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PhD Program in Innovation for The Circular Economy

Development of innovative materials and processes for cellulose-based food packaging in a circular economy perspective

Candidate:

Gioele Giordanengo

Supervisors:

Prof. **Claudia Barolo**

Dr. **Matteo Bonomo**

Jury Members:

Prof. **Marilena Carbone**

University of Rome, Tor Vergata

Department of Chemical Science ad Technologies

Dr. **Emilia Paone**

Department of Civil, Energy, Environmental and Materials Engineering

"Mediterranea" University of Reggio Calabria

Prof. **Francesca Bonino**

University of Turin

Department of Chemistry

Head of the Doctoral School: Prof. Eleonora Bonifacio

PhD Program Coordinator: Prof. Francesco Quatraro

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Premises

In recent years, concerns about the limited natural resources of fossil fuels and the environmental impact caused by the use of non-biodegradable plastic packaging materials have pushed research into the development of biopolymers based on agro-industrial products and/or food waste. [1]

In particular, significant interest in these innovative materials concerns the food packaging sector, where consumer demand for safe, high-quality, and long-lasting food products aligns with a strong ecological and environmental sensitivity regarding the issue of waste derived from food packaging.[1,2]

The development of sustainable packaging can be made possible and improved using renewable biocompatible materials, which will be the primary source for the development of environmentally sustainable packaging.[3]

It is also important to emphasize how packaging serves a social function, as it not only allows consumers to acquire information about the sustainability of what they purchase but also lays the groundwork for an indispensable role: the sustainability of the product is an inherent prerogative of the packaging itself.

The following paper presents the research and development activities regarding innovative paper-based food packaging developments and solutions carried out during my PHD studies in collaboration with Ecopack SPA.

Ecopack is a leader company in the design and development of paper baking forms for the bakery sector in the food packaging industry.

The PhD studies focused on the circularity of the packaging specifically addressing environmental sustainability related to the research and development of new raw materials and more sustainable packaging products.

These were proposed as viable alternatives for Ecopack, aligning with their commitment to sustainability in the food packaging industry.

Through effective awareness-raising facilitated by the doctoral program, it was possible to convey and disseminate to Ecopack fundamental concepts and approaches for the sustainable development of new packaging products to be proposed as alternatives for the global packaging market.

Through the implementation of more efficient machinery design, molds, and processes by the company, it is now possible to produce and offer lighter products tailoring the consumers' requests. These products are composed of 100% bio-based raw materials, easily recyclable and compostable, and exhibit high mechanical performance during their application phase.
These aspects also bring benefits in terms of reduced consumption of resources such as energy and water, fewer chemicals used, and less environmental impact in terms of CO_2 emissions.
Some examples of packaging products improved from an environmental sustainability standpoint are specifically descried below.

The compostable product lines have been certified according to the EN13432 regulation and awarded "OK COMPOST INDUSTRIAL and HOME" certifications following the certification standard by TUV AUSTRIA. This endeavor, which involved extensive analysis, laboratory testing, and data processing, has facilitated a better and validated end-of-life disposal and recycling phase for the product.
Through improved supply chain planning and advanced engineering of the product lifecycle, raw material waste has been optimized, along with the valorization of production by-products.
Process by-products and paper trimmings have been recovered and utilized in other process cycles as secondary raw materials, enhancing their value through a new purpose.
Thanks to the actions taken in recent years, Ecopack has opened various possibilities in shaping a wide range of paper materials, giving rise to innovative and sustainable packaging. These actions have allowed for the refinement of processes and technologies, enabling the production of solutions increasingly tailored to the needs of both the market and the environment.
Through a consistent commitment to research and development, Ecopack has enhanced its capabilities in creating products that not only meet required quality standards but also address growing environmental concerns.
As a result, the company is now able to offer a wide range of customized solutions designed to minimize environmental impact and maximize production efficiency.

This targeted approach has not only improved Ecopack's reputation as a leader in the packaging industry but has also laid a solid foundation for long-lasting partnerships with customers, built on trust and shared values of sustainability.

Introduction

History and Innovations in Food Packaging

Food packaging has evolved mimicking the changes in human lifestyle. Until the 1800s, there was minimal sophistication in packaging materials, with natural objects like gourds, shells, and leaves used to contain food, and grass, wood, and bamboo woven into baskets. Some of the earliest materials that could be shaped into food containers included ceramics, paper, glass, plastic.[1]

The first approaches toward functional packaging occurred during the Industrial Revolution, which led to the adoption of production processes that evolved over the years with progress. Initially, much of the packaging solutions were intended for food products.

Cardboard was first used to produce folding cartons in the early 1800s, while in 1850, the first corrugated boxes were developed, which are widely used today to contain a number of smaller packages.

In the 1950s and 1960s, thanks to significant discoveries such as the case of isotactic polypropylene, credit to Professor Giulio Natta, it became possible to create packaging for snacks with improved vapor barrier properties, better transparency, and increased rigidity.[4]

In 1970, with the contribution of PepsiCo, the concept of bottle was introduced, employing polyethylene terephthalate (PET) [5]as the most suitable and compatible material for containing liquid beverages. PET offers the necessary barrier to contain and preserve the content, preventing packaging components from interacting with the content itself and avoiding unwanted contamination by the end consumer.

In parallel with the development of materials and their related performances, packaging known today as "active packaging" has been developed.

An example is the food bag designed for containing items like popcorn or others usable through microwave heating. It's a laminated paper bag with a metallized film that interacts when exposed to microwave energy, enabling the cooking of the food content inside.[6]

Another packaging that has introduced a new category of products to the market is Modified Atmosphere Packaging (MAP). This type of packaging allows for extended preservation of foods. However, whenever one decides to adopt this packaging concept, it's necessary to use appropriate materials to maintain the desired atmosphere and prevent gases from penetrating through the packaging.[1,2]

Nowadays, the focus of the food packaging industry and research in this field is to find bio-based materials that are derived from non-synthetic sources. These materials can be configured to have a low environmental impact and can be easily recycled or composted, aligning with the approach towardsthe circular economy model.

The innovation of packaging is also greatly driven by market demand, which is increasingly attentive and sensitive to the issue of environmental sustainability.

In recent years, packaging innovation has contributed to creating new categories of food products and new packaging materials used.[3]

Packaging and main characteristic

Today, the market demands a significant amount of plastic, with an estimated 5% being recycled and 33% ending up in fragile ecosystems such as the ocean or the environment. This figure is steadily increasing.[3]

The packaging sector has always been the major contributor to industrial plastic consumption, leading to significant challenges in terms of disposal and recycling of the materials used, resulting in an unsatisfactory waste recovery rate despite many advancements in these terms. Hence, the importance of utilizing bio-based plastics.[7]

The production of bio-based plastics reached a quantity of 1.7 million tons in 2014 and 7.9 million tons in 2019, and it is estimated that after 2030, they will replace 100% not only conventional synthetic plastics but also enter the market of new innovative materials, whose invention will also be supported by technological development.[8]

In terms of environmental suitability, biopolymers are eco-friendly packaging materials, and the complete scheme containing the families and types of biopolymers is provided above. The main advantage of biopolymers over fossil-based polymers lies in their renewable natural cycle, where the end of one cycle leads to the beginning of the next cycle.[9]

The three main categories of biopolymers listed in the scheme differ in terms of origin and method of production and acquisition of the biopolymer. Biopolymers are divided into three main categories depending on origin and production method: those directly extracted from biomass, synthesized bio-derived monomers, and those produced by microorganisms. Polysaccharides and proteins are the most promising biopolymers to produce packaging materials. Proteins are heteropolymers consisting of α-amino acids as monomeric units. Combinations of 20 amino acids to form a protein sequence allow for an almost unlimited number of different polymer chains with different physical and chemical properties. Proteins also contain many functional groups that can be enzymatically, chemically, or physically modified to vary the properties of the films.[9]

Polysaccharides are good candidates for replacing petroleum-based polymers due to their ability to form a film, affinity for paper-based materials, appropriate barrier to gases and aromas, and good mechanical strength. Moreover, these biopolymers are biodegradable, non-toxic, and are used as a matrix for the inclusion of additives with specific functionalities, such as active antimicrobial properties. Polymer recycling is carried out a limited number of times, after which the issue of burial or incineration of these materials arises again.[10]

The prefix "bio" indicates that biopolymers are biodegradable. The term "biodegradable" means that materials can be decomposed by bacteria, fungi, and yeast into the final products of biomass under anaerobic conditions, hydrocarbons, and methane. These types of polymers are made up of monomers that are covalently linked, forming a chain of the molecule. They are also produced inside the cell because of complex metabolic processes. Biopolymers can be used for food packaging as substitutes for petroleum-based plastics due to their biodegradability, renewability, and wide distribution. Environmental sustainability is a key ideology nowadays. The use of biopolymers from renewable sources could solve the global plastic pollution problem. For many years, researchers have been trying to develop and design packaging materials based on natural biopolymers. However, animal proteins and natural polysaccharides are characterized by some undesirable properties caused by their chemical nature and structure.[11]

These disadvantages reduce their competitiveness compared to petroleum-based plastics but can be overcome. The most popular biopolymers include starch, cellulose, pectin, chitosan, alginate, casein, collagen, and gelatin, considered to produce films and coatings in general, and for each biopolymer. The main characteristics of packaging materials based on animal proteins and natural polysaccharides, suitability for a specific type of product based on polymer properties, and ways to improve them.[12]

A representative diagram of biopolymers is provided below.

