclable biodegradable

General properties	Processing technologies	Applications	References
heatsealable film semipermeability to moisture good barrier to oxygen and mineral oils superb dead-fold Ovenable	extrusion coating film lamination techniques	flexible packaging rigid packaging	http://www.futamuragroup.com/Futamura/media/Futamura/Press%20Releases/Futamura-Press-Release_New-product-launch-30NVO-Final.pdf
better oxygen barrier than metallized biaxially oriented polypropylene (BOPP) heat-seal ability designed to move from complex multilayer laminates to simpler packaging solutions	extrusion coating film lamination techniques	flexible packaging	https://www.natureworksllc.com/News-and-Events/Press-Releases/2016/03-17-16-Metalvuoto-Ingeo-PLA-barrier-film
carrying the highest class of certification from TÜV for its use of renewable materials slimmer and taller profile: optimization of materials, weight, and space	final product sale	rigid packaging	https://www.tetrapak.com/sustainability/planet/responsible-sourcing/polymers

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life
ecovio® PS by BASF Germany ~2013	Ecoflex/ PLA/additives	**bio-based** % NS	EVOH PVDC LDPE PE	paper recyclable compostable
Skalax® XH11 by Seelution Sweden 2013	natural polysaccharides/ additives	**bio-based** % NS	EVOH PVDC LDPE PE	paper recyclable compostable
Skalax® XD by Seelution Sweden 2013	natural polysaccharides/ additives	**bio-based** % NS	EVOH PVDC LDPE PE	paper recyclable compostable

General properties	Processing technologies	Applications	References
good barrier properties against liquids, fat and mineral oil low coating thickness at high coating rates and process stability temperature stability up to 100°C sealing capacity comparable to PE good adhesion	extrusion coating film lamination techniques	flexible packaging rigid packaging	https://plastics-rubber.basf.com/global/en/performance_polymers/products/ecovio.html
transparent and colourless preserves the food aromas grease barrier to prevent staining of packaging recommended for oxygen barrier applications no fluorocarbons used	extrusion coating film lamination techniques	flexible packaging	https://seelution.se/products/renewable-barriers/
preserves the food aromas particularly suitable for applications where you need a grease barrier no fluorocarbons used	extrusion coating film lamination techniques	flexible packaging	https://seelution.se/products/renewable-barriers/

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life
Skalax® XH11-4 by Seelution Sweden 2013	natural polysaccharides/ additives	**bio-based** % NS	EVOH PVDC LDPE PE	paper recyclable compostable
CelluForce NCC® by CelluForce Canada 2012	Cellulose nanocrystals (CNC) from wood	**bio-based** % NS	EVOH PVDC LDPE PE	paper recyclable biodegradable
BIOMEEASYFLOW by Biome Bioplastics UK 2012	starch-based paper coating	**bio-based** % NS	EVOH PVDC LDPE PE	paper recyclable biodegradable compostable

General properties	Processing technologies	Applications	References
aromas in food preserved grease barrier to prevent staining of packaging no fluorocarbons used designed for applications where a high flexibility is required	extrusion coating film lamination techniques	flexible packaging	https://seelution.se/products/renewable-barriers/
reduce the permeability of films and materials allows the stable suspension of particles. high particle crystallinity imparts strength and hardness crystal aspect ratio and surface charges create unique viscosity and shear thinning properties	extrusion coating film lamination techniques	flexible packaging rigid packaging	https://www.celluforce.com/
plasticiser-free GM-free excellent grease barrier Excellent sealing qualities at low coatweights	extrusion coating film lamination techniques	flexible packaging rigid packaging	https://biomebioplastics.com/product-ranges/coating/

Trademark/Project by Company, Country year	Type of material	Feedstock	Substituted material	circular-life
C☆Film Starches by Cargill USA launch ns	starch-based paper coating	**bio-based** % NS	EVOH PVDC LDPE PE	paper recyclable
C☆iFilm™ Coating Starches by Cargill USA launch ns	starch-based paper coating	**bio-based** % NS	EVOH PVDC LDPE PE	paper recyclable

General properties	Processing technologies	Applications	References
excellent viscosity stability chlorine-free completely AOX-free	blown film extrusion blow molding	flexible packaging	https://www.cargill.com/bioindustrial/paper-and-packaging/c-film
has a better stability compared to traditional coating starch 25% higher starch utilization than your current process with classical coating starch Good barrier against carbon dioxide (CO2) and oxygen (O2) comparable to PET	blown film extrusion blow molding	flexible packaging	https://www.cargill.com/bioindustrial/paper-and-packaging/c-ifilm

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Chapter V.

BIOPLASTICS MARKET ANALYSIS

5.1 Bioplastics market context in food packaging

Bioplastics play an important and unique role in food packaging because they represent a fundamental key shift to a circular economy in this sector. Bioplastics have opened an enormous spectrum of opportunities to finish with plastic pollution from single use articles, and therefore must be contemplated in all state's management plans. Additionally, the definition of standards, labels, guidelines, and legislations are vital to run a sustainable change for food packaging system.

Bioplastics are also supported on bioeconomy concept, which describes the industrial use of renewable biological resources through the sustainability principle. Being more than the substitution of fossil resources and reducing GHG emissions, bioeconomy seeks to create new jobs, and get new consumers while avoiding risks for these groups (Spierling et al., 2018). At the moment, bioplastics market is worth 2 trillion euros in annual turnover and accounts for 22 million jobs in the EU (Filho et al., 2021).

This last chapter provides the latest update of the global bioplastic's market analysis for the food packaging sector. Furthermore, it is considered the major representative regions and state-of-the-art companies which currently develop bioplastics food packaging solutions. Likewise, the key success factors and challenges that currently face bioplastics in food packaging segment are analyzed, and the market analysis concludes with a summarized SWOT analysis.

5.1.1 Market size and projection

Plastics are a large family of polymers, traditionally derived from fossil resources characterized by having a broad range of properties and characteristics. Currently, plastic production represents approximately 4-8% of oil consumption globally and is expected to reach 20% by 2050 (Narancic et al., 2020; Acquavia et al., 2021).

Since wide scale production in the 1950s, their low cost coupled with a wide range of properties. Global plastic production has steadily increased from 15 million metric tonnes in 1964 to 359 million metric tonnes in 2018 with a projected 2-fold increase within the next 20 years (Narancic et al., 2020; Acquavia et al., 2021).

Polymers have entirely integrated themselves into modern daily life as evidently seen from the increased production and usage volume over past decades. But from this vast plastic market, unfortunately, less than 1% corresponds to bioplastics (Andrady, 2015; Habel, et al., 2018; Narancic et al., 2020; Hatti-Kaul et al., 2020; Acquavia et al., 2021; Filho et al., 2021).

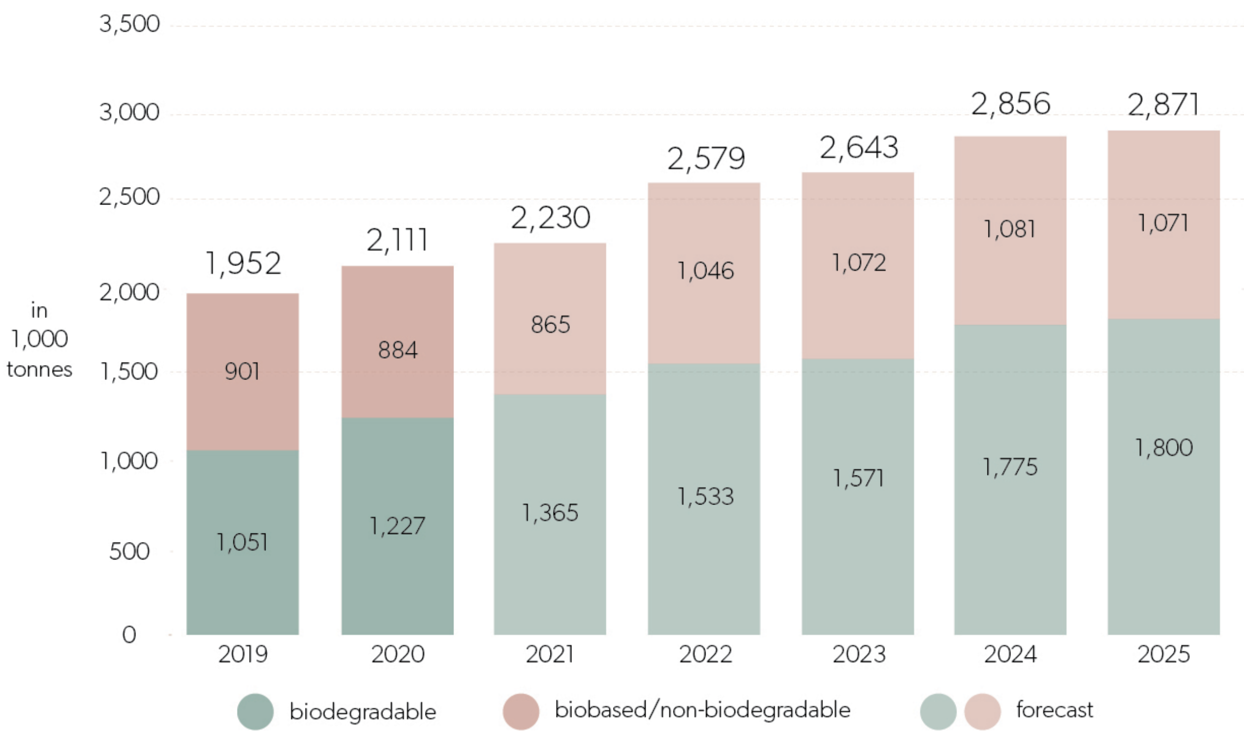
According to the latest market data compiled by European Bioplastics in cooperation with the Nova-Institute[1], global bioplastics production capacity was 2.11 million tonnes in 2020. However, due to the growing sensitivity towards a "green and circular economy" policy, the global bioplastics production capacity is expected to reach approximately 2.87 million tonnes by 2025

[1] Nova-Institute is a private and independent research institute focus on the transition of the chemical and material industry to renewable carbon. http://nova-institute.eu/

(European Bioplastics, n.d.; Acquavia et al., 2021). Even if bioplastic packaging is forecast to grow at a significantly higher rate than petrochemical-based polymers (Filho et al., 2021), its expected growth would remain below 1% of market share.

Among bioplastic's market segments, packaging remains the largest field of application since around 47% of the global bioplastic production (figure 38) is used in the packaging industry, including shopping bags and plastic bottles producers, and food packaging industry (Acquavia et al., 2021).

Figure 38

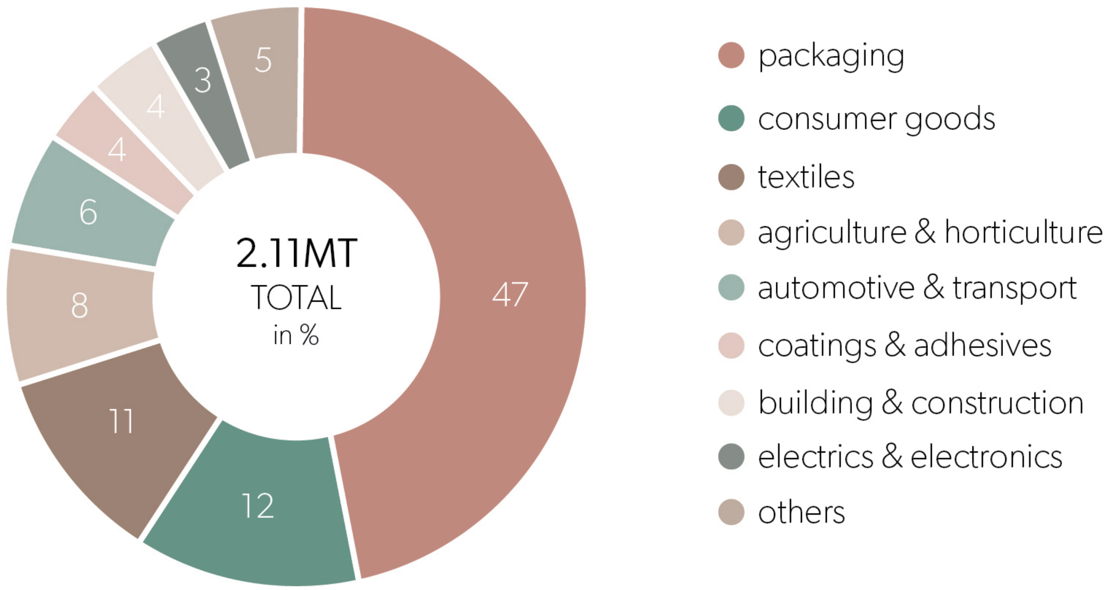


Global Production Capacities of Bioplastic

Source. (European Bioplastics & nova-Institute, 2020)
More information:www.european-bioplastics.org/market and www.bio-based.eu/markets

Food packaging represent approximately 60% of bioplastics global production (figure 39). The market for bioplastics is rapidly growing, and the packaging solutions, especially single-use food packaging, will have a great potential for the industrial application of bioplastics. In Europe, the demand for bioplastics in the packaging industry is expected to increase by more than 15% per annum over the next few years (Ceresana, 2015; Habel, et al., 2018).