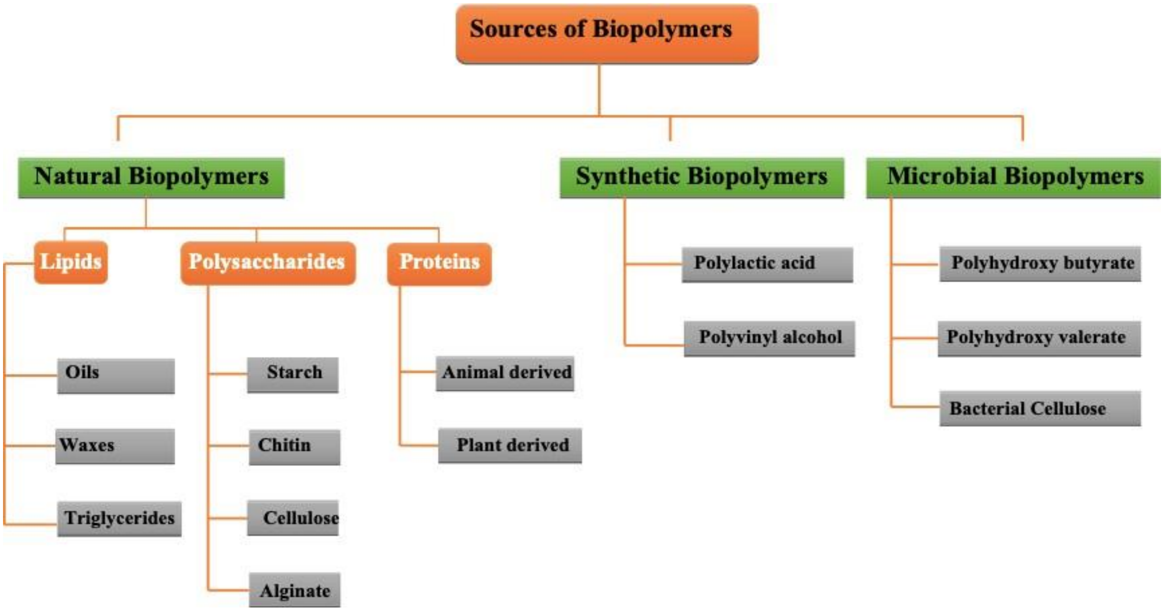


Figure 1 Graphic representation of the different categories of biopolymer[12]

Materials based on biopolymers must meet the basic requirements of health safety, mechanical and chemical resistance, and durability. Therefore, food packaging should not only be biodegradable but also functional.

Compared to synthetic polymers derived from petroleum, biopolymers have a more complex chemical structure and side-chain structure, which provide additional opportunities for the formation of packaging materials with specific characteristics for specific purposes.

Biopolymers directly extracted from biomass such as polysaccharides and animal proteins are the ones most often used for the preparation of food packaging materials, and a brief mention of this can be provided below.

Starch is one of the most readily available polysaccharides on the planet, and the plants from which it is extracted grow in virtually all temperate climate zones. The main sources of starch are corn, wheat, potatoes, and rice, which account for 84%, 7%, 4%, and 1% respectively. This biopolymer is a mixture of amylose and amylopectin, the ratio of which varies depending on the type of starch and finds numerous applications including the food packaging sector.[13]

Cellulose is the most common natural biopolymer and consists of β-(1–4)-D-glucopyranose monomers. It is biosynthesized by numerous living organisms, ranging from lower to higher plants, marine animals, bacteria, and fungus. It is estimated that 10^{11} to 10^{12} tons of cellulose are synthesized annually through photosynthesis in a rather pure form, for example, in the seed hairs of the cotton plant, but primarily cellulose combines with lignin and other polysaccharides in the cell wall of woody plants. Cellulose-containing materials also include agricultural residues, aquatic plants, grasses, and other plant matter. Commercial cellulose production is based on harvested sources, such as wood, or on natural sources with high biopolymer content, such as cotton. Cellulose is a linear polymer without winding and branching, and the numerous hydroxyl groups in cellulose form strong hydrogen bonds, which make the material unique. To achieve greater elasticity, a chemical modification is often applied, which involves the replacement of hydroxyl groups with acetate or methyl groups (esterification) to reduce the strength of the hydrogen bonds established by the hydroxyl groups. The most common cellulose esters are methylcellulose (MC) and carboxymethylcellulose (CMC), which have good film-forming properties and allow them to be used as materials for packaging food products.[14]

Pectin is a poly-α-1-4-galacturonic acid with varying degrees of methylation of carboxylic acid residues and/or amidated polygalacturonic acids, and it is one of the main components of plant cell walls, contributing to the integrity and rigidity of tissues. It is considered one of the most complex macromolecules in nature. Carboxylic groups of galacturonic acid are esterified with methanol, resulting in methoxylated carboxylic groups. Conversely, amidated carboxylic groups are obtained when galacturonic acid is converted with ammonia into carboxylic acid amide. Pectin is an ingredient used in the food industry without any restriction beyond current good manufacturing practices, and it is generally recognized as safe (GRAS) by the Food and Drug Administration (FDA) of the United States. Its ubiquitous presence, low cost, structural flexibility, and polymerization capacity contribute to its use as a matrix for active food packaging materials. Since bioactive packaging films made with pectin exhibit very weak antimicrobial properties, their antimicrobial potential can be enhanced by integrating and combining them with various functional compounds such as essential oils, phenolic compounds, nanomaterials, free fatty acids, and others.

Chitosan is a linear polysaccharide composed of randomly linked units of β-(1,4)-D-glucosamine and N-acetyl-D-glucosamine. Chitin is the second most common structural polysaccharide found in nature after cellulose and is typically deacetylated by an alkali for chitosan production. Chitin can be converted into chitosan through enzymatic treatment or via a chemical process. Chemical methods are widely employed for commercial purposes in chitosan production due to their low cost and suitability for mass production. Chitosan possesses numerous advantageous properties including biodegradability, antimicrobial properties, and non-toxicity.

Chitosan films can be produced, but limitations still exist when compared to plastic films as they are rigid and brittle, necessitating the use of plasticizers to enhance their mechanical properties. Compared to other biopolymers, chitosan has an advantage due to its ability to encapsulate functional substances such as minerals or vitamins and its antibacterial activity, which is important for maintaining product quality.[9]

Gelatin is a water-soluble protein obtained by partial hydrolysis of collagen, typically from pig skin for large-scale food production. However, in recent years, fish gelatin production has gained ground as an alternative, although it presents some challenges related to rheological properties and price competitiveness. Gelatin is composed of a heterogeneous mixture of α, β, and γ polypeptide chains.

Gelatin, obtained from collagen treated with acid (type A) or alkali (type B), possesses various functional properties. These include the ability to form protective external films for food, stabilize emulsions and foams, and gel and thicken. However, gelatin has limited thermal and mechanical stability during processing. Scientific studies have examined the addition of reinforcing agents and plasticizers to enhance its properties and increase the shelf life of food products using natural extracts as crosslinking agents to improve gelatin's strength. These modifications make gelatin more durable and versatile for a range of industrial and food applications.[9]

Collagen is a fundamental protein found in the extracellular matrix of vertebrates, accounting for approximately 30% of the total body protein mass. It is absent in plants and unicellular organisms and is found in the body walls of invertebrates. It is mainly extracted from bovine hides, which are rich in collagen in their inner layer, with a denaturation temperature higher than that of marine collagen. This protein consists of three chains of α-helices twisted into a right-handed triple helix, with characteristic amino acid sequences. Collagen self-associates to form highly organized and cross-linked fibrils, providing strength and mechanical integrity to the extracellular matrix. There are various types of collagens, grouped into families based on their structure and position in the organism.

Collagen, though constituting only a small fraction of the total, is water-soluble, with its solubility varying depending on tissue type and age. It is commonly extracted using neutral solutions or diluted acetic acid as solvents. It can be utilized to produce edible films in the meat industry due to its favorable mechanical properties. These films, primarily composed of fibrillar collagen, easily adapt to meat processing, aiding in maintaining quality and extending product preservation.[15]

Casein is the main protein extracted from milk, accounting for about 80% of its composition. It is divided into four different protein fractions: αS1-, αS2-, β-, and κ-casein, which together form colloidal micelles in milk. These micelles are stabilized by casein structures and calcium-phosphate bridges. Casein is appreciated for its nutritional properties, solubility in water, and emulsifying ability. It is also a suitable hydrocolloid for the formation of edible films among proteins, with its potential influenced by the four protein fractions comprising it.

Global production of caseins and caseinates is challenging to determine due to a lack of significant data. The major producers include New Zealand, the Netherlands, and Germany. Casein is derived from skim milk through either acid precipitation or rennet, while caseinates are water-soluble derivatives of acid caseins. Food-grade casein is manufactured under hygienic conditions and subjected to sufficient heat treatment for safety. The main types of casein are acid and rennet, each with distinct characteristics. Casein films exhibit good barrier properties against oxygen and other non-polar molecules due to the distribution of polar amino acids along the protein chain, allowing them to protect products prone to oxidation. Food-grade plasticizers like glycerin or sorbitol are added to the film-forming solution to address this issue. While plasticizers increase the thermoplastic of the protein film, they also decrease its strength. Although casein films have the potential for use in food packaging, some disadvantages must be addressed before they can be widely employed commercially. Casein-based films are highly sensitive to moisture, absorbing and releasing water molecules, which significantly impacts their mechanical and barrier properties. Additionally, they are predominantly soluble in water, limiting their application areas. It is also worth noting that plasticized casein films cannot provide high mechanical strength or good elasticity compared to synthetic polymer materials. Therefore, research is underway to identify new, cost-effective, and safe crosslinking agents for use in the food industry.[9]

The material that continues to be predominantly used in the world of food packaging is polyethylene (PE), a thermoplastic polymer derived from petroleum and thus synthetic, non-biodegradable, and mostly non-renewable. After briefly describing biopolymers and their importance, we understand how much work is needed to achieve the results of using these materials compared to currentones.[16]

The main critical aspect still present today is related to the barrier properties against external agents, which are crucial to prevent food deterioration typically caused by oxidation, microbial decay, and metabolism influenced by surrounding factors such as temperature and humidity.[7]

The primary function of food packaging is precisely to ensure the protection of the food throughout its storage duration and consequently food safety while maximizing the food's shelf life. There are numerous categories of food packaging that have been specifically developed based on the requirements requested by food producers and, in turn, by consumers.[17]

The importance of reducing gas transpiration to keep fruits and vegetables fresh is emphasized.

This is achieved by using materials with adequate barrier properties and carefully controlling factors such as humidity, temperature, light, and gases within the storage environment.

The need to protect dairy products from oxidation and microbial growth, which can compromise their freshness and safety, is highlighted.

Proper packaging can mitigate these factors by creating an effective barrier against oxygen, light, and moisture, which are the main causes of deterioration.

The issue of meat-based products, which can negatively impact their appearance and freshness, is discussed. Techniques such as vacuum packaging or modified atmosphere packaging can prevent this phenomenon by creating a controlled environment around the product that reduces oxidation and the action of bacteria responsible for discoloration.[18]

The purpose of food packaging is not only to protect the food from the external environment, but also to contain it, inform the consumer about nutritional values, and provide a convenient mode of transportation that meets the demands of both the industry and consumers while minimizing environmental impact. Below is an image depicting the overall functions of food packaging.[19]

FIGURE 2 FUNCTIONS OF PRIMARY PACKAGING[19]

Protecting food from the external environment is crucial for packaging. Without it, food could deteriorate, become unappealing, or unsafe to consume. Packaging must guard against physical damage, physiochemical deterioration, microbial contamination, and tampering. While active packaging can enhance protection with technologies like oxygen scavengers, it's important to note that packaging can't always shield against internal factors, such as moisture migration within foods. In these cases, innovative solutions like edible coatings may be necessary.

Food packaging plays a fundamental role in adapting to the hectic lifestyle of modern consumers by offering convenience through solutions like ready-to-eat meals and packaging that allows for easy opening and closing.[19]At the same time, it serves as an essential communication tool for branding and nutritional information, with mandatory labels providing data such as calories and fats. Its containment function is crucial, ensuring that food is held securely, considering its physical characteristics, and maintaining freshness and product quality. Ultimately, the success of a food packaging technology depends on its ability to fulfill at least one of these essential functions.[20]

Currently, food packaging is undergoing dynamic changes, becoming increasingly functional and innovative, containing active substances that interact with the packaged product. The traditional and passive protective function of packaging, intended as a passive barrier to protect food from harmful external factors, has been replaced by active protection.

Today's active packaging extends the functionality of traditional packaging with new elements. Traditional packaging maintains the shape, color, and flavor of the product, protecting it from mechanical, microbiological, physical, and chemical impurities and preventing the loss of product ingredients or the entry of unwanted substances from the outside. Additionally, it advertises the product through appropriate selection of packaging marketing values, such as shape, color, and typography. Traditional packaging is there to protect food and preserve its shelf life as long as possible, minimizing the interaction between packaging and the product. Active packaging, meanwhile, deliberately utilizes existing interaction.[19,20]

Active packaging integrates traditional packaging with new functionalities that optimize the conditions inside the packaging of a food product, significantly extending its shelf life. It is expected that food producers will increasingly resort to active packaging to better protect food and enhance the attractiveness of their products on one hand and provide greater safety to their customers on the other. In addition to protecting the product itself, active packaging plays an additional role in protection against external influences. Its main functional principle is to interact with the packaged product. The concept of active packaging is based on modifying the conditions inside the package, thus prolonging the life of the products. The interaction between the product and the packaging is very important and extends the shelf life or improves the sensory properties of the product. In this type of packaging, two methods are used to introduce active agents: they are placed in small bags in the packaging or directly in the packaging material.[20]

The selection of the right packaging material and shape for a particular food product depends on many factors. The most important of these are directly related to the physicochemical properties of the packaged item, including, for example, chemical composition, physical condition, texture, porosity, as well as storage time and conditions under which the product will remain until consumption. Equally important is understanding the processes (mechanisms) and factors that cause physical, chemical, biochemical, and biological changes that occur in the product during storage, limiting its expiration date.[21]

Active packaging represents a broad and diverse group both in terms of its purpose and the solutions applied. The use of proper active packaging extends the shelf life of products through its impact on emerging processes in food:

- Physiological processes, such as the respiration of fresh fruits and vegetables.
- Chemical processes, like fat oxidation.
- Physical processes, in the case of bread hardening.
- Microbiological changes due to the impact of microorganisms.
- Insect infestations.

The atmosphere inside the packaging can be actively controlled by substances that absorb (scavengers) or release (emitters) gases or vapor.

Scavengers are designed to remove unwanted components from the environment inside the packaging. There is no direct migration between a scavenger and a product but rather an improvement of conditions inside the packaging, which extends the product's shelf life. Depending on the application, it can be associated with the absorption of oxygen, moisture, ethylene, or carbon dioxide. To achieve specific effects, substances such as zeolite, cellulose, activated carbon, silica gel, iron ions, ascorbic acid, potassium permanganate, and calcium hydroxide are applied.

The second group of packaging consists of emitters. The operating principle of emitters is based on the release of desired substances that have a positive impact on food within the packaging environment. Such packaging contains and produces compounds capable of penetrating inside the packaging and inhibiting adverse processes.[21]

They are intended to ensure stable conditions during storage and should guarantee the extension of shelf life. It is thanks to emitters that moisture inside the packaging (vegetable packaging) can be controlled, the growth of harmful microorganisms (CO_2, SO_2, and ethanol emitters) can be inhibited, and bacterial deterioration can be prevented (antibacterials). Emitters can be fragrant substances, food additives, food ingredients, moisture regulators, and biologically active substances that prevent the growth of microorganisms.[19]

The most used antimicrobial substances are ethanol, sulfur dioxide, and carbon dioxide.

It should be noted, however, that the development of new products increases the risk of the emergence of new packaging-related hazards. For this reason, active packaging is not without disadvantages, among which one should mention especially higher costs of use and excessive migration of chemicals (Fig.3). In addition, improper labeling poses a significant threat to consumer safety. The safety of active materials and products intended to meet food is, therefore, regulated by law. All such products must be produced according to good manufacturing practice.[21]

Active packaging	
Disadvantages	Advantages
More expensive than the traditional packaging Certain substances released because of activepackaging can affect the composition of food In the event of damage to the packaging harmfulchemical reactions may occur Requires more knowledge and consumerawareness Some of the compounds used for their production may be deposited on the surface of fruits and due to their strong characteristic smell,they can lead to rejection of the product by the consumer Can be used in an unethical manner for food adulteration -using lower class raw materials ornot observing and neglecting strict principles ofgood manufacturing practice	Can control the internal conditions, reacting to them accordingly by emitting beneficial substancesof absorbing those that negatively affect the product. Detects the presence of metabolites of microorganisms, carbon dioxide, ammonia, sulfur dioxide, hydrogen sulphide, ethanol, and organic acids or amines. Allows for longer shelf life and maintaining the product intact, including sensory properties of food products. Can reduce the use of food preservatives Contributes to the protection of the environment using solutions that are biodegradable and biocompatible

TABLE 1 ACTIVE PACKAGING—ADVANTAGES AND DISADVANTAGES.[21]

Over the past years, many innovations in the packaging industry have led to the transition from the concept of passive food packaging, active packaging (AP) described above, to the model of intelligent or smart food packaging (IOSP) and sustainable or green packaging (SOGP).[22]

Passive packaging has focused more on mechanical strength, barrier performance, and thermal stability, while active packaging has involved the integration of oxygen/radical/ethylene scavengers, moisture absorbers, carbon dioxide emitters, and antimicrobial compounds into the packaging.

In this case, IOSP has been realized using time-temperature indicators, gas indicators, microwave cooking indicators, and radiofrequency identification, among others, and SOGP has gained importance in the market due to urgent and significant concerns about the environmental impacts of food packaging waste.

SOGP aims to develop materials for food packaging with minimal environmental impact. The impact generally depends on how materials are produced and processed, and the end-of-life stage of the packaging material, including recycling, incineration, landfill disposal, and composting. SOGP involves three aspects: 1) raw materials, 2) the production process, and 3) waste management.[22]

SOGP prefers 1) materials from renewable resources or recycled materials to eliminate CO2 emissions and reduce the use of petrochemicals, 2) lighter and thinner packaging with economically and energetically efficient processes, and 3) materials that are biodegradable, compostable, recyclable, or reusable to minimize harmful effects on the environment.[22]

Although various techniques have been applied to food packaging, safety issues are widely concerning, especially for active packaging and packaging techniques involving nanotechnology. Some regulations, such as EU Regulation 1935/2004, EU Regulation 10/2011, and Chapter 21 of the Code of Federal Regulations in the United States, establish concrete regulations, specific details, and requirements for food packaging materials.[21]

The migration of harmful substances and other hazardous food additives is the main concern. The objective for the coming years will be to highlight bio-based materials for food packaging so that they can find good scalability and application in this sector. Bio-based polymers are chemically unique polymer molecules, and the sizes of polymers are at the molecular level, which can vary with the category and origin of the polymers.[22]

Most bio-based polymers should have a length ranging from tens to thousands of nanometers and can be enhanced in terms of properties and performance using bio-based nanomaterials such as nanocrystals and nanofibers, which contain many polymer chains with sizes ranging from several nanometers to several micrometers. Practical applications of these food packaging can involve various types of packaging, and properties such as mechanical strength, barrier properties, antimicrobial activity, and antioxidant functions are discussed. The primary advantages of each material and the related operating mechanisms are mainly due to environmental sustainability aspects, which are extremely important and delicate nowadays.[22]

The progressive development towards sustainable packaging arises from the dual need to source materials other than fossil resources (which are constantly diminishing) and to mitigate the ecological damage caused by their use.[22]

There are numerous advantages derived from this approach, such as biodegradability, compostability, the ability to reuse waste materials, and the use of materials that, by showing minimal moisture loss, reduce oxidation processes and enhance the aromatic qualities of food. Additionally, they preserve the original color and resistance to attacks by pathogens.