Figure 39



Global Production Capacities of Bioplastics in 2020

Source. (European Bioplastics & nova-Institute, 2020)
More information:www.european-bioplastics.org/market and www.bio-based.eu/markets

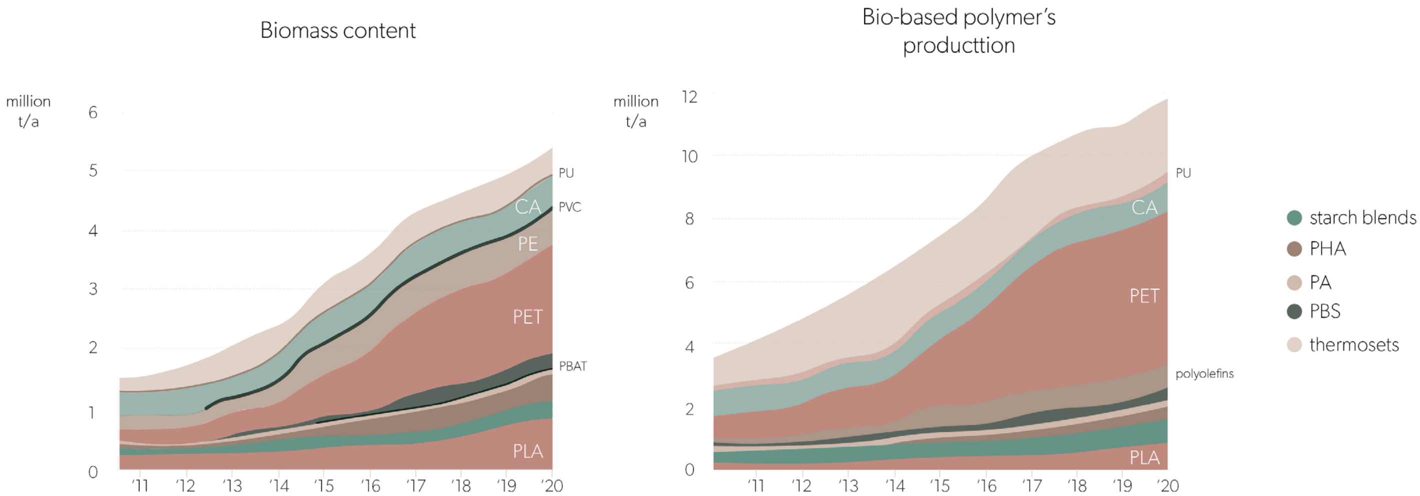
On the other hand, the food packaging market is also expected to grow. For instance, Europe is predicted to grow to a market volume of about 38.2 million tons in 2022. Moreover, it is a dynamically changing market where canning or metalized foils are increasingly replaced by transparent flexible packaging (Habel, et al., 2018).

Despite its low growth estimated (2.87MT by 2015), the potential of bioplastic production is especially promising considering the renewable feedstocks not used. Abundant biomass is available to be used as a raw material. Out of the 170 billion tons of biomass produced annually by nature, less than 4% is used by humans, mostly for food and wood-based industries (Andrady, 2015).

Bioplastic's market share and growth per type

Even though bioplastics market is still very low, during last 10 years have growth vertiginously (figure 40). With increasing environmental concerns and awareness of stress on fossil fuel, other studies estimate a compounded annual growth rate (CAGR) of 11.28% from 2019 to 2025 for bioplastic (Rai et al., 2021). Considering last year production-2.11MT (European Bioplastic & nova-Institute, 2020)- this means a bioplastic's production of 3.23 MT by 2025.

Figure 40

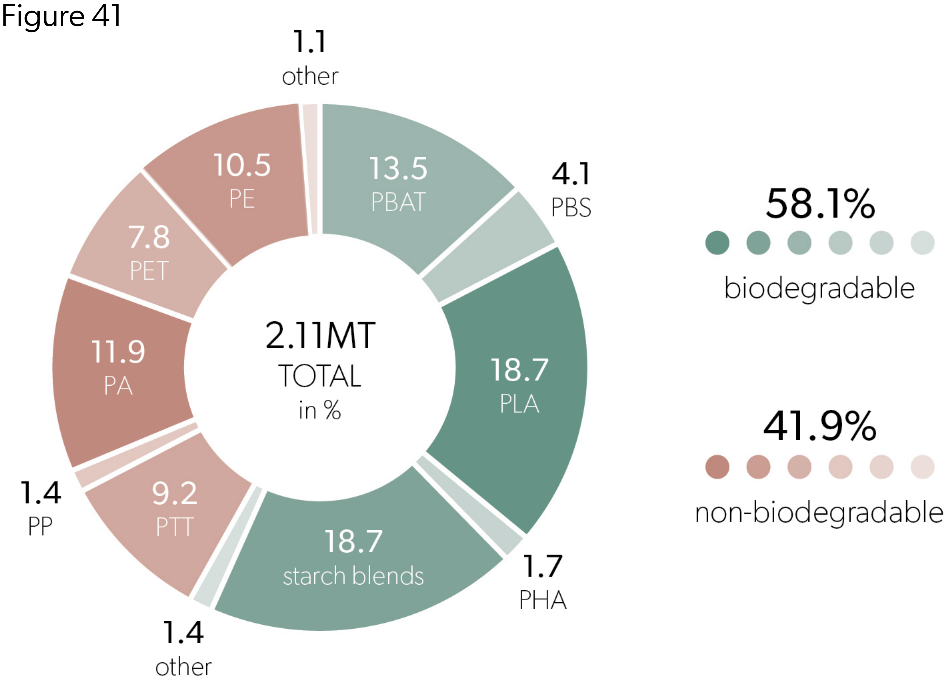


Biobased Bioplastics: Biomass Content and Production Evolution from 2011 to 2020

Source. (Luzi et al., 2019).

As the figure shows, biodegradable bioplastics represent the majority production capacity with almost 60% of the bioplastic market share. The participation of three principal contributors can be highlighted: starch-based bioplastics, PLA and PBAT. The production of biodegradable bioplastics is expected to increase to 1.8 million in 2025 especially due to PHA's significant growth rates and new investments for PLA production in the US and in Europe (European Bioplastic & nova-Institute, 2020).

Figure 41



Global Production Capacities of Bioplastics 2020 by Material Type

Source. (European Bioplastics & nova-Institute, 2020)
More information:www.european-bioplastics.org/market and www.bio-based.eu/markets

Among biodegradable bioplastics, starch-based polymers represented 18.7% of bioplastic's global production capacities in 2020 (Narancic et al., 2020; Rai et al., 2021; European Bioplastics, n.d). Along with starch blends, PLA report similar market share, being the main contributors to the growth of biodegradable bioplastics. PLA is expected to see an 8% increase in production from 293 290 tonnes in 2019 to 317 000 in 2024. PBAT also contributes strongly to this advance with a 13.5% of participation (Narancic et al., 2020; Rai et al., 2021; European Bioplastic & nova-Institute, 2020).

PHA's show a market share of 1.7% (% wt of 2.11 million tonnes of bioplastics produced), but although, PHA's current market share is very small, PHAs is expected to see a 6.3-fold increase in global production from 25 320 tonnes 2019 to 159 700 tonnes by 2024 (Narancic et al., 2020; European Bioplastic & nova-Institute, 2020).

Cellulose is the most abundant natural biopolymer on earth sourced predominantly from trees and cotton, nonetheless only approximately 1.5×10^{12} tonnes is produced annually (Narancic et al., 2020), but was not found an estimation of cellulose-based bioplastic production. However, cellulose and its derivates represent a huge opportunity for a sustainable food packaging industry, especially for films and coating as have been seen in chapter IV.

On the other hand, biobased and non-biodegradable bioplastics represent near 42% of bioplastic's global market (European Bioplastic & nova-Institute, 2020). Some spectacularized literature classified this group as the *New Economy* of bioplastics market, naming them "Novel bioplastics" or "Drop-Ins" (Spierling et al., 2018), because even if they are biobased can perform similar to their counterpart fossil-based plastic. Additionally, due to their properties and currently high demand, they promise to be an important material category in food packaging applications. The next table 17 represents this classification.

Table 17. *Framework of Bioplastics*

		Durable	Biodegradable
Biobased	Old Economy	e.g. Cellulose Acetate	natural rubber Linoleum i.a
Biobased	New Economy	bio-PE bio-PA bio-PUR/PU i.a.	PHAs PLA Starch blends i.a.
Fossil-based		PE PP PVC i.a.	PBAT PBS PCL i.a.

Note. Old Economy bioplastics indicate previous materials, that were in use before fossil-based plastics had even been developed, and still exist on the market today (e.g., rubber, cellophane, viscose, celluloid, cellulose acetate, linoleum). Cellulose is part of this group; its research development is considered part of the state-of-art bioplastics for food packaging applications.
Source. (Spierling et al., 2018).

Among them, bio-PE and its derivates such as bio-LDPE, bio-LLDPE and bio-HDPE; bio-PET, and PEF, are the most important for food packaging applications (Andrady, 2015; Spierling et al., 2018).

Nevertheless, these drop-in solutions are predicted to slightly decrease even further to just over 37% by 2025 (around 1 million tonnes) as the forecast for biodegradable plastics production shows a higher level of growth (European Bioplastic & nova-Institute, 2020).

Production capacities for biobased PET continue to decline as they have not been nearly meeting the predicted rates from previous years. Instead, the focus has shifted to the development of PEF, which expected to notably enter the market in 2023 (European Bioplastic & nova-Institute, 2020).

5.1.2 Customer perspective

Awareness and education play a crucial role in diminishing plastic pollution especially of daily products such as disposable food packages. Education, cultural, and behavioral changes are essential to promote sorting and prevent moral hazards such as littering (Kawahima et al., 2019).

Consumer perspective, and attitudes are vital to fulfill the circular economy of food packaging made of bioplastics. In fact, at the purchase phase and at the point of end-of-life disposal, consumers play a key role in the life cycle of food products and its packaging (Filho et al., 2021).

Fostering a culture of circularity in manufacturers is intrinsically linked with buyer's choices. Moreover, the design measures adopted for recycling are only effective if the consumer makes the right decision at the purchase and disposal phases (Filho et al., 2021).

Additionally, biodegradable plastics represent a great opportunity since they can be disposed of and recycled as organic matter. However, their benefits can be effective only if combined with an accurate consumer awareness campaign (Filho et al., 2021).

Furthermore, as the term sustainability is gaining importance for evermore consumers, the demand volume of food packaging solutions made of bioplastics will experience strong growth rates with the probability to be higher than the estimated. This also includes packaging solutions made of recycled plastics (Habel, et al., 2018; Ceresana, 2015).

From the functional packaging side, consumers expect lightweight and practical packaging, assuring a long shelf life and allowing them to visually inspect the product. Furthermore, they prefer complete information about the packaged product and an appealing design (Ceresana, 2015; Habel, et al., 2018). Consumers are currently better informed and wish to have as many details about the product as possible, such as the origin, manufacturing conditions, freshness, atmospheric composition inside the packaging, ingredients, additives, etc (Ceresana, 2015). Therefore, in case of bioplastics, is essential demonstrates this information to capture consumer awareness and guide them to the right final designed disposition.

5.2 Representative regions and companies

This section provides the most important companies on bioplastic market which have dedicated trademarks for food packaging applications. Among them are large transnational companies as well as smaller start ups in its beginnings. Table 18 summarizes a market research of 152 companies which currently are present in the bioplastic market, however only 67 already have bioplastics which meet food and beverage packaging requirements. The present analysis of these bioplastics' food packaging producers is the result of a specialized research done from the literature obtained and further individual investigation of each company official websites (Smith & Scion, n.d.; Bioplastics News, n.d.; Fapesp, 2012; Andrady, 2015; Luzi et al., 2019 Rai et al., 2021; Acquavia et al., 2021).

It is relevant to mention that among the companies investigated, 52% are based in Europe, and 22% in North America (USA and Canada), nevertheless, USA is the country with a greater number of companies specialized in this sector.