There is a strong focus on reducing environmental impact, which typically depends on how materials are produced and processed, as well as the end-of-life stage of packaging materials, including recycling, incineration, landfill disposal, and composting.[23]

In recent years, there has been a growing global interest in environmental protection. Sustainable packaging plays a crucial role in reducing waste and pollution and promoting sustainable development. Eco-green packaging, also referred to as 'eco-friendly packaging', 'sustainable packaging', or 'recyclable packaging', utilizes environmentally friendly materials for packaging purposes, always keeping in mind that products must be effective and safe for both human health and the environment.[23]

Research documents in this area can be broadly categorized into two main groups: those addressed from the consumer perspective and those that focus on businesses. The study of how consumers demand the use of eco-friendly packaging strategies to mitigate the negative environmental impact of packaging is as important as the actions taken by companies to implement eco-friendly packaging. These actions cover a wide range of dimensions, including technological, organizational, and human capabilities that contribute to the implementation of innovation in packaging eco-design and its benefits in terms of brand innovation and environmental protection.[24]

Sustainable packaging is a relatively new concept that has garnered significant attention in recent years and is a key theme to consider for achieving Sustainable Development Goals, with social and economic implications.[24]

To be recognized as environmentally friendly, packaging must meet several key environmental aspects. Firstly, it's essential to use sustainable raw materials sourced from renewable, biodegradable, or recyclable sources. This involves utilizing materials like sustainably managed forest-derived cellulose, biodegradable polymers obtained from plant sources such as corn or sugarcane, and recyclable materials like aluminum and glass and paper.[24]

Moreover, it's crucial to minimize the use of natural resources during packaging production and usage by optimizing production processes to reduce waste and adopting advanced technologies requiring less energy and resources. This helps reduce the overall environmental impact of packaging and preserves natural resources for future generations.[23]

Another important consideration is energy efficiency, which entails reducing the energy required to produce, transport, and dispose of the packaging. This can be achieved by adopting renewable energy sources to power production and logistics processes and implementing energy-efficient practices such as using low-energy machinery and optimizing supply chains.

Packaging recyclability is fundamental for facilitating material separation and recycling after use. This requires using materials that can be easily separated and processed in existing recycling systems, along with proper labeling to indicate correct disposal methods.[23]

Biodegradability and compostability are other important features to consider. Biodegradable materials can naturally decompose without causing environmental harm, while compostable materials can be transformed into usable compost for agriculture. This helps reduce the overall amount of waste sent to landfills and promotes material circularity.[19]

In addition to environmental aspects, it's important to consider social impacts throughout the entire packaging supply chain, including workers' rights and local communities. This involves ensuring safe and fair working conditions for workers involved in packaging production and distribution and involving local communities in decisions regarding the environmental impact of packaging.[24]

Finally, packaging should be designed to be robust and functional, effectively protecting the product during transportation and use, and traceable to monitor and make transparent the entire supply chain.

When all these aspects are taken into consideration and implemented, sustainable packaging can be achieved, significantly reducing environmental impact compared to before.

It's possible to assess environmental aspects through Life Cycle Assessment (LCA) studies and by analyzing environmental indicators such as those listed below, recognized as drivers of packaging eco-design[25]:

1. Globa Warming Potential (GWP): assesses the emission of all gases that contribute to the greenhouse effect jointly with CO_2. It is measured in mass of CO_2 equivalent by converting emissions of the various gases to CO_2 emissions based on conversion factors defined by the IPPC in 2013 (www.ipcc.ch)[26]

2. Gross Energy Requirement (GER) is an indicator expressed to megawatts of the total energy extracted throughout the life cycle of a functional unit of the product and/or service. Contributing to this indicator are the shares of energy consumed to power production processes, to produce the fuels used in processes and transportation stages, and the energy contained in raw materials.[26]

3. Water consumption: expressed in liters, it defines the amount of process water used in the production and marketing of consumer goods. This is blue water or net process water consumption.[25,26]Fare clic o toccare qui per immettere il testo.

Alongside sustainability and innovation aspects, all food packaging introduced to the market must comply with and meet the requirements established by legislation concerning safety and health. It is within the European Union that the requirements for materials and articles intended to come into contact with food, including food packaging, are contained in the framework regulation of the European Parliament and Council (EC) No. 1935/2004 of 27th October 2004 on materials and articles intended to come into contact with food and in Commission Regulation (EC) No. 450/2009 of 29th May 2009 on active and intelligent materials and articles intended to come into contact with food.

Regulation No. 1935/2004 was introduced to ensure quality and legal certainty for the application of materials that may actively influence the behavior of food or improve its condition, as well as for materials used to monitor its condition. According to this regulation, active materials and articles intended to meet food are designed to include ingredients that are gradually released into food or absorb substances contained in it. Materials and articles may modify the composition or organoleptic characteristics of food only if the changes are in accordance with the community provisions applicable to food.[21]

Article 3 of the framework regulation stipulates that all materials and articles intended to meet food must be safe; therefore, active packaging must also be sufficiently inert to prevent the transfer of their constituents into food in quantities that could endanger human health, cause unacceptable changes in the composition of food, or deterioration of the organoleptic characteristics of food under normal and foreseeable conditions of use. Meanwhile, according to Article 4 of the regulation, active materials and articles may lead to changes in the composition or organoleptic characteristics of food provided that these changes are in line with the provisions on food (food additives, flavors, and enzymes). However, such changes must not mask signs of the initial spoilage of food or mislead consumers.[21]

Active materials have been deliberately designed to contain active ingredients that may be gradually released into food or its environment or absorbed from food products in accordance with Regulation (EC) No. 1935/2004. It is crucial to distinguish materials that have natural properties to absorb or release substances, such as cellulose or wood, from materials that have been intentionally designed so that their components interact with the food they meet. Materials such as cellulose, despite interacting with food or its environment, are not considered active materials if not functionalized through surface chemical treatments and/or specific treatments that allow functional barriers to the cellulose substrate.[21]

Regarding the safety and health aspect of packaging, a very stringent regulation, often used as a reference in Europe, is the FDA regulation of the Food and Drug Administration (FDA), which establishes regulations and requirements to ensure the safety of packaging used for food products in the United States. These provisions are contained in Title 21 of the Code of Federal Regulations (CFR) of the United States of America. The CFR Title 21 covers a wide range of topics related to food and drugs, including standards for food packaging.[19]

The FDA regulation stipulates specific requirements for food packaging to ensure they are safe for contact with food and do not transfer harmful or unsafe substances to the food. These requirements include guidelines on the use of safe and approved materials for food packaging, as well as restrictions on the use of certain chemicals that could be harmful to human health. Additionally, the FDA regulation establishes criteria for the labeling of food packaging, which must be clear and accurate to inform consumers about the correct use of the packaging and the materials used in its production. These labels must comply with FDA standards to ensure that the information provided is truthful and not misleading.[19]

Overall, the FDA regulation on food packaging aims to protect public health by ensuring that packaging used for food products is safe, compliant with manufacturing standards, and adequately labeled to inform consumers.[21]

Categories of packaging: primary, secondary, tertiary, and specific materials.

The fundamental shift from traditional packaging to that of grade and functionality has challenged the concept of packaging ownership and introduced different packaging categories.

Packaging is recognized at different levels depending on its function, and the categories are listed below based on the layer and functionality.

Primary packaging is the packaging that wraps the product first and protects it, coming into direct contact with the food, therefore, it must comply with the safety and health regulations mentioned above.[27]

Secondary packaging is the outer packaging layer of the primary packaging and can be used to prevent theft or to group primary packages together.

Tertiary packaging or transport packaging is used for bulk handling, warehouse storage, and shipping. Recently, associations like "Stop Waste" and the "Coalition for Pallets" and "Reusable Containers" have provided a list of virtues that tertiary packaging, if designed to be reusable, brings to this sector's supply chain.[27]

Reusable transport packages improve workers' safety and ergonomics because their material and design reduce injuries that can occur if the tertiary packaging is designed to be disposed of after its use. Reusable transport packaging also provides just-in-time delivery of finished products because it provides standardized ordering quantities that can improve ordering procedures and inventory tracking.[27]

Reusable secondary packaging can have common advantages when combined with the mentioned tertiary packaging because the risk of packaging breakage during transportation is reduced, and the quality of the finished product delivered to the end user (consumer) is improved, as ventilated reusable containers increase shelf-life and freshness. The use of these packaging systems for shipping products in a supply chain can generate significant cost savings since the cost of reusable packages can be spread over several years. Additionally, both packaging systems can be advantageous from a waste management perspective, as they produce less waste to manage for recycling or disposal. Finally, one of the main reasons for using such packaging systems is their environmental impact. By using this type of containers, the need to build disposal facilities or recycling centers is reduced. Using this type of containers for product delivery can also reduce greenhouse gas emission rates and the overall energy consumption of the entire system.[27]

Regarding primary packaging, we can list three virtues for this category of packaging systems if designed following the approaches adopted for the other two categories of packaging: long-term food preservation ensuring excellent shelf-life, virtuous recycling once its use is finished becoming a new resource for new raw materials, bringing new specific functionalities according to market needs. Below is an image highlighting the packaging categories.[27]

FIGURE 3 CATEGORIES OF PACKAGING[27]

Additionally, special types of food packaging made using edible films and/or coatings are added, as well as packaging in modified atmosphere (Modified Atmosphere Packaging, MAP).[21]

A highly performing example is the so-called "active packaging" often used to preserve fresh foods such as fruits, which is achieved through a protective film coating often biobased.[21]

The characteristics of active packaging for the food sector include a series of properties designed to preserve the freshness, safety, and quality of foods, some of which are listed below:[28]

- Protective barrier: the packaging must provide an effective barrier against moisture, oxygen, light, and other external agents that could deteriorate the food product.
- Gas absorption: it may include active components that absorb gases such as oxygen to prolong the shelf-life of the product and reduce oxidation.
- Controlled release of substances: some active packaging can release antimicrobial or antioxidant substances in a controlled manner to prevent bacterial growth or food oxidation.
- Antimicrobial activity: antimicrobial agents can be integrated into the packaging to inhibit bacterial growth and prolong the shelf-life of foods.
- Moisture absorption: absorbent materials can be incorporated to control moisture inside the packaging and preserve the crispness or freshness of foods.
- Freshness indicator: some active packaging includes freshness indicators that change color or show visible signs when the product has exceeded its shelf-life.[17]

These are just some of the common characteristics of active packaging in the food sector, but specifications may vary depending on the type of food, production process, and customer requirements.

In the EU Directive 94/62/EC, it is written : packaging designed to constitute, at the point of sale, the grouping of a certain number of sales units regardless of whether it is sold as such to the end user or consumer, or whether it only serves to facilitate the replenishment of shelves at the point of sale. It can be removed from the product without altering its characteristics.[20]

Edible Films and Coatings as packing materials

Recently, innovative configurations have been developed compared to conventional primary packaging, involving the use of edible films and coatings capable of providing the key characteristics that biodegradable packaging should meet for specific uses in the preservation and enhancement of various food products.[29]

Special attention must be paid to the main components used (e.g., biopolymers, additives, bioactive components, and probiotics), production methods (for edible films or coatings), and their application to specific products. The applications of edible films and coatings as quality indicators for perishable products are still under development, with growth prospects expected in the coming years. An edible film or coating is any material with a thickness of less than 0.3 mm, formed from a combination of biopolymers and various additives dispersed in aqueous media. The edible coating is formed directly on the food, while the edible film is previously made and then adhered to the product.[29] The figure below illustrates the main characteristics that edible films and coatings can exhibit: protection againstUV light; transport of solutes (e.g., salts, additives, and pigments), water vapor, organic vapors (e.g.,aromas and solvents), and gases (e.g., oxygen, carbon dioxide, nitrogen, and ethylene) between foodand the atmosphere; barrier against mechanical damage (e.g., dents or cuts); increase in the product'sshelf-life; bioactive components (e.g., antioxidants); antimicrobial effect against bacterial reproductionand fungal contamination (e.g., silver nanoparticles); healthy microorganisms (e.g., probiotics) conferring benefits to the consumer; and biodegradable natural materials.[29]

FIGURE 4 ILLUSTRATING THE MAIN CHARACTERISTICS OF EDIBLE FILMS AND COATINGS.[29]

Below is a table listing the most used biopolymers and additives in the production of edible films and coatings, along with their properties and functionalities in packaging.

Active packaging		
Materials	Examples	Properties
Polysaccharides	Starch Cellulose Chitosan Alginate	They form the base structure of a solid polymer matrix
Proteins	Gelatin Casein Whey protein	They help in the transport of antioxidants. They control the transport of gases
Lipids	Waxes Paraffin Glycerides	They help to avoid drying or dehydrationof the edible film providing flexibility
Plasticizers	Glycerol Aloe	They also modify the viscosity and the rheological properties
Others	Polyphenos	They work as stabilizers as well as protection for the products

TABLE 2 MAIN MATERIALS USED AND FUNCTIONALITY IN THE MANUFACTURE OF EDIBLE FILMS AND COATINGS.[29]

As can be seen from the table, starch is considered the universal biopolymer for biodegradable packaging, which has been widely used for decades, thanks to its characteristics and gelatinization properties.[29]

Alginate is another important biopolymer that demonstrates the ability to form hydrogels and encapsulation barriers.

Recently, chitosan has attracted attention for the development of edible films and coatings due to its properties as a gelling agent, as well as its chemical (ability to form hydrogen bonds and hydrophobic interactions) and biological (biocompatibility, biodegradability, and bioactivity) properties.[29]

On the other hand, the role of additives (such as plasticizers or stabilizers) in the formulation of edible films and coatings is to modify their mechanical properties. Additionally, the incorporation of antioxidant, fungicidal, or microbial additives allows to produce bioactive biodegradable packaging, which is expected to see growth in terms of application development in the coming years.[29]

It is worth noting that this concept of packaging can be further improved and advanced in terms of functional performance through the development of composite polymers. These are simple films combined with functional additives to achieve mechanical and barrier properties that meet the desired targets. For example, by adding vegetable oils to simple materials such as lecithin or casein films, high barrier performance to water vapor and/or stabilization of the film solution can be achieved.[30]

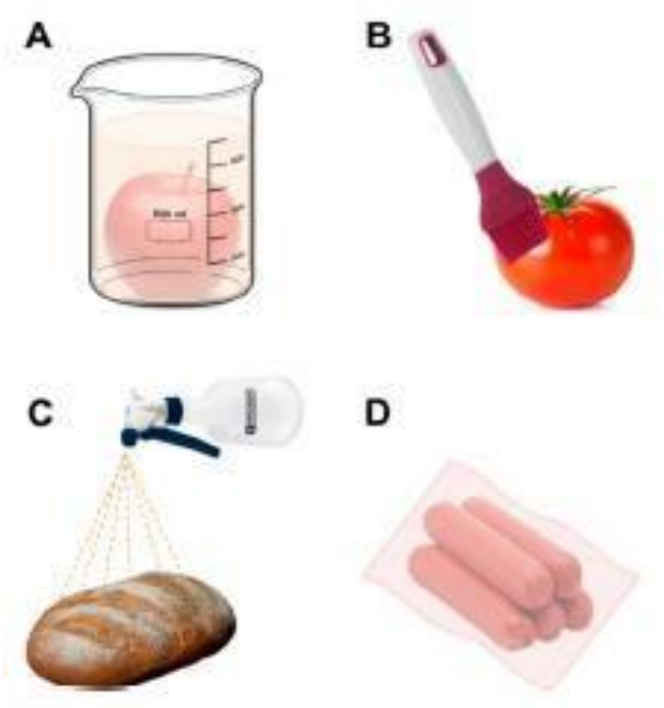


FIGURE 5 MAIN TECHNIQUES USED FOR FOOD COATING. (A) DIPPED; (B) SPREAD; (C) SPRAYED; (D)WRAPPED.[29]

These systems, presented in dispersed form, need to be incorporated and/or coated into the product to allow for proper drying and thus generate a rigid matrix that will act as an edible film or coating. This will strictly depend on the type of application protocol. The most used application techniques are outlined below and are of different types: immersion (A), spreading (B), spraying (C), and wrapping (D). Edible coating formulations are added and dried directly onto the surface of the food, while edible film formulations are poured into a mold and dried to be subsequently incorporated into the product (Figure D).[29]

It is also possible to add substances to these packaging systems that serve as colorimetric indicators to detect any product alterations due to external factors such as temperature and humidity variations.[23]

Bio-based materials and their composites for packaging applications

The food packaging industry in recent years has undergone a significant transformation to align with the model and approach towards the circular economy, replacing non-renewable packaging with more sustainable and biodegradable renewable materials.[31]

This approach is leading to a reduction in the use of virgin plastic and a consequent decrease in plastic waste, with more sustainable alternatives whose waste becomes biomass destined to become the basis for new materials and products.[32]

These materials and products will not deplete because they are continuously renewable and regenerable.[32]

The significant development of biopolymers has been observed in recent years primarily due to the increased environmental awareness among citizens as well as end consumers.

This aspect has encouraged polymer producers and companies across various sectors to utilize renewable raw materials to produce high-value-added products.[32,33]

The production capacities of polymers have consistently shown growth over the years, although their growth forecasts have changed in the last five years. Initially, there was talk of exponential growth in biopolymers, particularly concerning materials sourced from renewable and biodegradable sources.[31]

The bioplastics market for the packaging sector is currently experiencing an exponential growth phase, with the prospect of significant global growth in the coming years.

Currently, bioplastics constitute only about 0.5 percent of the vast annual plastic production, which exceeds 400 million tonnes but is destined to grow significantly in the coming years.

After a period of stagnation in recent years, the total global plastic production is once again registering an increase starting from 2023.[34]

This trend is driven by the increasing demand for plastic, combined with the emergence of increasingly sophisticated applications and products that require more sustainable and biodegradable solutions.[34]

The global production capacity of bioplastics is projected to experience significant growth in the period under consideration, increasing from around 2.18 million tonnes in 2023 to approximately 7.43 million tonnes in 2028.

This increase represents a response to the growing demand for sustainable alternatives to conventional plastic and reflects the industry's efforts to adopt more eco-friendly practices.[34]

Below are two representative graphs regarding the projected production growth of bioplastics.

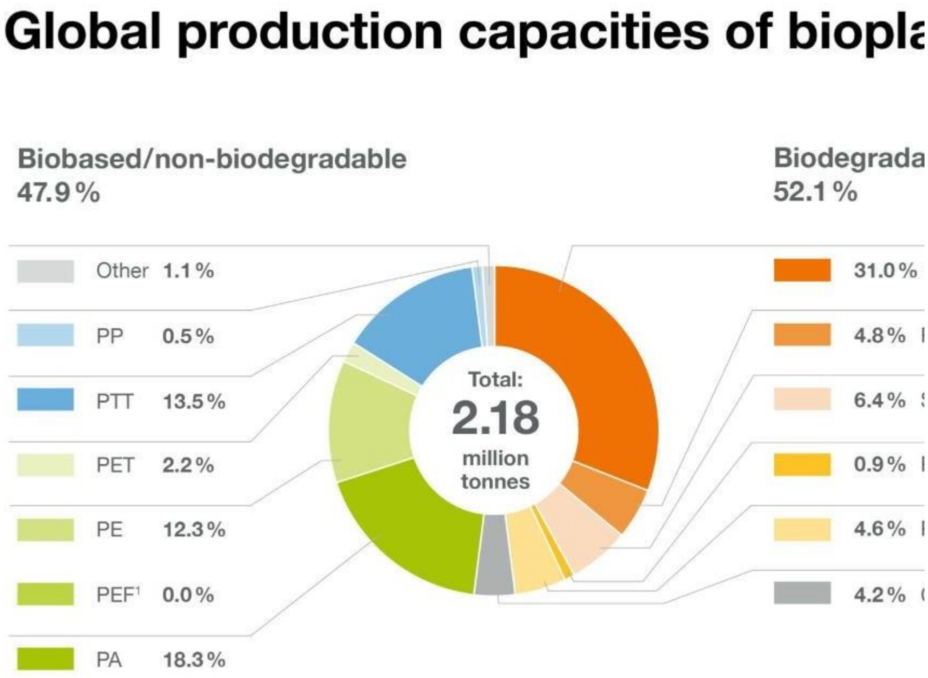


FIGURE 6 OVERVIEW OF GLOBAL PRODUCTION CAPACITIESOF BIOPLASTICS2023.[33,34]