Table 18. The Most Representative Bioplastics Producers Specialized in Food Packaging Applications

Company	Country Headquarter	Feedstock	Type of Bioplastic Produced	Bioplastic circular-life	reference
Agrana	Austria	bio-based	starch-based	compostable	https://www.agrana.com/produkte/alle-produktportfolios/staerke-portfolio/produkte-fuer-technische-anwendungen/biobasierte-kunststoffe
Anellotech	USA	bio-based agri-waste, wood, plastic-waste	bio-PET	recyclable	https://anellotech.com/
Archer Daniels Midland (ADM)	USA	bio-based	PEF	recyclable	https://www.adm.com/sustainability/innovating-sustainable-materials
Arctic Biomaterials	Finland	bio-based	PE PP	biodegradable	https://abmcomposite.com/technical-produc
Arkema	France	bio-based	R&D	R&D	https://www.arkema.com/global/en/arkema-group/innovation/natural-resources-management/bio-based-products/
Ava Biochem	Switzerland	bio-based	PEF	recyclable	https://ava-biochem.com/
Avantium	Netherlands	bio-based	PEF	recyclable	https://www.avantium.com/
BASF	Germany	bio-based	PEF PLA	recyclable	https://plastics-rubber.basf.com/global/en/performance_polymers/products/ecoflex.html
Braskem	Brazil	bio-based sugarcane	bio-HDPE bio-LDPE bio-LLDPE	recyclable	https://www.braskem.com.br/imgreen/home-en

Company	Country Headquarter	Feedstock	Type of Bioplastic Produced	Bioplastic circular-life	reference
Biobent	USA	bio-based soy, canola, agave, algae	bio-PP bio-PE bio-PBS bio-PS PHA PLA	recyclable biodegradable(PLA)	https://www.biobent.com/price_list
Biologiq	USA	bio-based potatoes, corn, cassava	bio-LDPE bio-LLDPE bio-HDPE bio-PS bio-PP bio- PBAT	recyclable	https://www.biologiq.com/
Biome Bioplastics	UK	bio-based potato,corn	bio-LDPE	biodegradable compostable	https://biomebioplastics.com/product-ranges/
Biomer	Germany	bio-based	PHB	biodegradable compostable	https://www.biomer.de/IndexE.html
Bosk Bioproducts	Canada	bio-based	PHA cellulose-based	biodegradable compostable	https://www.bosk-bioproducts.com/compostable-bioplastics.html
Biotec	Germany	bio-based potato	starch-based	compostable biodegradable recyclable	www.biotec.de
Bio-Mi	Croatia	bio-based	n.s	biodegradable compostable	https://bio-mi.eu/
Bio-on	Italy	agri-waste	PHA	biodegradable compostable	http://www.bio-on.it/
Carbiolice	France	bio-based sugarcane, corn	PLA	biodegradable compostable	https://www.carbiolice.com/en/evanesto-inside/

Company	Country Headquarter	Feedstock	Type of Bioplastic Produced	Bioplastic circular-life	reference
Cardia Bioplastics	Australia	bio-based corn sugarcane	starch-based	compostable biodegradable recyclable	www.cardiabioplastics.com
Cargill	USA	bio-based	starch-based	recyclable	https://www.cargill.com/bioindustrial
CelluForce	Canada	wood-based	Cellulose nanocrystals (CNC)	biodegradable	https://www.celluforce.com/
Cereplast	USA	bio-based corn, potato	starch-based	biodegradable compostable	https://www.cereplast.com/bioplastics/
Compostpack	Colombia	bio-based corn	starch-based	biodegradable compostable	https://www.compostpack.com/index.html
Corbion	Netherlands	agri-waste sugarcane, corn, sugar beet, cassava	PEF PLA	recyclable(PEF*) biodegradable compostable	https://www.total-corbion.com/
Danimer Scientific	Georgia	bio-based canola soy	PHA PLA	biodegradable compostable	https://danimerscientific.com/
Eastman	USA	bio-based	cellulose-based	n.s	https://www.eastman.com/Markets/Food-Beverage-Packaging/Pages/Food-Packaging.aspx
Eranova	France	bio-based	algae-based	compostable biodegradable recyclable	https://eranovabioplastics.com/

Company	Country Headquarter	Feedstock	Type of Bioplastic Produced	Bioplastic circular-life	reference
FKuR	Germany	wood-based	bio-PE	recyclable	https://fkur.com
Futamura	UK	biobased wood pulp	cellulose-based	biodegradable compostable	http://www.futamuragroup.com/
Futerro	France & Belgium	biobased sugar beet, sugar cane, wheat, maize, cellulose.	PLA	compostable biodegradable recyclable	http://www.futerro.com/endoflifemanagement.html
GC Innovation America	USA	biobased	PLA bio-PBS bio-PET	compostable recyclable	https://productsandsolutions.pttgcgroup.com/products/packaging_detail/Mw==
Gema Polimer	Turkey	biobased	starch-based	biodegradable compostable	https://www.gemabio.com/
Global Biopolymers	Thailand	agri-based	PLA	biodegradable	http://www.globalbiopolymers.com/index.php
Goglio	Italy	biobased	cellophane	biodegradable	https://www.goglio.it/en/
Grabio	Taiwan	biobased	starch and cellulose-based	biodegradable compostable	http://grabio.com.tw/index_esp.asp
Green Dot Bioplastic	USA	biobased	starch-based	biodegradable compostable	https://www.greendotbioplastics.com/

Company	Country Headquarter	Feedstock	Type of Bioplastic Produced	Bioplastic circular-life	reference
Green Science Alliance	Japan	biobased wood waste-based	PLA/ nanocellulose starch-based	biodegradable compostable	https://www.gsalliance.co.jp/en/
Kaneka	Japan	biobased	PHBH	biodegradable compostable	https://www.kaneka.co.jp/en/business/material/nbd_001.html
Lactips	France	biobased	casein protein-based	biodegradable compostable	https://www.lactips.com/?lang=en
Lyspackging	France	biobased sugarcane	n.s	compostable recyclable	https://lyspackaging.com/?lang=en
Maistic	Denmark	plant-waste	n.s	biodegradable compostable	https://www.maistic.com/en/
Metabolix	USA	biobased sugarcane	PHA	biodegradable compostable	https://www.yield10bio.com/Corporate-Overview
Naturetec	USA	biobased	PLA	biodegradable compostable	https://www.naturtec.com/products/
Natureworks	USA	biobased cassava, corn starch, sugar cane, beets. R&D: Lignocellulosics: bagasse, wood chips, switch grass or straw	PLA	compostable recyclable	https://www.natureworksllc.com/
Novamont	Italy	biobased	starch/PCL	biodegradable compostable	https://www.novamont.com/eng/

Company	Country Headquarter	Feedstock	Type of Bioplastic Produced	Bioplastic circular-life	reference
Nurel	Spain	biobased	n.s	compostable	https://biopolymers.nurel.com/en/nurel/inzea
Oimo	Spain	biobased	PLA	biodegradable	http://oimo.co/
Omya	Switzerland	biobased	PHA PLA	biodegradable compostable	https://polymers.omya.com/
Plantic Technologies	UK	starch-based	PHA	recyclable	http://www.plantic.com.au/product/plantic-flexible
Rodenburg Biopolymers	Netherlands	biobased potato	starch-based	biodegradable	http://biopolymers.nl/biopolymer/
Roquette	France	biobased	PBS	biodegradable	https://www.roquette.com/industries/performance-materials/polyesters-pbs
Seelution	Sweden	biobased	natural polysaccharides/additives	recyclable	https://seelution.se/
Synbra Thecnology	Netherlands	biobased sugarcane, beets	bio-EPS	recyclable biodegradable reusable	https://www.synprodo.com/what-is-biofoam/
Nature2Need	Germany	biobased	n.s	biodegradable compostable	https://nature2need.com/biomaterials/

Company	Country Headquarter	Feedstock	Type of Bioplastic Produced	Bioplastic circular-life	reference
Spectalite	India	agri-waste agri-based	biocompounds	biodegradable compostable	https://www.spectalite.com
Stora Enso	Finland	biobased	cellulose foam	biodegradable recyclable	https://www.storaenso.com/en/products/bio-based-materials/cellulose-foam
Sulzer	Switzerland	biobased	PLA	biodegradable	https://www.sulzer.com/en/applications/oil-gas-chemicals/downstream/bioplastics
Taghleef Industries	USA	biobased	bio-PP	recyclable	https://www.ti-films.com/en/sustainability/biobased-PP
Tecnaro	Germany	biobased lignin, natural resins, natural waxes, natural oils, natural fatty acids, cellulose	bio-PET bio-PA bio PE starch-based PLA	biodegradable recyclable	https://www.tecnaro.de/arboblend-arbofill-arboform/
Terraverdae	Canada	biobased	PHA	biodegradable compostable	https://terraverdae.com/
TIPA	Israel	biobased	n.s	compostable	https://tipa-corp.com/portfolio/
Total	France	biobased sugarcane, corn, sugar beet, cassava	PLA	biodegradable compostable	https://www.total-corbion.com/
Treemera	Germany	biobased	PEF	recyclable	https://www.treemera.com/#applications

Company	Country Headquarter	Feedstock	Type of Bioplastic Produced	Bioplastic circular-life	reference
Venvirotech	Spain	organic-waste	PHA	biodegradable	https://www.venvirotech.com/en/
Woodly	Finland	wood-based	cellulose-based	recyclable	https://woodly.com/woodly/

Note. This list does not contemplate intermediate producers or companies producing bioplastics additives. Some companies presented also provide biodegradable bioplastics based on petrochemical resources, nevertheless, the information gave is based in its biobased trademarks.

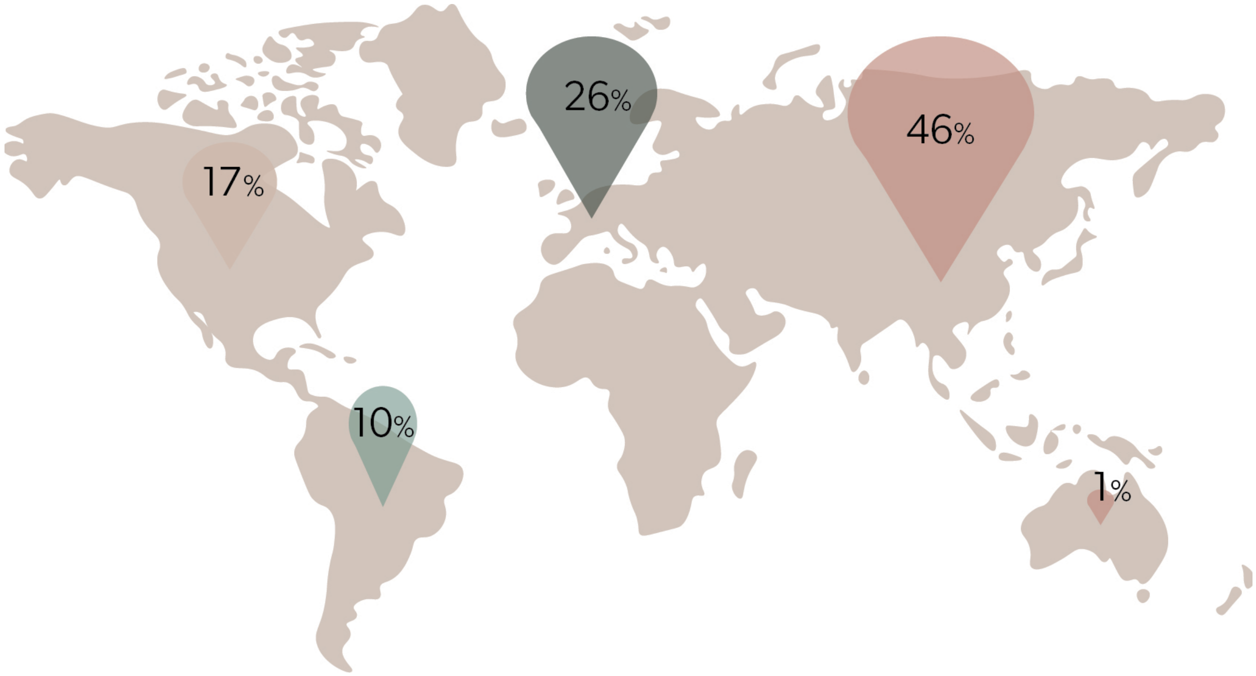
Source. (Smith & Scion, n.d.; Bioplastics News, n.d.; Fapesp, 2012; Andrady, 2015; Luzi et al., 2019; Rai et al., 2021; Acquavia et al., 2021)

Europe strengthened its position as one of hub for the entire bioplastics industry once again; it ranks highest in the field of research and development and is the industry's largest market worldwide. By now, one-fourth of the global bioplastics production capacity is in Europe (European Bioplastic & nova-Institute, 2020; Filho et al., 2021). This share is predicted to grow up to 27% by 2023, which will be supported by recently adopted policies in several European Member States, such as Italy and France (Acquavia et al., 2021).