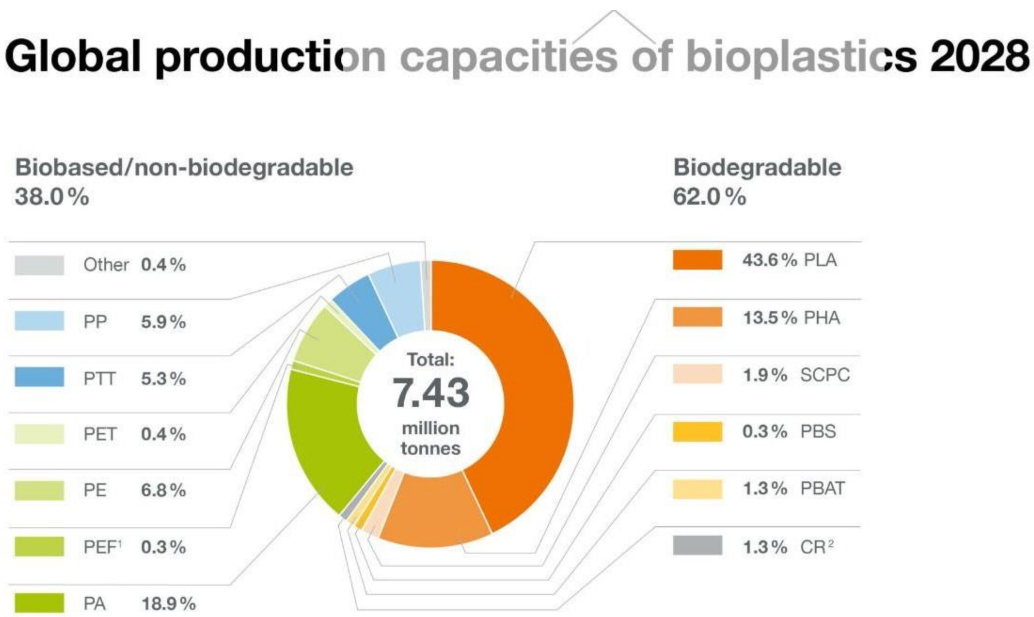


FIGURE 7 OVERVIEW OF GLOBAL PRODUCTION CAPACITIES OF BIOPLASTICS 2028.[34]

*PEF is currently in development and predicted to be avaible in commercial scale in 2024; CR = regenerated cellulose films

As can be observed in the fig.8-9 above, in previous years there have been global development implementations regarding the production and market introduction of bioplastics.

The increase in bioplastics production that was anticipated has been confirmed, driven by the growing demand for sustainable alternatives to conventional plastics. This increase has been fueled by greater environmental awareness and the increasing demand for biodegradable and compostable materials.[34] Bioplastics are gaining popularity in a wide range of sectors, including packaging, automotive, electronics, and construction, as companies seek more eco-friendly solutions to reduce the environmental impact of their products and processes.[33] The growth prospects for bioplastics by 2028 compared to 2023 are even more promising. There is expected to be a further significant increase in global bioplastics production capacity over these years, from 2.18 million tonnes to 7.43 million tonnes. As can be seen from the graphs, the increase in bioplastics will involve both non-biodegradable and biodegradable biobased materials. The overall advantage of further increasing the production and use of bioplastics is closely linked to the renewability factor of the raw material, and therefore the aspect related to the raw material that will never be depleted and will never have an end. Continuous research and development of new, more effective bioplastic technologies and materials will also support this growth. Additionally, increased consumer awareness of the importance of environmental sustainability could accelerate the adoption and use of bioplastics in the coming years.[34]

The development and diversification of bioplastic materials represent a significant opportunity to replace virtually every type of traditional plastic material and meet a wide range of application needs. Thanks to the continuous advancement of bioplastic polymers, such as polylactic acid (PLA), polyhydroxyalkanoates (PHA), polyamides (PA), and the steady growth of polypropylene (PP), it is expected that the production capacities of these materials will continue to expand significantly over the next 5 years.[24,31]

This development is driven by the increasing demand for sustainable alternatives to conventional plastics, both from consumers and businesses seeking more eco-friendly solutions to reduce the environmental impact of their products and processes. Bioplastic materials offer advantages such as biodegradability, compostability, and derivation from renewable sources, compared to traditional petroleum-derived plastics.[33] Furthermore, innovation and ongoing research in the bioplastics sector are leading to the development of materials with ever-improving performance, suitable for a wide variety of industrial and commercial applications. This trend is expected to contribute to the growth and dissemination of bioplastics as a sustainable and responsible alternative in the coming years.[34]

It is essential to clarify the terminology governing this field since it does not involve a single material but rather a family of polymers with sometimes similar, sometimes contrasting characteristics. Once again, the "European Bioplastic" has provided specific terminology, defining the term "biopolymer" to encompass both bio-based materials and biodegradable materials, as well as the combination of the two, as can be observed from the graphical representation they issued[34].

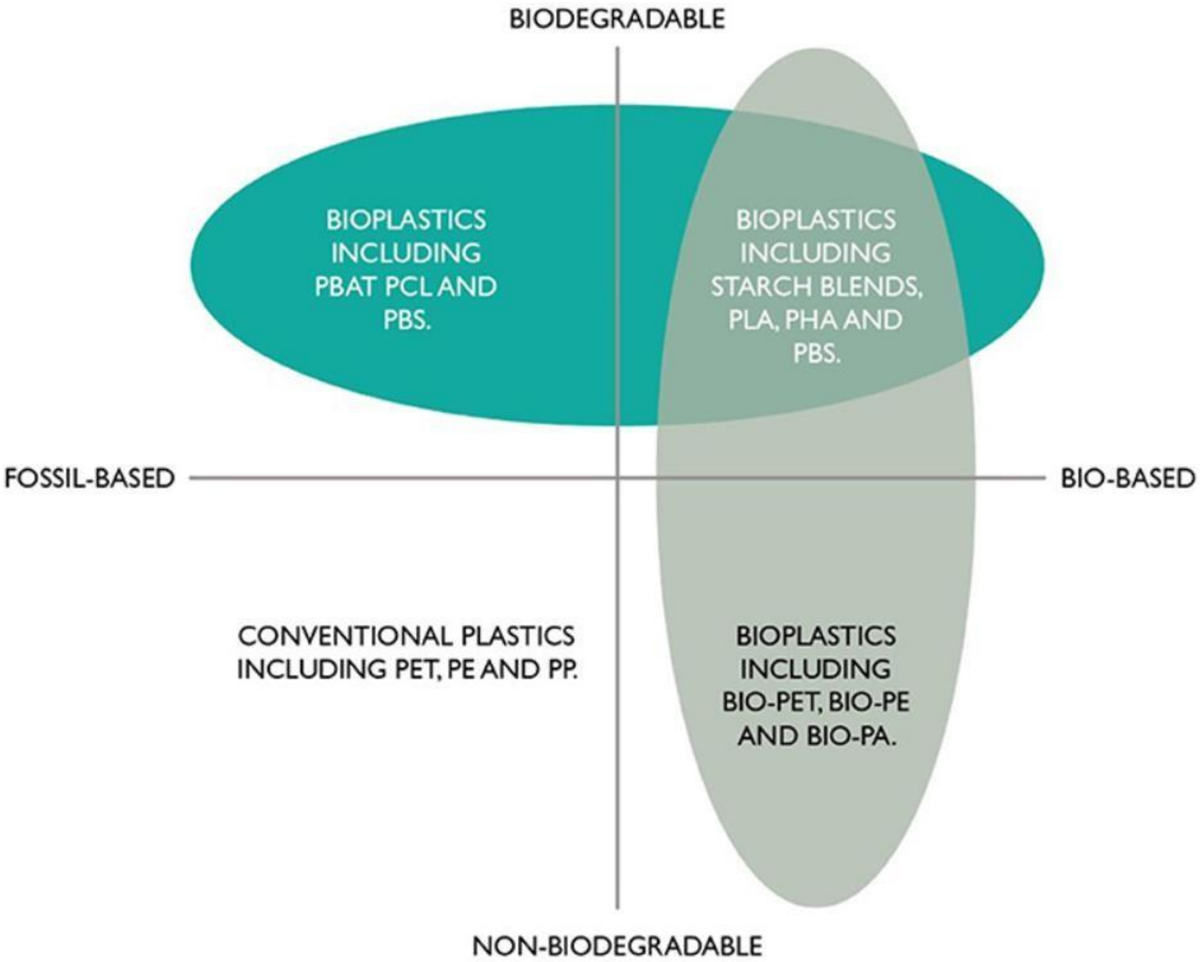


Figure 8 Overview of Bioplastics; macro category and the main polymers it includes.[35]

Above, we present a representative chart that aims to encapsulate the meaning of bioplastic.

The term "biobased" is not synonymous with "biodegradable"; therefore, a biobased plastic can be either biodegradable or non-biodegradable, and vice versa. This distinction is crucial in the field of bioplastics, as it helps define the specific properties and characteristics of each type of plastic material.[35]

There are three types of bioplastics as follows[35]:

- Non-biodegradable bioplastics: These plastics are made entirely or partially from biobased materials but are not biodegradable. For example, biobased polyethylene and polyethylene terephthalate, produced from sugar cane, fall into this category.
- Biobased and biodegradable bioplastics: These plastics are composed of biobased materials and can degrade into compost or other biodegradable materials. A common example of this type is starch blends.
- Fossil-based biodegradable bioplastics: These plastics are biodegradable but not biobased, meaning they are made from fossil raw materials but can still be decomposed by microorganisms. This type of bioplastics includes, for example, oxo-biodegradable polyethylene.[35]

Biobased bioplastics can be produced partially or entirely using plant biomass, for example, using renewable resources such as plants, algae, marine organisms, or organic waste (such as sugar cane or corn starch).

These materials are fermented in contact with bacteria and yeasts to create many products.

Some bioplastics, defined as durable, are instead mixed with petroleum-based plastic polymers.[36]

Within these three main categories, various biopolymeric materials are classified, as illustrated in the figure below, where specific biopolymers are identified.[36]

Bio-based materials can be entirely or partially sourced from renewable sources and may not necessarily be biodegradable or compostable at the end of their life cycle. Some examples include bio-polyolefins, particularly bio-PE, bio-PA, and bio-PET.[36]

Above is a representation of the macro families of biopolymers and their properties, specifically listing the most common names of bioplastics based on their origin and properties.