However, with a view to the actual production of bioplastics and regional capacity development (figure 42), Asia continues to be in the lead. In 2020, 46% of bioplastics were produced in Asia, and the region will remain to be the central production hub over the next five years (European Bioplastic & nova-Institute, 2020; Acquavia et al., 2021).

Furthermore, the main driver for bioplastic's increase is the increasing demand for biodegradable polymers in emerging economies such as India, Brazil, and China (Andrady, 2015; Narancic et al., 2020). Nonetheless, the market trends indicate that Asia-Pacific region is expected to have the fastest growth due to the easy availability and low cost of raw material required to produce bioplastics (Rai et al., 2021).

Figure 42



Global Production Capacity of Bioplastics by Region

Source. (Fapesp, 2012; European Bioplastics & nova-Institute, 2020)
More information:www.european-bioplastics.org/market and www.bio-based.eu/markets

The *Bio-Based Polymers Producer Database* frequently updated by the Nova-Institute, exhibits that Europe's running situation in creating biobased polymeric matrices is restricted to a few polymeric matrices. So far, the European community has determined a solid role, principally in starch blending materials and is expected to continue in this specific sector for the following few years (Luzi et al., 2019).

On the other hand, in North America production is estimated to slightly increase from 17% in 2020 to 18% by 2025 (European Bioplastic & nova-Institute, 2020).

5.3 Bioplastics' market: Key success factors and challenges

5.3.1 Biodegradability and mechanical properties

Biodegradability has been a valuable feature for bioplastics providing a solution to reducing plastic waste by decomposing into CO_2 (or CH_4) and H_2O (Hatti-Kaul et al., 2020). In fact, several standards can certify if a bioplastic can be compostable at a residential level, industrial, or if they can follow an anaerobic degradation. There are even bioplastics that can biodegrade in soil or marine environments (section 2.2.3). Although these standards are measured in specific conditions, and take into account the current composting technologies, they also depend on the specific municipality composting facilities if bioplastics are enabled to biodegrade there. Therefore, an alignment between these parts must be settled before the official issue of the corresponded labels.

Nevertheless, biodegradable bioplastics are susceptible to microorganisms, resulting in good biodegradability, but could also be disadvantageous for food preservation product for their inherent low barrier properties (Habel, et al., 2018).

In addition, bioplastic's practical use is hindered by their relatively poor thermal and mechanical properties (Hatti-Kaul et al., 2020). Since numerous biopolymers show water affinity, their barrier and mechanical characteristics are subject to the humidity and ambient atmosphere. The exposure may decrease their overall performance and the quality of packages compared with petroleum-based polymers (Luzi et al., 2019).

Furthermore, molecular weight, physical properties (crystallization phenomenon and crystallization degree), visco-elasticity, and rheological characteristics may induce disadvantages, several modifications or adjustments during the processing steps are necessary to modulate the final performance (Luzi et al., 2019).

On the other hand, even though significant development has been made to improve bioplastics properties to fit food packaging functions, there is still a need for further advancements. Almost all the practiced processes nowadays also use traditional petrochemical and non-compostable ingredients (Luzi et al., 2019).

5.3.2 New biopolymeric blends

As shown in previous chapters, to improve bioplastics properties, these can be modified, through new polymeric blends combining two or more different polymeric matrices or fillers, at the micro/nanoscale level (Luzi et al., 2019; Hatti-Kaul et al., 2020). These solutions are an opportunity to expand the bioplastic's market in food packaging applications.

In many cases, the different "bioplastic formulations" need to be blended with additives to optimize some properties of the materials, such as thermal instability, high water vapor, brittleness, and low melt strength. Plasticizers, like glycerol, for example, are often required to improve the processability and mechanical properties (Acquavia et al., 2021).

Therefore, safer additives, co-polymers, and other chemicals useful for biobased polymeric blends are necessary to meet the required food packaging performance. Methods of green chemistry and design will be vital in this arena (Álvarez-Chávez et al., 2012).

5.3.3 Sustainability of Bioplastics

With an evolving biobased bioplastic market and application range, its sustainability has come to the fore and is questioned by different stakeholders (Spierling et al., 2018).

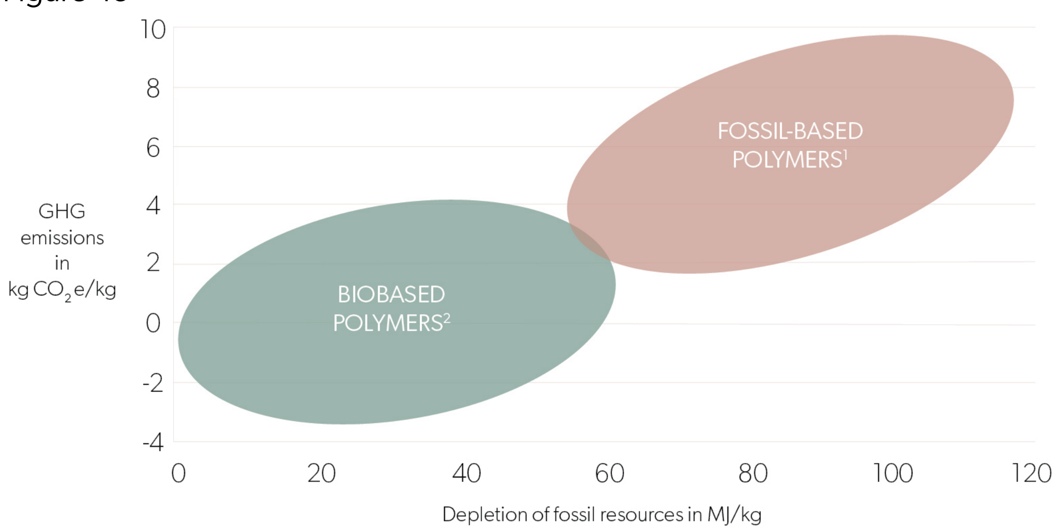
The savings in primary energy and avoided CO2 emissions in using bio-based feedstock compared to conventional petroleum feedstock are significant. Cradle-to-grave life cycle assessment (LCA) of several bio- and fossil-based plastics has shown the production and use of biobased plastics to be generally advantageous in terms of saving fossil resources and reducing GHG emissions (Andrady, 2015; Spierling et al., 2018; Hatti-Kaul et al., 2020).

Despite different scopes and boundaries of these cradle-to-gate studies done, the trend is clear. Biobased can, under certain conditions, decrease carbon dioxide emissions and potentially act as a carbon sink throughout their life cycle (Ellen MacArthur Foundation, 2016).
For biobased plastics, plants capture carbon dioxide from the atmosphere as they grow and this carbon is then harnessed in the polymer (Ellen MacArthur Foundation, 2016).

For example, significant savings of 40-50% less nonrenewable energy use and 45-55% less GHG emissions have been reported to produce PEF compared with the cradle-to-grave impact of PET (Hatti-Kaul et al., 2020). The GHG emission for conventional PET in a cradle-to-grave LCA estimate was 3.36 (kg equiv. CO2/kg plastic). Whereas the corresponding number for bio-based PET was 2.34-2.67 (kg equiv. CO2/kg plastic) depending on feedstocks used (Andrady, 2015).

A comparison of fossil-based and biobased polymers in terms of their greenhouse gas emissions and depletion of resources is shown in figure 43.

Figure 43



Environmental impacts of fossil-based and biobased polymers

Source. (Ellen MacArthur Foundation, 2016)

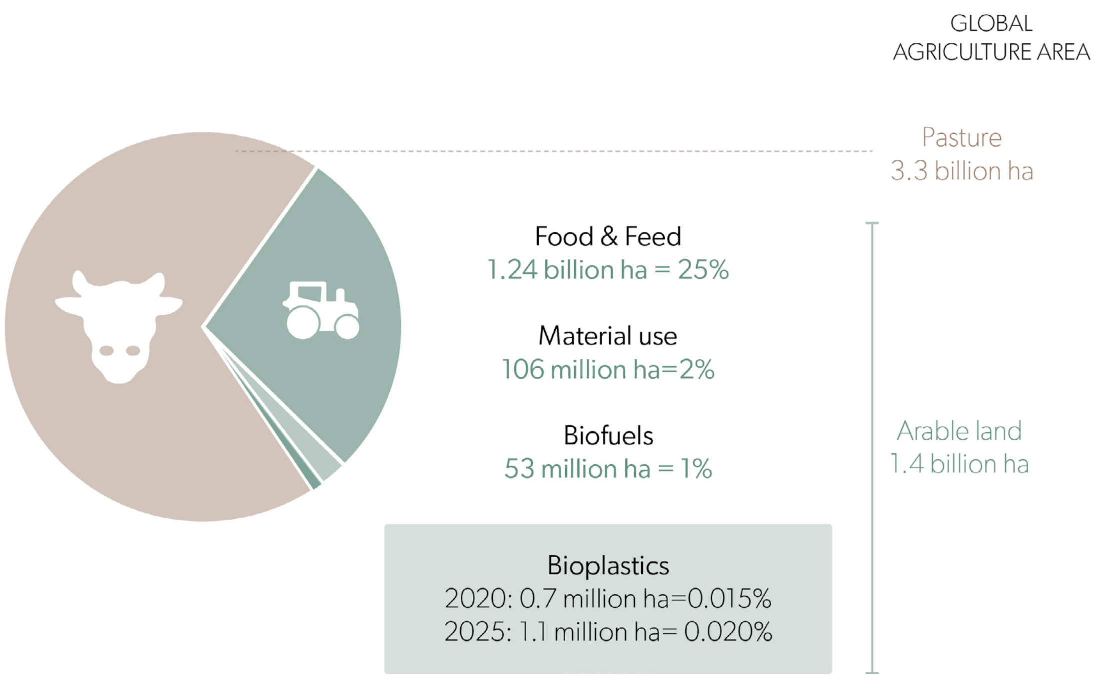
However, the externalities associated with biobased plastic production that includes intensive agriculture to produce the biomass, can exceed those for conventional plastics. The environmental impact of biobased plastics production is typically dominated by primary agricultural feedstock production in terms of fossil fuel, fertilizer, and pesticide inputs resulting in higher GHG emissions (Álvarez-Chávez et al.,2012; Andrady, 2015; Hatti-Kaul et al., 2020).

Studies have reported the acidification of soil, ecotoxicity, eutrophication, ozone depletion, and PM 2.5 particulates to be significantly higher for PLA compared to conventional PE and PET. Similar results have been reported for PHA (Álvarez-Chávez et al.,2012; Andrady, 2015; Van den Oever et al., 2017; Hatti-Kaul et al., 2020).

The water footprint of biobased plastics is usually quite significant due to most commercial biobased plastics. Their monomers are produced from first-generation[2] agricultural feedstocks (e.g., cornstarch and sugar cane) that are easy to process compared to other biomass feedstocks (Hatti-Kaul et al., 2020).

In addition, there are some concerns about a possible issue of land use triggered by the production of biobased plastics made of agricultural feedstocks (Álvarez-Chávez et al., 2012; Hatti-Kaul et al., 2020). Nevertheless, current arable land use and estimations do not mark a competition (figure 44).

Figure 44



Land Use Estimation for Bioplastics 2020 and 2025

Source. (nova-Institute, 2020; FAO Stats, 2005-2014 & Institute for Bioplastics and Biocomposites, 2019 as cited in European Bioplastics, 2020)

More information:www.european-bioplastics.org

Most of bioplastic's companies produce sustainable bioplastics made from plant-based renewable resources, like corn, potatoes, and wheat (Acquavia et al., 2021). The land used to grow the renewable feedstock to produce bioplastics amounted to approximately 0.7 million hectares in 2020. It represents 0.015% of the global agricultural area of 4.8 billion hectares of which 94% is used for pasture, feed, and food (Andrady, 2015; European Bioplastic & nova-Institute, 2020).

Nevertheless, despite the market growth predicted in the next five years, the land use share for bioplastics will only slightly increase to 0.02%. This forecast eliminates the possibility of a competition between the renewable feedstock for food, feed, and the production of bioplastics (Andrady, 2015; European Bioplastic & nova-Institute, 2020).

5.3.4 Second generation of biomass feedstock

The second generation of bio-refineries feedstocks corresponds to non-food biomass (Smith & Scion, n.d.). The biomass used can be food crops (corn and soy), non-food crops (switchgrass), or agricultural waste. Using the first category that include corn, sugarcane, sugar beet, potato, cassava, rice, wheat, and sweet potato, will directly or indirectly compete with land use for food production (Andrady, 2015), making it a valid concern on biobased plastics technology. While the percentage of arable land needed to support this growth is claimed to be the minimum of the global arable land, adverse regional impacts of water use associated in the future cannot be ruled out (Álvarez-Chávez et al.,2012; Andrady, 2015).

Alternative non-food sources of biomass are needed to support plastics' growth. Efforts are being made to convert agricultural waste into PLA and other biobased plastics. A second generation biomass feedstock that is indenpednt of food crops based on agricultural waste would lend considerable impetus to future growth in bioplastics (Andrady, 2015).

The carbon footprint of the agricultural biomass such as jute, banana peels, corn stalks, vegetable waste, wood chips, grain husks, stubble etc. can be reduced substantially by redirecting the waste towards the production of bioplastics. The raw agro-waste is converted into usable biomaterial by facilitating their microbes' breakdown in a fermentation chamber under specific environmental and nutrition conditions. Natural polyesters like PLA, PHA, and PHB have also been isolated by mass producing microorganisms that store these polyesters in the intracellular inclusions. In both cases, the physiochemical properties of the monomer and the macromolecule made by this process are affected by the microbial strain and the growth medium being used (Rai et al., 2021).

Since, the bioplastics use monomers sourced from agricultural waste, the carbon footprint for both the raw and the finished product is reduced. For instance, the straw leftovers from paddy fields can be used to produce biobased plastics. Under-developed and developing countries produce enormous quantities of crop waste that is incinerated despite their potential for valorization due to the lack of alternatives. Rice straw obtained post-harvest can be used to extract cellulose polymer and fabricating blended biopolymeric membranes for food packaging and preservation. Industrial stakes in the project will benefit the farmers, enrich society, and safeguard the environment (Rai et al., 2021).

The most recent research concerning bioplastic production focuses on by-products and waste materials of food industries. According to the Food and Agriculture Organization (FAO) of the United Nations, every year, an estimated 1.3 billion tons of food is wasted globally from all stages of the food supply chain including post-production, handling/storage, manufacturing, wholesale/retail, and consumption. Since food waste (FW) landfilling yields undesirable results, such as greenhouse gas (GHG) emissions and groundwater contamination, their valorization through bioplastics production could help overcome their disposal problem by renewable sustainable processes. In addition, the production of value-added products while reducing the volume of waste is expected to decrease the production cost of biodegradable plastics, e.g., compared to conventional routes of production using overpriced pure substrates (Acquavia et al., 2021).

It is often used as a substrate for bacterial fermentation to obtain natural polyesters, namely PHA and PLA. When used to produce PHAs, food waste is a prime candidate for an inexpensive carbon source, due to its widespread availability and the potential to solve significant waste problems. In this case, physical, thermo-chemical, and biological pre-treatments of the FW are requested. A preliminary liberation of monomers from the FW (e.g., lignocellulosic components) with increasing accessibility of proteins, lipids, and polysaccharides (e.g., starch and cellulose), for subsequent enzymatic hydrolysis and fermentation, are essential. After the pre-treatment, the FW is ready for fermentation step in presence of bacteria, by using several cultivation strategies (Acquavia et al., 2021).

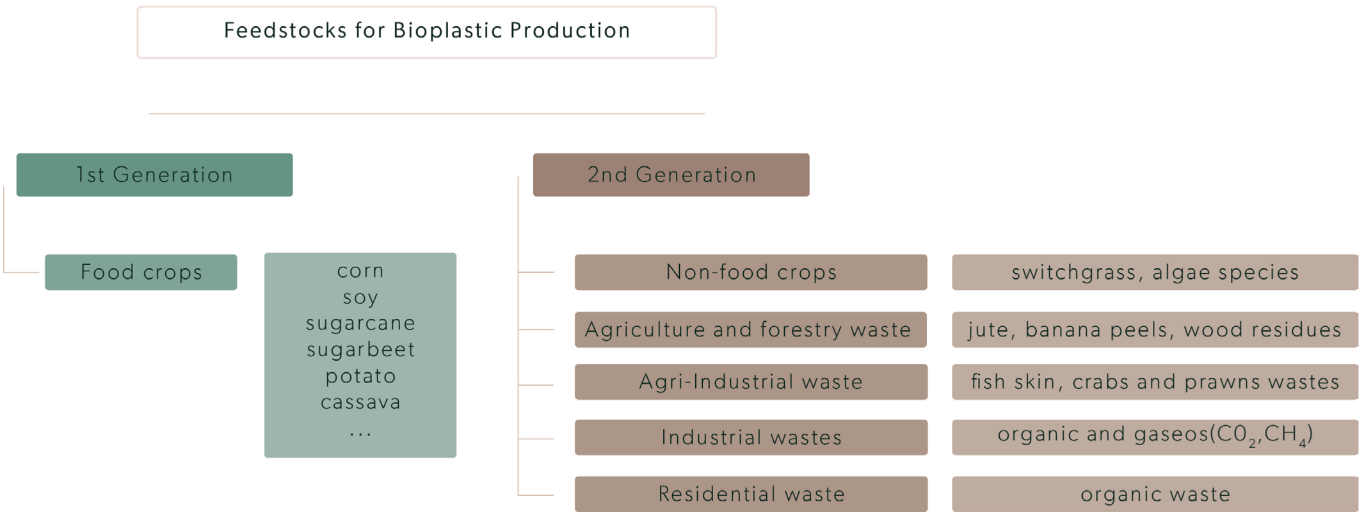
FW valorization can create opportunities to produce new valuable bioplastics, which represent an eco-friendly alternative to conventional petroleum-based plastics. Moreover, they could create positive synergies between industry and the agri-food sector, with considerable advantages for environmental pollution.

Currently, there are companies that exploit waste as feedstock for bioplastics production; an example is NaturePlast. Since 2015, NaturePlast has been producing and marketing a range of biocomposites consisting of by-products and plant fibers (such as hemp), sourced mainly from the French territory. The objective is to incorporate by-products or local waste materials in different polymers to ensure a circular economy and the reclamation of waste materials (Acquavia et al., 2021).

Additionlly, research on the utilization of industrial wastes, both organic and gaseous (syngas, CO_2, and CH_4), as the future feedstocks for non-fossil products has gained momentum. Production of PHA from diverse biomass streams, municipal wastewater, CO_2, and CH_4 is an important case, and the transformation of CO_2 and CH_4 provides further benefits of utilizing GHGs to form useful products (Hatti-Kaul et al., 2020).

Utilization of by-products and waste flows as raw materials or integrating production in a biorefinery would provide a dramatic ecological advantage and reduce pressure on arable land and water. Wood and other lignocellulosic residues from agriculture and forestry are expected to be more sustainable alternatives because they are a major source of polysaccharides and lignin in nature for providing both aliphatic and aromatic chemical platforms for the chemical and material industries (Hatti-Kaul et al., 2020). Next figure 45 sumarizes the diverse feedstocks streams for bioplastics production.

Figure 45



Feedstocks for Bioplastic Production

Source. Álvarez-Chávez et al.,2012; Andrady, 2015; Hatti-Kaul et al., 2020; Acquavia et al., 2021; Rai et al., 2021

5.3.5 Collection, labelling and recycling

One important challenge for bioplastics is the undeveloped specialized recycling streams (Hatti-Kaul et al., 2020). As shown in chapter III, due to bioplastics' similar look to conventional plastics, it is easy to contaminate their existing recycling system if not labeled with proper instructions (Andrady, 2015; Hatti-Kaul et al., 2020; Van den Oever et al., 2017).

On the other hand, biodegradable plastics are not a solution to littering problems. For instance, when lost at sea or when only partly recovered from the land, marine or soil biodegradable plastics would at least result in a lower risk of harmful consequences than if they fail to break down at all. Nonetheless, a certified claim such as OK biodegradable MARINE or OK biodegradable SOIL could stimulate consumers to leave a certified product behind in the environment. Therefore, a clear distinction should be made between certification of the claim and authorization to communicate about this certification (Van den Oever et al., 2017).

Collection and sorting, which starts with consumers and their behavior, largely determine the efficiency of waste management systems. To facilitate consumers to choosing the right route of disposal for food packaging waste, clear labels and logos must indicate the preferred disposal route (Van den Oever et al., 2017). This will highly influence the collection and sorting of bioplastics, and the quality of final compost or recycled plastics (Kawashima et al., 2019).

Despite the disadvantages discussed, the use of bioplastics has increased considerably over the past decades. But to achieve sustainable large scale production, use and management of bioplastics, proper standards and guidelines need to be established. Therefore, work with governments and municipalities to put in place adequated infrastructure is mandatory. Only in this manner bioplastics can be successfully composted and/or recycled and efficiently separated from petroleum-based recycling streams if is necessary(Álvarez-Chávez et al.,2012).

5.3.6 Economic feasibility

While bioplastics offer several advantages over their nonbiodegradable counterparts, the disadvantages associated with their potential applications should also be considered. Though bioplastics reduce the stress on fossil fuel consumption, their popularity and demand are marred by the economic feasibility of production. They are currently unavailable for use at competitive prices (Rai et al., 2021).

Currently, low biobased bioplastics production can be attributed primarily to the competition with cheap virgin plastics made from low-cost fossil feedstock that go untaxed despite their carbon content (Hatti-Kaul et al., 2020).

A large amount of green food packaging systems is quite expensive compared to fossil-based systems. The price for commodity plastics is primarily centered in the range of $1.32–$3.3/kg. Unfortunately, no exact evaluation of price for traditional and biobased systems is accessible. In general, biobased, and biodegradable plastics are more expensive than fossil-based plastics on a weight basis. It was estimated that biobased materials are three to five times more expensive in comparison to traditional packaging systems (Luzi et al., 2019).

However, specific material properties can allow costs reductions in the use or end-of-life phase. There are several examples of bioplastic products being cost competitive already today. Further, the price of fossil-based plastics is depending on oil prices and fluctuates with oil prices while in general the prices of biobased plastics depend on more stable biomass prices (Van den Oever et al., 2017)

The higher drivers of the cost to realize green plastics include the cost for mobilizing biomass feedstocks, cost for technical and scientific innovations, and the lack of economies-of-scale (Luzi et al., 2019). Another barrier to reducing costs is the increase in production of biofuels which in many cases are competing for the same raw materials (corn and maize) as biobased packaging, putting upwards pressure on raw material costs (Robertson, 2012).

Cost is undoubtedly a limitation to the widespread adoption of biobased packaging materials, however, with the gaining momentum of industrial-scale production of bioplastics, the production cost is expected to reduce considerably (Robertson, 2012; Van den Oever et al., 2017; Rai et al., 2021). As the scaling-up of monomers production for biopolymers analogous to conventional polymers is successful, the production of biobased polymers will also be scaled up. This will result in prices that are competitive with those of petroleum-derived polymers (Nakajima et al., 2017).

SWOT analysis

As a conclusion to the market analysis of bioplastics for food packaging solutions, a SWOT analysis for each bioplastic category is presented.