Bio-based materials are derived from renewable sources such as plants, algae, or organic waste and can be used partially or entirely to produce plastic polymers. These materials can help reduce dependence on non-renewable resources and mitigate the environmental impact of plastic waste derived from fossil sources. However, it's important to note that the presence of bio-based components does not automatically imply biodegradability or compostability properties at the end of the material's life cycle.[35]

Exclusively biodegradable materials are designed to biologically degrade through the action of microorganisms present in the disposal environment. These materials may come from non-vegetable and natural sources, such as polybutylene adipate terephthalate (PBAT), and can be broken down into simpler forms until reaching a state of final biodegradability, converting into water and carbon dioxide. This process of biological decomposition contributes to the sustainable management of plastic waste and the conservation of environmental resources.[37]

There are materials that combine both characteristics, being both bio-based and biodegradable. These materials, such as polylactic acid (PLA), polyhydroxyalkanoates (PHA), polyhydroxybutyrates, and starch-based blends, are produced from renewable sources and can biologically degrade at the end of their life cycle. These materials represent a more sustainable choice compared to materials derived entirely from non-renewable sources, thereby contributing to responsible plastic waste management and environmental resource conservation.[37]

A particular form of biodegradability is composability. The biological process of composting generates carbon dioxide, biogas, water, inorganic substances, and biomass. It's worth noting that biodegradation is a natural process occurring wherever conditions are favorable, while composting is a controlled process.

The European standard EN 13432 (Requirements for packaging recoverable through composting and biodegradation - testing scheme and evaluation criteria for final packaging acceleration) determines whether a plastic can be defined as compostable and treated as such.[31]

Here's a further breakdown in the classification of biobased polymers, along with a list of polymers classified by the source of their renewable starting material:

1) Polymers of polysaccharide origin such as chitin, chitosan, starch, and cellulose, and of protein origin such as casein.

2) Polymers synthesized from monomers derived from biomass, such as polylactic acid (PLA) and bio-polyethylene (Bio-PE).

3) Polymers produced by microorganisms, such as polyhydroxyalkanoates (PHA) e.g. Poly hydroxybutyrates (PHB), bacterial polyesters.

4) Biodegradable polymers synthesized from petrochemical monomers, such as polycaprolactone (PCL), poly (butylene succinate-co-adipate) (PBSA), polybutylene adipate terephthalate (PBAT), poly (glycolic acid) (PGA), polybutylene succinate (PBS), and polycarbonate propylene (PPC).

Below are the different categories of biopolymers and their uses in the food packaging sector[38].

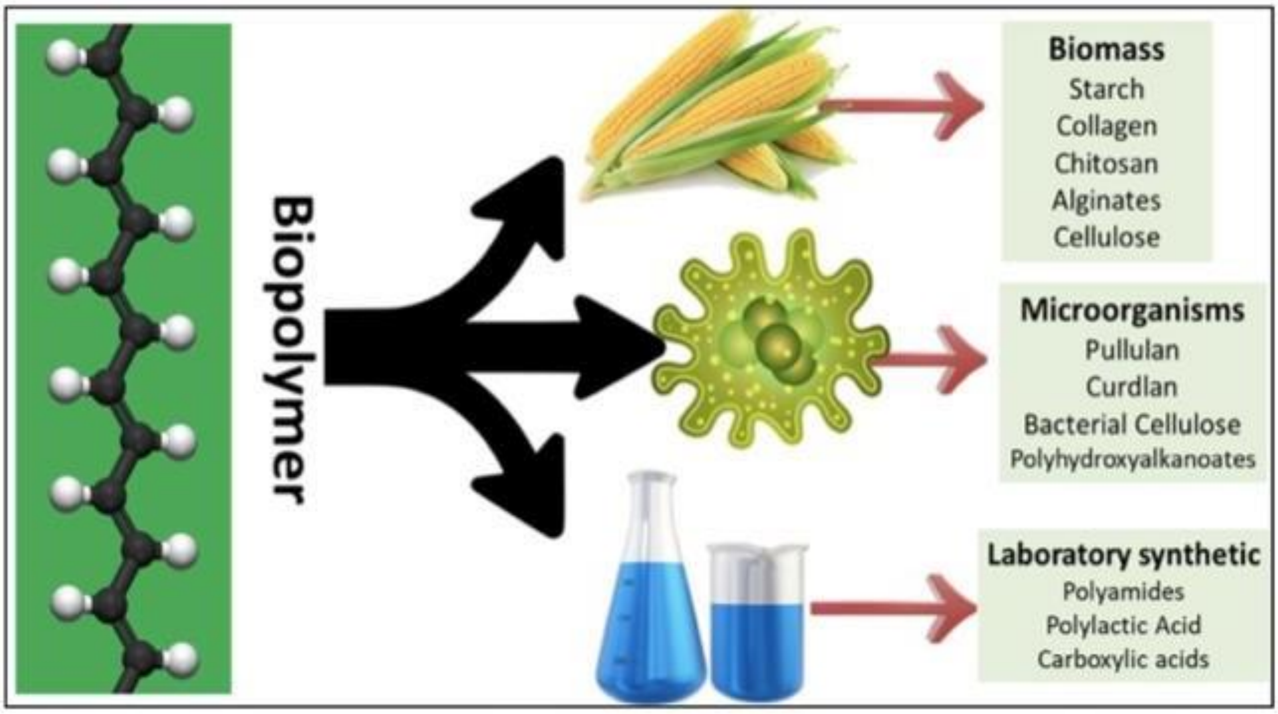


FIGURE 9 CLASSIFICATION AND ORIGIN OF BIOPOLYMERS.[38]

Biopolymers, naturally occurring renewable and abundant polymers, are classified into three groups based on their origin. Some biopolymers, such as starch, cellulose, chitosan, and alginates, can be extracted from biomass through various treatments (chemical, mechanical, or biological).

Similarly, biopolymers, including pullulan, curdlan and bacterial cellulose, can be derived from microorganisms.

A new class of biopolymers, such as polyamides, polylactic acid, and carboxylic acids, can be synthetically prepared in laboratories.[38]

Composition and properties of some common biopolymers that are used in food packaging applications below:

Polysaccharide	Composition	Properties
Alginate	Mannuronic glucuronic acid	Biodegradable High water permeabilityCross-link with calcium
hitin	N-acetylglucosamine	Transparent Non-toxic Biodegradab le Biocompatibl e Antifungal and antibacterial
Cellulose	Glucose	Biodegradable Transparent Sensitive to water Good mechanical properties
Chitosan	D-Glucosamine N-aceyl-D-glucosamine	Biodegradable Non-toxic Barrier to gases
Xanthan gum	Glucose, mannose, pyruvate	Edible Biodegradle

TABLE 3 COMPOSITION AND PROPERTIES OF SOME COMMON BIOPOLYMERS THAT ARE USED IN FOOD PACKAGING APPLICATIONS.[38]

As can be observed in the table, all the polysaccharides listed are biodegradable, and some of them inherently possess good barrier properties against gases and lipid substances.

The properties of some important biopolymers are presented in the table above, and among various biopolymers, cellulose is certainly one of the most abundant, sustainable, and promising bio-based polymers. Millimeter-sized strings composed of continuous microfibers and microfibers containing nanometer-sized microfibrils form the fundamental structure of cellulose.[35]

Cellulose is expected to be the least expensive, non-toxic, and have a remarkable strength-to-weight involve the food packaging sector using specially designed composite materials in accordance with the end application and current legal requirements and regulations.[36,38]

Biopolymers

The origin and production process of biopolymers play a fundamental role in their classification and typology, directly influencing their biodegradability and compostability properties.

These aspects are of vital importance in addressing the issue of plastic waste and promoting sustainable solutions in the industry.[11]While many of the listed biopolymers demonstrate good gas barrier properties, there is still room for improvement that can be explored through various approaches. For example, functionalization of functional groups or composite production can further optimize their performance. Through targeted chemical modifications, it is possible to enhance the barrier capabilities of biopolymers by intervening in factors such as hydrophilicity and crystallinity. These modifications will not only contribute to improving food preservation properties, extending shelf life, but will also make biopolymers even more competitive than traditional polymers.[39] To illustrate the effectiveness of biopolymers compared to conventional materials, a comparison of barrier properties has been prepared, highlighting the potential and advantages offered by these innovative materials.[11]

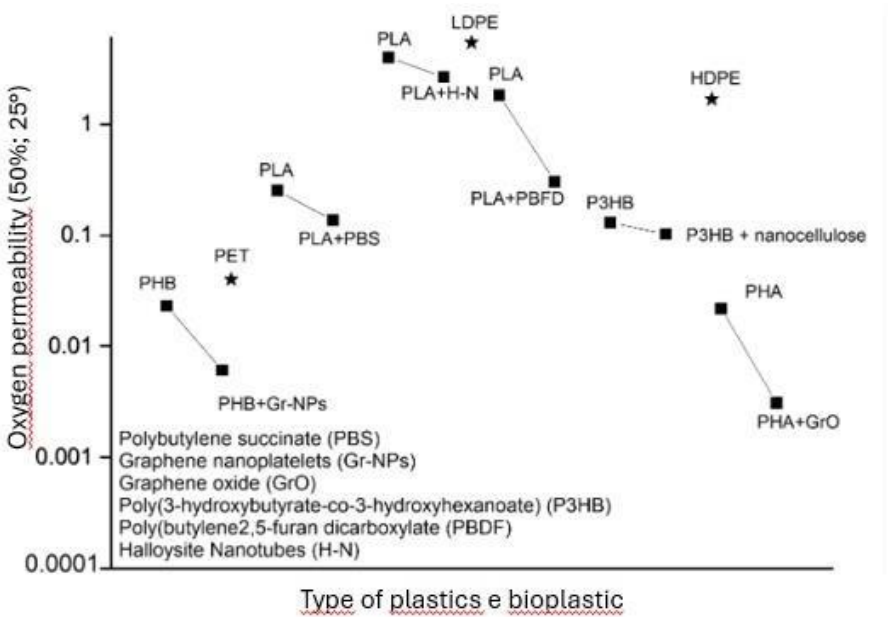


FIGURE 10 OXYGEN PERMEABILITY PROPERTIES BY TYPE OF POLYMER MATERIAL.[39]

As can be seen from the graph, biopolymers such as PHB, PHA, and PLA, when supplemented with substances exhibiting good barrier properties and good interaction with the functional groups of the biopolymer, can achieve high levels of gas barrier, specifically to oxygen. This property would allow them to find applications in numerous food packaging configurations, especially where long-term food preservation is a primary requirement.[39]

Most of the listed biopolymers have satisfactory gas barrier properties but exhibit aspects that need improvement through different approaches, such as functionalization of functional groups and/or composite production.[33]

Through chemical modifications, it is possible to enhance barrier capabilities by altering their hydrophilic and crystallinity aspects, features that will contribute to prolonging the shelf-life of fresh foods in addition to already optimal gas barrier properties.