Table 19. *SWOT analysis of Bioplastics for Food Packaging*

Strengths

- Nontoxicity and non health hazards
- biocompatible with other biobased bioplastics
- Biodegradable in home and industrial composting facilities and even some in soil and water
- Renewable resources origin
- GHG emission reduction
- Higher savings in fossil resources through its lifecycle
- Same processability of conventional plastics with smaller adjustments depending on the biopolymer formula
- Provide a circular bioeconomy
- More than 30 years of strong research and development

Weaknesses

- Low barrier, thermal and mechanical properties.
- Exist specific standards and labels but are not diffuse and well commonly understood
- Specific regulations for biodegradable disposal treatments
- Lack of economies of scale
- Higher mean price compared to fossil-based plastics
- Difficult processability due to natural composition properties

Opportunities

- Biopolymeric blends, hybrid blends with other biobased and/or fossil-based biodegradable bioplastics.
- Nanotechnology advances in biopolymers
- Used of non-biomass as feedstock:
  - Non-food crops
  - Agricultural and forestry waste
  - Industrial agriculture waste
  - By-products
  - Food-waste

Threats

- Increment of raw materials due to possible competition of agriculture crops between food and bioplastic production.
- Quality reduction of recyclable plastics if biodegradable plastics are not properly separated
- Externalities associated to agricultural-based bioplastics:
  - Increase of GHG emissions due to the use of fossil fuel, fertilizers, and pesticides
  - Risk of acidification of soil, ecotoxicity, eutrophication, ozone depletion
  - Higher water footprint
  - Concern about use of arable land competition with food crops and their market prices

Strengths

- Nontoxicity and non health hazards
- Can act as a drop-in material because its properties and performance are very similar and can sometimes even better than its conterparts fossil-based plastics
- Can be properly recyclable by type of bioplastic as its counterparts
- High scale production feasibility
- Easy to produce and management
- Provide a circular bioeconomy
- More than 30 years of strong research and development

Weaknesses

- Non-biodegradable
- Current recyclable systems are not efficient

Opportunities

- Biopolymeric blends
- Nanotechnology advances in biopolymers
- Used of non-biomass as feedstock:
  - Non-food crops
  - Agricultural and forestry waste
  - Industrial agriculture waste
  - By-products
  - Food-waste
- Improvements in research of chemical and mechanical recyclability systems

Threats

- Increment of raw materials due to possible competition of agriculture crops between food and bioplastic production.
- Quality reduction of recyclable plastics if biodegradable plastics are not proper separated
- Externalities associated to agricultural-based bioplastics:
  - Increase of GHG emissions due to the use of fossil fuel, fertilizers, and pesticides
  - Risk of acidification of soil, ecotoxicity, eutrophication, ozone depletion
  - Higher water footprint
  - Concern about use of arable land competition with food crops and their market prices

Strengths

• Nontoxicity and non health hazards
• Excellent barrier, thermal, and mechanical properties for food packaging
• Biodegradable in industrial composting facilities
• Cheaper than biobased biodegradable bioplastics
• High scale production feasibility
• Easy to produce and management
• Provide a circular bioeconomy

Weaknesses

• Currently industrial composting facilities are not widely spread out, and need more research and industrial investment to shift to a high scale composting production

Opportunities

• Biopolymeric blends
• Nanotechnology advances in biopolymers
• Are used to reinforce biobased biodegradable properties and make them more economic

Threats

• Most of them are not even recycled due to lack of proper labels and clear segragation systems for customers
• No savings in fossil resources
• Higher CO_2 and GHG emissions than biobased bioplastics

Strengths

• Nontoxicity and non health hazards
• Suitable barrier, thermal and mechanical properties for food packaging
• Cheaper than other alternatives
• Whole system of plasctic packaging is currently adapted for traditional plastics
• High scale production feasibility
• Easy to produce and management
• Can provide a circular economy only through a lean recyclability system
• More than 60 years of strong research and development

Weaknesses

• Durable, non-biodegradable
• Current collection and separation systems are not efficient

Opportunities

• Improvements in research of chemical and mechanical recyclability systems

Threats

• No savings in fossil resources
• Higher CO_2 and GHG emissions than biobased bioplastics
• Quality reduction of recyclable plastics if biodegradable plastics are not proper separated

Source. (Robertson, 2012; Álvarez-Chávez et al., 2012; Andrady, 2015; Van den Oever et al., 2017; Nakajima et al., 2017; Habel, et al., 2018; Spierling et al., 2018; Luzi et al., 2019; Kawashima et al., 2019; Hatti-Kaul et al., 2020; Acquavia et al., 2021; Rai et al., 2021).

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Conclusions

This study was done based on the latest updates corresponding to the development of sustainable plastic solutions used for the packing of food. And from the information presented, it is possible to determine how these are a source of novel products that can contribute to the problems associated with contamination generated by disposable or single-use plastics.

These sustainable solutions have the safeguarding characteristic of balance between economic, social, and environmental forces throughout their development, from raw material to the final disposal of the product after its use.

Nowadays, the sustainable solutions that can be considered to cooperate with a circular economy in the food packaging sector in plastic materials are the following:

1. Plastic materials grouped by the consensual name of "bioplastic" which in turn consists of three kinds of plastics:

    1.1 Bioplastics originated from renewable sources and which can biodegrade
    1.2 Bioplastics originated from renewable sources but cannot biodegrade
    1.3 Bioplastics originated from non-renewable sources but can biodegrade.

2. Plastic materials from petroleum resources that do not own the biodegrading property but can be adequately recycled based on the type of plastic that constitutes them.

Within these two large groups, bioplastics from renewable sources represent the most sustainable solutions for packaging from an environmental and safe perspective for human health. This classification is based on the CO_2 footprint reduction issued compared to conventional plastics. Moreover, coming from renewable resources, some can also biodegrade, thus constituting a potential reduction of plastic pollution.

Bioplastics from renewable resources represent a strong potential to contribute to a sustainable and circular economy in food packaging, although they have defiant challenges in moving to an economy of scale. Among the main barriers in this market is the difficulty of processing and manufacturing due to raw material's natural characteristics. Moreover, because of this, bioplastic's cost of development and innovation make the final total cost of bioplastic generally between 3 to 5 times higher than commodity plastic's prices.

On the other hand, further research on bioplastics is needed in terms of barrier and mechanical properties. Due to the natural properties of their renewable feedstock, they must be blended with other conventional plastics, which diminishes their biodegradability and sustainability. However, the latest advances in nanotechnology have greatly contributed to combating these disadvantages by forming sustainable and safe biobased copolymers for food packaging which enhance the moisture and gas barrier properties of bioplastics.

Thus, many companies in the biorefinery and green materials sector have now established encouraging formulas that can be found on the market at competitive prices despite having higher added value.

One of the great fears; regarding bioplastics; is that water and land are used for the harvest of their raw materials, because most companies developing these green plastics use agricultural products such as maize and sugar cane. Although only around 5% of arable soil is intended to be used, if 100% of plastics come from crops, the use of the necessary fresh water could pose a risk to many countries.

However, in recent years, other ways of obtaining bioplastic materials that do not have the minimum risk of competing with agricultural products for human and animal consumption, as well as water use, have been studied. Scientific advances indicate that it would be possible to obtain plastic materials even from organic waste discarded daily from houses and from agricultural waste obtained during the weaning process. Wood and other lignocellulosic residues from agriculture and forestry are also expected to be more sustainable alternatives.

The use of this biomass source would radically change the sustainability of the economics of bioplastics not only for applications in the food industry such as packaging but in other packaging sectors. This would change the context of global plastic pollution due to the reduction of plastic waste in natural environments, the diminution of GHG effect, and the shrinkage of marine and terrestrial animal species deaths.

Using these alternative biomass sources would help to avoid indirectly associated problems such as malnutrition, famine, and poverty in the countries from which these agricultural products are obtained. Most of which are located throughout Asia and Latin America.

Additionally, the use of non-edible sources would significantly reduce the cost of production. That way, the final price of bioplastic could compete and gain greater market share especially in the plastics sector for food packaging. This should therefore be the route to be followed for the development of more sustainable bioplastics that truly contribute to a circular bioeconomy.

On the other hand, although plastics derived from petrochemical sources can continue a circular course of life, their recycling process is difficult in the current recycling systems operating in most countries. This difficulty is due to their diversity and characteristics as a final product, meaning their design and constituents. Europe is the most advanced region in terms of plastic recycling rates; approximately 30 to 40% of plastics annually discarded are recycled. However, these figures, despite being the highest compared to other countries, still reflect an inefficient industrial system for plastic recycling.

More investment in D&R is needed and in technology that contributes to greater recycling efficiencies, which is entirely possible. Investment by governments in current recycling systems, whether mechanical and/or chemical is of crucial importance. Furthermore, the waste management and packaging industries should be promoted to effectively address the issue of production, use and management of bioplastics from a life cycle analysis perspective.

Also, more support is needed in regards to the new regulations that allow the correct classification of the consumer at the time of discarding the used wrapper to the subsequent logistics, segregation and industrial process system that guarantees high recycling processing efficiencies.

Another important action from corresponding public identities, as well as businesses and stakeholders associated with the food packaging sector is to align a standard designation for labeling and proper separation of biodegradable, compostable and/or recyclable packaging.

Because of this current void and confusion, most consumers have limited knowledge of the differences between bioplastics, biodegradable plastics, or biobased plastics. The consumer's attitudes and actions in choosing the product's final disposition is absolutely crucial to carrying out subsequent processes. If consumers are unable to select the appropriate final disposal of packaging, they lack environmental awareness or proper information, then an efficient and latest generation of industrial processes related to the after treatment of these wastes will have no greater impact on the material recirculation rates.

Recirculating these often used and discarded packages not only benefit the environment and the good health of communities. It notably contributes to the country's economy because the value of these rapidly discarded but serviceable plastics is reconstituted and revalued after the corresponding treatment. Being them mechanical or chemical recycling, home, or industrial composting. Reused plastic means added economic value for a nation and must be valued to contribute to the nation's sustainability.

Changing the way plastics are produced is not an easy task. Despite the progress made at the moment, there are many factors that determine the feasibility of this shift. The current supply and production chain of plastics is designed and established for plastics from fossil sources. A gradual change can occur. However, the collaboration of the industrial sector, the government and its policies are needed to facilitate this possibility on a large scale. In addition, responsible consumer education is one of the key pieces. Bioplastics are a sustainable alternative if the axes of change are aligned with their development and expansion.

Bioplastics are not a universal solution to all the world's plastic problem, but its contribution is vital to shifting towards a sustainable food packaging system. Considering food packaging is the main sector in the plastic packaging industry, its impact could drastically diminish plastic pollution.

Because of the nature of the food packaging market, these are on the front line to initiate a conversion to biobased and/or biodegradable plastics. Food packaging has an accelerated frequency of use, the properties of bioplastics can be designed based on the estimated food product shelf life and particular barrier properties requirements. Besides, contributing to sustainable final disposal alternatives such as composting, biodegradation and recycling.

As a conclusive delivery, the table 20 presented below summarizes the sustainable alternatives for conventional plastics used for food packaging applications. This table 20 could have only been done after the research work carried out in this thesis.

Table 20. *State-of-the-Art Sustainable Alternatives for Food Packaging made of Plastic Material*

Sustainable Alternative Bioplastic	Replaced Conventional Plastics	Differential Properties	Companies material suppliers	Production capacity tons/year
PLA	PE HDPE PP PS PET	• Rigid, brittle, and low ability to deformation, therefore, need a plasticizer • Excellent aroma barrier • Appearance: transparent to white and high gloss • Processability: thermoforming, coextrusion, injection molding • Printable • Can be produced cheaply • Biodegradable under certain conditions	NatureWorks Total Corbion Futerro BASF Biobent Carbiolice Danimer Scientific GC Innovation America Global Biopolymers Green Science Alliance Naturetec Oimo Omya Sulzer Tecnaro	>217 000
Starch blends	PE LDPE HDPE PP PS PVC	• Biocompatible • Appealing balance of properties • Suitable for flexible packaging • Higher commercial availability • Processability: extrusion, injection molding, blow molding, sheet extrusion, thermoforming • Competitive price within renewable source material market • Biodegradable under certain conditions	Novamont Sphere BASF Biome Bioplastics Cargill Agrana Biotec Cardia Bioplastics Cereplast Compostpack Gema Polimer Grabio Green Dot Bioplastic Green Science Alliance Plantic Technologies Rodenburg Biopolymers Tecnaro	>384 000
PHAs (PHB, PHBV)	PE LDPE HDPE PET PP PS PVC	• Biocompatible • Appealing balance of properties • Suitable for flexible packaging • Higher commercial availability • Processability: extrusion, injection molding, blow molding, sheet extrusion, thermoforming • Competitive price within renewable source material market • Biodegradable under certain conditions	Metabolix ADM Biobent Biomer Bosk Bioproducts Bio-on Danimer Scientific Kaneka Omya Tecnaro Terraverdae Venvirotech	>30 000
Cellulose based	PP PVC PS PET	• Is the most abundant biopolymer in nature • Present high mechanical strength and durability, and low density • Can mechanically reinforce and enhance the barrier properties of other biopolymers • Low price of feedstock • High extraction technology • Biodegradable in certain conditions	Bosk Bioproducts CelluForce Eastman Futamura Grabio Green Science Alliance Seelution Stora Enso Woodly	n.s

Sustainable Alternative Bioplastic	Replaced Conventional Plastics	Differential Properties	Companies material suppliers	Production capacity tons
bio-PE (bio-LDPE bio-LLDPE bio-HDPE)	PE (LDPE LLDPE HDPE)	• Characteristics and properties equivalent to fossil-based PE and its derivates. • Easy to recycle in PE recycling system • Applied in the same applications of PE • Same processability as conventional plastics • Is not biodegradable	Bosk Bioproducts CelluForce Eastman Futamura Grabio Green Science Alliance Seelution Stora Enso Woodly	>200 000
bio-PET	PET	• Performs similar to fossil-based PET • Easy to recycle in PET recycling system • Applied in the same applications of PET • Same processability as conventional plastics • Is not biodegradable	Anellotech FKuR Kunststoff GmbH GC Innovation America Tecnaro	>560 000
PEF	PET	• Performs better than fossil-based PET • Has better barrier, thermal and mechanical properties than PET • Easy to recycle in PET recycling system • Can be applied in the same applications of PET • Same processability as conventional plastics • Is not biodegradable	Avantium Archer Daniels Midland (ADM) Ava Biochem BASF Corbion Treemera	n.s

Note. Green color represent biodegradable and pink represents non-biodegradable bioplastics
Source. (Robertson, 2012; Álvarez-Chávez et al., 2012; Andrady, 2015; Van den Oever et al., 2017; Nakajima et al., 2017; Habel, et al., 2018; Spierling et al., 2018; Luzi et al., 2019; Kawashima et al., 2019; Hatti-Kaul et al., 2020; Acquavia et al., 2021; Rai et al., 2021).

In order to shift plastic food packaging to more sustainable alternatives at a larger scale, it is necessary to make more scientific research and development. The research required must focus mainly on mechanisms to enhance barrier properties of biobased bioplastics, the use of non-food feedstocks for bioplastic production, and the impact of post-consumer final treatments of these bioplastics (recyclability and compostability). Additionally, the development of standard regulations, system and clear labels for traditional plastics and bioplastics are crucial to support consumers' correct segregation. It is also compulsory to have more investment and development in the industrial efficiency of the current recyclable systems for plastic.

Glossary

Chapter I

Absorption: The process of absorption means that a substance captures and transforms energy. The absorbent distributes the material it captures throughout whole and adsorbent only distributes it through the surface. The process of gas or liquid which penetrate into the body of adsorbent is commonly known as absorption (Wikipedia, n.d.).

Adsorption: Adsorption is the adhesion of atoms, ions or molecules from a gas, liquid or dissolved solid to a surface. This process creates a film of the adsorbate on the surface of the adsorbent. This process differs from absorption, in which a fluid is dissolved by or permeates a liquid or solid, respectively (Wikipedia, n.d.).

Aerobic respiration: Cellular respiration is a set of metabolic reactions and processes that take place in the cells of organisms to convert chemical energy from oxygen molecules[1] or nutrients into adenosine triphosphate (ATP), and then release waste products. Aerobic respiration requires oxygen (O2) in order to create ATP. (Wikipedia, n.d.).

Bacteriocins: Bacteriocins are proteinaceous or peptidic toxins produced by bacteria to inhibit the growth of similar or closely related bacterial strain (Wikipedia, n.d.).

Catalysts: Catalysis is the process of increasing the rate of a chemical reaction by adding a substance known as a catalyst. Catalysts are not consumed in the catalyzed reaction but can act repeatedly. Often only very small amounts of catalyst are required (Wikipedia, n.d.).

Chlorophyll: Any member of the most important class of pigments involved in photosynthesis, the process by which light energy is converted to chemical energy through the synthesis of organic compounds. Chlorophyll is found in virtually all photosynthetic organisms, including green plants, cyanobacteria, and algae (Britannica, n.d.).

DP: degree of polymerization

Enzyme: A substance that acts as a catalyst in living organisms, regulating the rate at which chemical reactions proceed without itself being altered in the process (Britannica, n.d.).

Ethylene: Ethylene ($H_2C=CH_2$), the simplest of the organic compounds known as alkenes, which contain carbon-carbon double bonds. It is a colourless, flammable gas having a sweet taste and odour. Natural sources of ethylene include both natural gas and petroleum; it is also a naturally occurring hormone in plants, in which it inhibits growth and promotes leaf fall, and in fruits, in which it promotes ripening. Ethylene is an important industrial organic chemical (Britannica, n.d.).

HDPE: High Density Polyethylene

LDPE: Low Density Polyethylene

Metabolites: In biochemistry, a metabolite is an intermediate or end product of metabolism. The term metabolite is usually used for small molecules (Wikipedia, n.d.).

MW: Molecular Weight

Nucleic acids: Is a naturally occurring chemical compound that is capable of being broken down to yield phosphoric acid, sugars, and a mixture of organic bases (purines and pyrimidines) (Britannica, n.d.).

OML: Overall migration limit

Organoleptic properties: Are the aspects of food, water or other substances that create an individual experience via the senses—including taste, sight, smell, and touch (Wikipedia, n.d.).

PET: Polyethylene Terephthalate

PH: Quantitative measure of the acidity or basicity of aqueous or other liquid solutions (Britannica, n.d.).

Polyolefins: Are a family of polyethylene and polypropylene thermoplastics. They are produced mainly from oil and natural gas by a process of polymerisation of ethylene and propylene respectively (Plastics Europe, n.d.).

PP: Polypropylene

PP: Polypropylene

PS: Polystyrene

PVC: Polyvinyl chloride

Refractive index: also called index of refraction, measure of the bending of a ray of light when

passing from one medium into another (Britannica, n.d.).
Resins: Any natural or synthetic organic compound consisting of a noncrystalline or viscous liquid substance. Natural resins are typically fusible and flammable organic substances that are transparent or translucent and are yellowish to brown in colour. They are formed in plant secretions and are soluble in various organic liquids but not in water. Synthetic resins comprise a large class of synthetic products that have some of the physical properties of natural resins but are different chemically. Synthetic resins are not clearly differentiated from plastics (Britannica, n.d.).

RFID: Radio frequency identification

SML: Specific migration limit

TTIs: Time-temperature indicators

Chapter II

Aliphatic polyester: Aliphatic polyesters include poly(hydroxy acid)s, such as poly(lactic acid) (PLA) and poly(glycolic acid) (PGA); polyhydroxyalkanoates (PHAs), derived mainly from microorganisms; and poly(alkylene dicarboxylate)s, derived from both fossil fuel and renewable resources, such as poly(butylene succinate) (PBS) and poly(butylene adipate-co-terephthalate) (PBAT) (Niaounakis, 2015).

Anaerobic digestion: Anaerobic digestion is a sequence of processes by which microorganisms break down biodegradable material in the absence of oxygen (Wikipedia, n.d.).

Bacteria: Bacteria lack a membrane-bound nucleus and other internal structures and are therefore ranked among the unicellular life-forms called prokaryotes. Prokaryotes are the dominant living creatures on Earth, having been present for perhaps three-quarters of Earth history and having adapted to almost all available ecological habitats. As a group, they display exceedingly diverse metabolic capabilities and can use almost any organic compound, and some inorganic compounds, as a food source. Some bacteria can cause diseases in humans, animals, or plants, but most are harmless and are beneficial ecological agents whose metabolic activities sustain higher life-forms (Britannica, n.d.).

Biocompatibility: Biocompatibility is a general term describing the property of a material being compatible with living tissue. Biocompatible materials do not produce a toxic or immunological response when exposed to the body or bodily fluids (Spine-health, n.d.).

Compost: Organic amendments or soil improvers which are obtained through a biodegradation of a mixture of vegetable residues and of other organic materials with a limited amount of minerals (Plastics Europe, 2017).

Composting: Aerobic process for the production of compost (Plastics Europe, 2017).
Copolymer: Copolymer, any of a diverse class of substances of high molecular weight prepared by

chemical combination, usually into long chains, of molecules of two or more simple compounds (the monomers forming the polymer) (Britannica, n.d.).

Dicarboxylic acids: is an organic compound containing two carboxyl functional groups (−COOH). A carboxylic acid is an organic acid that contains a carboxyl group (C(=O)OH) (Wikipedia, n.d.).

Disintegration: is the physical falling apart of the biodegradable plastic material, or more precisely the product that has been made from it, into fine visually indistinguishable fragments at the end of a typical composting cycle (Rujnić-Sokele & Pilipović, 2017).

Ester linkages: An ester is the compound obtained when the hydrogen atom in at least one hydroxy group in an oxoacid or a hydroxoacid is replaced by an alkyl group (alkyl ester) or an aryl group (aryl ester). In an ester molecule, the bond connecting the atom doubly bonded to oxygen and the oxygen atom bearing the alkyl or aryl group is called the ester bond or, in biochemistry, ester linkage (Chemistry, n.d.).

Glucose: Glucose, also called dextrose, one of a group of carbohydrates known as simple sugars (monosaccharides). Glucose (from Greek glykys; "sweet") has the molecular formula $C_6H_{12}O_6$. It is found in fruits and honey and is the major free sugar circulating in the blood of higher animals. It is the source of energy in cell function, and the regulation of its metabolism is of great importance (see fermentation; gluconeogenesis). Molecules of starch, the major energy-reserve carbohydrate of plants, consist of thousands of linear glucose units. Another major compound composed of glucose is cellulose, which is also linear (Britannica, n.d.).

Hydrogenation: Is a chemical reaction between molecular hydrogen (H_2) and another compound or element, usually in the presence of a catalyst such as nickel, palladium or platinum. The process is commonly employed to reduce or saturate organic compounds (Wikipedia, n.d.).

Hygroscopy: Is the phenomenon of attracting and holding water molecules via either absorption or adsorption from the surrounding environment, which is usually at normal or room temperature. If water molecules become suspended among the substance's molecules, adsorbing substances can become physically changed, e.g., changing in volume, boiling point, viscosity or some other physical characteristic or property of the substance (Wikipedia, n.d.).

Maleic anhydride: Is an organic compound with the formula $C_2H_2(CO)_2O$. It is the acid anhydride of maleic acid. It is a colorless or white solid with an acrid odor. It is produced industrially on a large scale for applications in coatings and polymers (Wikipedia, n.d.).

Mineralization: The breakdown of a chemical substance or organic matter by microorganisms in the presence of oxygen to carbon dioxide, water and mineral salts of any other elements present (Plastics Europe, 2017).

PBAT: Poly (butylene adipate-co-terephthalate)

PBS: Polybutylene succinate

PE: Polyethylene
PGA: Poly(glycol acid)

PHA: Polyhydroxyalkanoates

PHB: Poly(hydroxybutyrate)

PHBV: (hydroxybutyrate-co-hydroxyvalerate)

Photo-oxidation: Is the degradation of a polymer surface in the presence of oxygen or ozone, facilitated by radiant energy such as UV or artificial light (Wikipedia, n.d.).

Plasticizers: Is a substance that is added to a material to make it softer and more flexible, to increase its plasticity, to decrease its viscosity, or to decrease friction during its handling in manufacture (Wikipedia, n.d.).

Protozoa: Organism, usually single-celled and heterotrophic (using organic carbon as a source of energy), belonging to any of the major lineages of protists and, like most protists, typically microscopic. All protozoans are eukaryotes and therefore possess a "true," or membrane-bound, nucleus. They also are nonfilamentous (in contrast to organisms such as molds, a group of fungi, which have filaments called hyphae) and are confined to moist or aquatic habitats, being ubiquitous in such environments worldwide, from the South Pole to the North Pole. Many are symbionts of other organisms, and some species are parasites (Britannica, n.d.).

PVOH: Poly(vinyl alcohol) (PVOH, PVA, or PVAl) is a water-soluble synthetic polymer. It has the idealized formula [CH2CH(OH)]. PVA is used in a variety of medical applications because of its biocompatibility, low tendency for protein adhesion, and low toxicity (Wikipedia, n.d.).

Radiocarbon method: Carbon-14 dating, also called radiocarbon dating, method of age determination that depends upon the decay to nitrogen of radiocarbon (carbon-14). Carbon-14 is continually formed in nature by the interaction of neutrons with nitrogen-14 in the Earth's atmosphere; the neutrons required for this reaction are produced by cosmic rays interacting with the atmosphere (Britannica, n.d.).

Succinic anhydride: Succinic anhydride, is an organic compound with the molecular formula (CH2CO)2O. In the laboratory, this material can be prepared by dehydration of succinic acid (Wikipedia, n.d.).

Sucrose: Or table sugar, organic compound, colourless sweet-tasting crystals that dissolve in water. Sucrose (C12H22O11) is a disaccharide; hydrolysis, by the enzyme invertase, yields "invert sugar" (so called because the hydrolysis results in an inversion of the rotation of plane polarized light), a 50:50 mixture of fructose and glucose, its two constituent monosaccharides. Sucrose occurs naturally in sugarcane, sugar beets, sugar maple sap, dates, and honey. It is produced commercially in large amounts (especially from sugarcane and sugar beets) and is used almost entirely as food (Britannica, n.d.).

TPS: Thermoplastic starch

Chapter III

5-hydroxymethylfurfural: Hydroxymethylfurfural, also 5-furfural, is an organic compound formed by the dehydration of certain sugars. It is a white low-melting solid which is highly soluble in both water and organic solvents. The molecule consists of a furan ring, containing both aldehyde and alcohol functional groups (Wikipedia, n.d.).

Additives: The most common polymer additives are stabilizers, plasticizers, lubricants and flame retardants (Hunt, 2000).

Bionaphtha: Is a mixture of C5 - C10 hydrocarbon compounds which are volatile and flammable with boiling points in the range of 30-200°C resulting from bio-based feedstock processing such as biomass and palm oil (Widikrama & Rachmawati, 2019).

Cross contamination: inadvertent transfer of bacteria or other contaminants from one surface, substance, etc., to another especially because of unsanitary handling procedures (Merriam-Webster, n.d.).

Depolymerization: Is the process of converting a polymer into a monomer or a mixture of monomers (Wikipedia, n.d.).

Dye: substance used to impart colour to textiles, paper, leather, and other materials such that the colouring is not readily altered by washing, heat, light, or other factors to which the material is likely to be exposed. Dyes differ from pigments, which are finely ground solids dispersed in a liquid, such as paint or ink, or blended with other materials. Most dyes are organic compounds (i.e., they contain carbon), whereas pigments may be inorganic compounds (i.e., they do not contain carbon) or organic compounds (Britannica, n.d.).

Covalent bond: in chemistry, the interatomic linkage that results from the sharing of an electron pair between two atoms (Britannica, n.d.).

EG: ethylene glycol

Electrostatics: Is the study of electromagnetic phenomena that occur when there are no moving charges—i.e., after a static equilibrium has been established (Britannica, n.d.).

Epoxy groups: Any of a class of thermosetting polymers, polyethers built up from monomers with an ether group that takes the form of a three-membered epoxide ring (Britannica, n.d.).

EPS: Expanded polystyrene

FCM: food contact materials

GHG: greenhouse gases

Carboxyl group: in organic chemistry, a divalent chemical unit consisting of a carbon (C) and an oxygen (O) atom connected by a double bond (Britannica, n.d.).

Lignocellulosic: Refers to plant dry matter, so called lignocellulosic biomass. It is the most abundantly available raw material on the Earth for the production of biofuels, mainly bioethanol (Wikipedia, n.d.).

Microplastics: Very small (<5mm) non-biodegradable plastic particles formed through mechanical degradation of larger pieces of plastics. Biodegradable plastic should not yield microplastics as these will be assimilated by microorganisms (Annemette et al., ca. 2019).

MSW: Municipal solid waste

Polycarbonates: Are a group of thermoplastic polymers containing carbonate groups in their chemical structures. Polycarbonates used in engineering are strong, tough materials, and some grades are optically transparent. They are easily worked, molded, and thermoformed (Wikipedia, n.d.).

Polyesters: Polyesters are polymers made by a condensation reaction taking place between monomers in which the linkage between the molecules occurs through the formation of ester groups. The major industrial polyesters include polyethylene terephthalate, polycarbonate, degradable polyesters, alkyds, and unsaturated polyesters (Britannica, n.d.).

Polymerization: Any process in which relatively small molecules, called monomers, combine chemically to produce a very large chainlike or network molecule, called a polymer (Britannica, n.d.).

Polyurethanes: Polyurethane (PUR and PU) is a polymer composed of organic units joined by carbamate (urethane) links (Wikipedia, n.d.).

PSW: plastic solid waste

RPM: Recycled Plastic Material

RPET: Recycled PET

Spectral signatures: Is the variation of reflectance or emittance of a material with respect to wavelengths (i.e., reflectance/emittance as a function of wavelength) (Wikipedia, n.d.).

Stabilizers: Stabilizers are added to prolong the useful life of a polymer formulation by protecting it from thermal and light-assisted oxidation (Hunt, 2000).

Vitrimer: are a class of plastics, which are derived from thermosetting polymers (thermosets) and are very similar to them. Vitrimers consist of molecular, covalent networks, which can change their topology by thermally activated bond-exchange reactions (Wikipedia, n.d.).

β-hydrogen bonds: Hydrogen bonding, interaction involving a hydrogen atom located between a pair of other atoms having a high affinity for electrons; such a bond is weaker than an ionic bond or covalent bond but stronger than van der Waals forces (Britannica, n.d.).

Chapter IV

Acetylation reaction: Acetylation is an organic esterification reaction with acetic acid. It introduces an acetyl functional group into a chemical compound. Such compounds are termed acetate esters or acetates (Wikipedia, n.d.).

Ach: Alkyl chitosan

AgNPs: Silver nanoparticles

Amino acid: Amino acid, any of a group of organic molecules that consist of a basic amino group, an acidic carboxyl group, and an organic R group (or side chain) that is unique to each amino acid. The term amino acid is short for Î±-amino [alpha-amino] carboxylic acid (Britannica, n.d.).

Amphiphilic: An amphiphile is a chemical compound possessing both hydrophilic and lipophilic properties (Wikipedia, n.d.).

Biocide: A biocide is defined in the European legislation as a chemical substance or microorganism intended to destroy, deter, render harmless, or exert a controlling effect on any harmful organism (Wikipedia, n.d.).

By-product: A by-product or byproduct is a secondary product derived from a production process, manufacturing process or chemical reaction; it is not the primary product or service being produced (Wikipedia, n.d.).

CA: Cellulose acetate

Calcium chloride (CaCl2): Is a colourless or white solid produced in large quantities either as a by-product of the manufacture of sodium carbonate by the Solvay process or by the action of hydrochloric acid on calcium carbonate (Britannica, n.d.).

CCh: Carboxymethyl chitosan

Cellobiose: Is a disaccharide classified as a reducing sugar (Wikipedia, n.d.).

chemical oxidation: Chemical oxidation is a process involving the transfer of electrons from an oxidizing reagent to the chemical species being oxidized (Shammas et al., 2005)

Compression molding: Compression Moulding is a method of moulding in which the moulding material, generally preheated, is first placed in an open, heated mould cavity (Wikipedia, n.d.).

CM: Methyl cellulose

CMC: Carboxymethyl cellulose

Cross-link: In chemistry and biology a cross-link is a bond that links one polymer chain to another. These links may take the form of covalent bonds or ionic bonds and the polymers can be either synthetic polymers or natural polymers (Wikipedia, n.d.).

Curtain coating: Curtain coating is a process that creates an uninterrupted curtain of fluid that falls onto a substrate. The substrate is transported on a conveyor belt or calender rolls at a regulated speed through the curtain to ensure an even coat of the die (Wikipedia, n.d.).

Denaturation: Is a process in which proteins or nucleic acids lose the quaternary structure, tertiary structure, and secondary structure which is present in their native state, by application of some external stress or compound such as a strong acid or base, a concentrated inorganic salt, an organic solvent (e.g., alcohol or chloroform), radiation or heat (Wikipedia, n.d.).

Dip coating: Is an industrial coating process where the substrate is immersed in the solution of the coating material at a constant speed (preferably jitter-free) (Wikipedia, n.d.).

EC: Ethyl cellulose

Endosperm: Is a tissue that surrounds and nourishes the embryo in the seeds of angiosperms (flowering plants) (Britannica, n.d.).

EVOH: Ethyl vinyl alcohol

Globular proteins: Or spheroproteins are spherical proteins and are one of the common protein types. Globular proteins are somewhat water-soluble, unlike the fibrous or membrane proteins (Wikipedia, n.d.).

Glycerol: a clear, colourless, viscous, sweet-tasting liquid belonging to the alcohol family of organic compounds; molecular formula HOCH2CHOHCH2OH (Britannica, n.d.).

Glycine: the simplest amino acid, obtainable by hydrolysis of proteins. Sweet tasting, it was among the earliest amino acids to be isolated from gelatin (1820) (Britannica, n.d.).

Glycosidic bond: A glycosidic bond or glycosidic linkage is a type of covalent bond that joins a carbohydrate molecule to another group, which may or may not be another carbohydrate. A glycosidic bond is formed between the hemiacetal or hemiketal group of a saccharide and the hydroxyl group of some compound such as an alcohol (Wikipedia, n.d.).

HEC: Hydroxyethyl cellulose

HNTs: Halloysite nanotubes

HPC: Hydroxypropyl cellulose

HPMC: Hydroxypropyl methyl cellulose

Hydrocolloid: Hydrocolloids are moisture-retentive dressings, which contain gel-forming agents such as sodium carboxymethylcellulose and gelatin. Many products combine the gel-forming properties with elastomers and adhesives are applied to a carrier such as foam or film to form an absorbent, self-adhesive, waterproof wafer (Weller, 2009).

Hydrophobe: In chemistry, hydrophobicity is the physical property of a molecule that is seemingly repelled from a mass of water. In contrast, hydrophiles are attracted to water. Hydrophobic molecules tend to be nonpolar and, thus, prefer other neutral molecules and nonpolar solvents (Wikipedia, n.d.).

LbL: Layer-by-Layer

MAH: Maleic anhydride

NaCas: Sodium caseinate

NC: Nanocellulose

NFC: Nanofibrillated cellulose

Phosphoproteins: A phosphoprotein is a protein that is posttranslationally modified by the attachment of either a single phosphate group, or a complex molecule such as 5′-phospho-DNA, through a phosphate group (Wikipedia, n.d.).

Polarity: In chemistry, polarity is a separation of electric charge leading to a molecule or its chemical groups having an electric dipole moment, with a negatively charged end and a positively charged end (Wikipedia, n.d.).

QCh: Quaternary chitosan

SPI: Soy protein isolate

Proline: Proline, an amino acid obtained by hydrolysis of proteins. Its molecule contains a secondary amino group (>NH) rather than the primary amino group (>NH2) characteristic of most amino acids (Britannica, n.d.).

Prolamin: any of certain seed proteins known as globulins that are insoluble in water but soluble in water-ethanol mixtures. Prolamins contain large amounts of the amino acids proline and glutamine (from which the name prolamin is derived) but only small amounts of arginine, lysine, and histidine (Britannica, n.d.).

PVA: vinyl alcohol

PVDC: poly (vinylidene chloride)

R&D: Research & Development

Sizing agents: Sizing agents are applied into the paper to impart certain desirable qualities. The main function of sizing agent is to increase the resistance to penetration of water or other liquids into the paper so that the paper is suitable for printing, writing and other purpose (Wikipedia, n.d.).

Sodium alginate (SA): Alginic acid, also called algin, is a polysaccharide distributed widely in the cell walls of brown algae that is hydrophilic and forms a viscous gum when hydrated (Wikipedia, n.d.).

Surface sizing: Surface sizing agents are amphiphilic molecules, having both hydrophilic (water-loving) and hydrophobic (water-repelling) ends (Wikipedia, n.d.).

Surface energy: Surface free energy or interfacial free energy or surface energy quantifies the disruption of intermolecular bonds that occurs when a surface is created (Wikipedia, n.d.).
Transglutaminases: Form extensively cross-linked, generally insoluble protein polymers (Wikipedia, n.d.).

Xylitol: Is a chemical compound with the formula $C_5H_{12}O_5$, or $HO(CHOH)_3OH$; specifically, one particular stereoisomer with that structural formula. It is a colorless or white crystalline solid that is soluble in water. It can be classified as a polyalcohol and a sugar alcohol, specifically an alditol (Wikipedia, n.d.).

NnO: Zinc oxide nanoparticles

Chapter V

Circular economy: A concept of an economy that is restorative or regenerative in contrast to the concept of end-of-life or waste. It is based on a shift to renewable energy, superior design of materials and products without any toxic chemicals to allow their reuse or recycling, and innovative services and business models (Hatti-Kaul et al., 2020).

Bioeconomy: Encompasses the production of renewable biological resources and the conversion of these resources and waste streams into value-added products, such as food, feed, biobased products, and bioenergy (Hatti-Kaul et al., 2020).

Circular Bioeconomy: Combines the common features of bioeconomy and circular economy, (i.e., improved resource and ecoefficiency, low GHG footprint, reducing the demand for fossil carbon, and valorization of waste and side streams) (Hatti-Kaul et al., 2020).

SWOT analysis: Is a strategic planning technique used to help a person or organization identify strengths, weaknesses, opportunities, and threats related to business competition or project planning (Wikipedia, n.d.).

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Author motivation

This research work has been carried out mainly by personal and academic motivation based on the design of sustainable materials that contribute to the economic development of nations, scientific advancement and above all the well-being of our natural ecosystems and the good health of the population.

Questo lavoro da ricerca è stato svolto principalmente diretto da motivazione personale e accademica basata sulla progettazione di materiali sostenibili che contribuiscono allo sviluppo economico delle nazioni, al progresso scientifico e soprattutto al benessere dei nostri ecosistemi naturali e alla buona salute della popolazione.

Este trabajo de investigación ha sido llevado a cabo principalmente direccionado por la motivación personal y académica basada en el diseño de materiales sustentables que contribuyan con el desarrollo económico de las naciones, el avance científico y sobre todo el bienestar de nuestros ecosistemas naturales y la buena salud de la población.