These compounds exhibit a degree of hydrophilicity and crystallinity, which may cause issues during processing.[40]

Moreover, these polymers have poor performance, especially concerning the packaging of moist food products. Conversely, biopolymers serve as excellent gas barriers and are consequently utilized in the food packaging industry.[40]

Polysaccharides, such as starch, cellulose, chitosan, gums, and other polysaccharides, proteins, such as animal-extracted protein (casein, collagen, gelatin, etc.) and plant-derived protein (zein, gluten, soy, etc.), and lipids, including related triglycerides, are among the commonly available natural polymers derived from animal, marine, and agricultural sources.[40]

Most of these polymers are hydrophilic and crystalline, which might explain why there are numerous issues with wet food packaging. Nevertheless, despite these limitations, these polymers serve as robust barriers to gas transport.[8,37]

Biopolymers derived from entirely renewable organic substances are used to produce biodegradable packaging, which must decompose at the end of its lifecycle. Although biodegradable packaging can be primarily made from fossil-based substances, fossil-based chemicals, or a combination of renewable and fossil materials, the material must fully decompose and return to the natural world.

Based on specific criteria, biodegradable packaging material can be classified into various types. They can be divided into numerous categories based on the presence of essential elements. Inherently biodegradable polymers have a specific level of biodegradability.[36]

These are essentially artificial oil-derived polymers such as polyhydroxybutyrate (PHB), polycaprolactone (PCL), and polyvinyl alcohol (PVA).[11]

Biodegradable compounds are divided into three groups based on this classification. Polymers derived from natural materials, mainly plants, are known as natural polymers. Natural polymers include polysaccharides such as starch and glucose. Synthetic polymers are chemically produced from renewable polymers, such as polyacetate from lactic acid monomers.[31]

Polymers generated from microorganisms or genetically modified bacteria are known as microbial polymers. Polyhydroxyalkanoates are a famous example of microbial polymers in the energy substrate function.[39] Below is a depiction of the molecular structure of the biopolymers described earlier. For each polymer listed, a brief description of its key properties and characteristics relevant for packaging applications has been provided.[8]

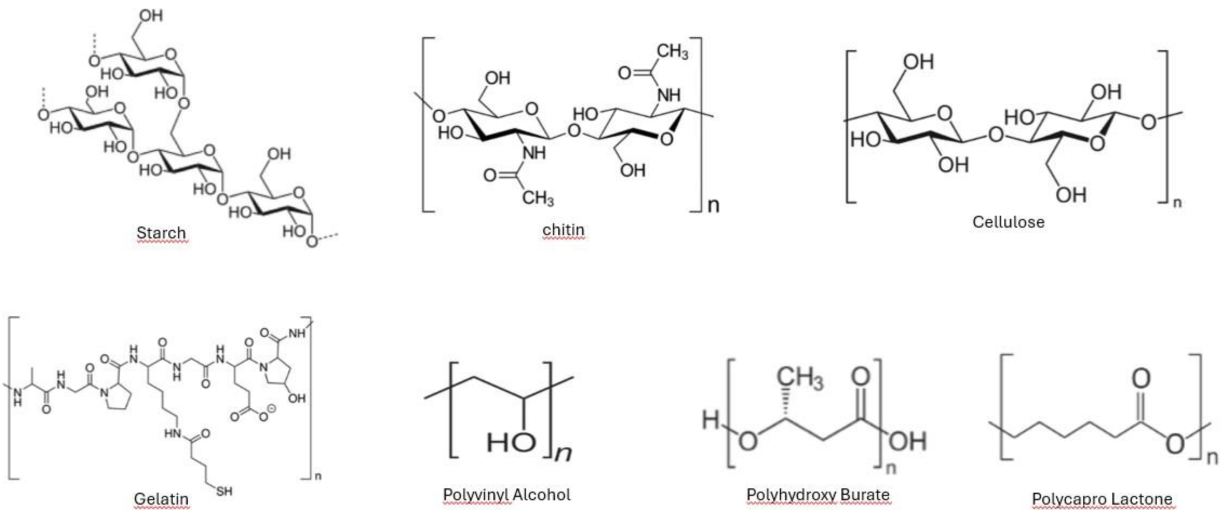


FIGURE 11 MOLECULAR STRUCTURES OF BIOPOLYMERS.[8]

Starch

Starch is known as one of the economically feasible and widely available groups of biodegradable polymers. It is considered an economical source of polysaccharides and is classified as a hydrocolloid

Generally, starch is used as a thermoplastic. Various varieties of starch, including rice, corn,cassava, potato, and tapioca, are employed for the synthesis of biodegradable polymers. Casting and extrusion are common methods for creating starch-based films. Polylactic acid (PLA), polycaprolactone(PCL), or polyethylene are commonly used to blend starch with less moisture-sensitive polymeric compounds (PE). Due to starch's high moisture sensitivity, blending is required to create a biobased product that is completely biodegradable in the environment. UV-irradiated starch films exhibitincreased moisture sensitivity.[13]

The physical and mechanical properties of UV-based starch films have also been altered. Plasticizers are used to minimize brittleness. Commonly used plasticizers include glycerol, sorbitol, and xylitol.

The physical and mechanical properties of starch films blended with glycerol have been evaluated as superior. Higher concentrations of xylitol and sorbitol have shown variations in film characteristics.[13]

Starch is a polysaccharide composed of two different polysaccharides: amylose (poly-alpha-1,4-D-glucopyranose) and amylopectin (70%-85% of the quantity in starch, poly-alpha-1,4-D-glucopyranose and alpha 1,6-D-glucopyranose units), distinguished by the linear structure of the former and the branched structure of the latter.[41]

Natural starch exists in the form of granules, which are the most common morphology found in nature and the most economical. However, it lacks industrial application due to its progressive decomposition in the presence of water. Moreover, because of the hydrophilic nature of its chains, its properties diminish in the presence of moisture, and it does not exhibit thermoplastic characteristics.[41]

To be used as a biopolymer for food packaging, starch needs to be blended with other polymers, such as adding PLA, to form polymer blends. This modification is achieved by incorporating crosslinkers that reduce water permeability and moisture absorption. The addition of PLA results in the formation of a homogeneous polymer blend, increasing not only the values of elongation at break and tensile strength but also the material's brilliance, thus enhancing its visual appeal to consumers.[13]

The addition of PLA does not alter the biodegradability of starch but improves its workability during processing. This biopolymer exhibits excellent biodegradable capabilities compared to synthetic polymers because it is easily and rapidly decomposed by various microorganisms present in the environment. The biodegradation of TPS (Thermoplastic Starch) occurs through the hydrolysis of acetate bonds facilitated by amylase, which breaks alpha 1,4 bonds, while glucosidases break alpha 1,6 bonds.[41]

Below, the mechanism of biodegradation of starch/PLA film is reported.[41]

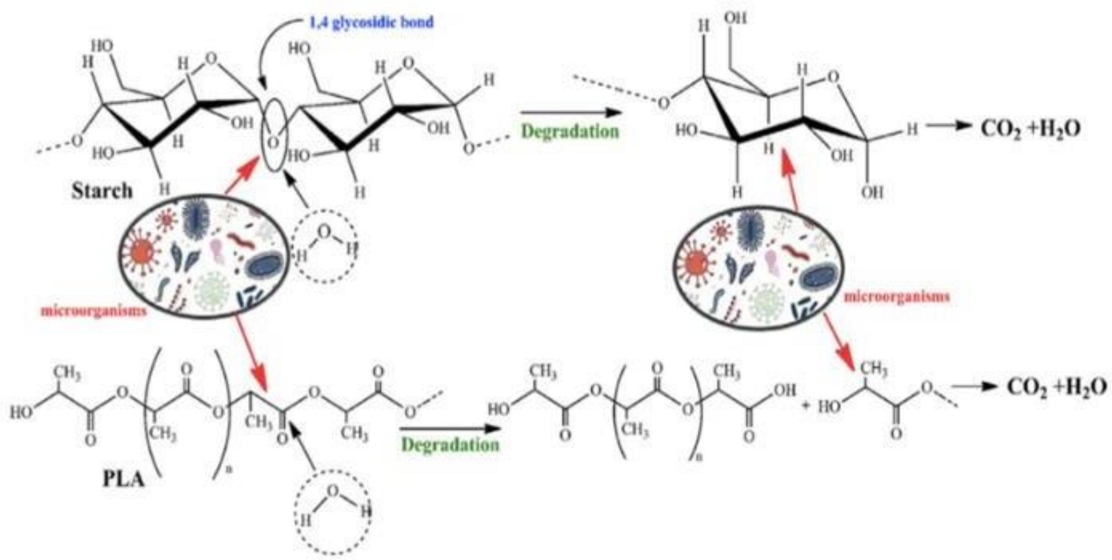


FIGURE 12 BIODEGRADABILITY ACTION IN RELATION TO HUMIDITY AND TEMPERATURE CONDITIONS.[41]

As can be observed from the representation above, under chemical and physical conditions such as temperature, pressure, and humidity, biodegradation proceeds at suitable rates aligned with specific requirements and results in desired end products such as CO_2 and water.[36,41]

Cellulose

Cellulose is a natural polymer composed of beta D-glucose. In the petrochemical industry, cellulose has been used as a necessary alternative. This organic material is abundantly available, degradable, and reusable. [42]

Fiber strength is generated through hydrogen interchain bonds, which prevent melting. The cellulose structure can absorb a large amount of water, but it cannot be dissolved in water.[14]

Due to the existence of hydroxyl structures and a regular structure that leads to the creation of crystalline microfibrils and fibers with strong hydrogen bonds, cellulose is commonly used in packaging as paper or cardboard.[43]

Cellulose its natural state might be difficult to employ as packaging material since it is hydrophilic and crystalline, with undesirable mechanical qualities. Consequently, it must be treated with chemicals such as NaOH, H_2SO_4 to produce cellophane with suitable mechanical properties for use as packaging material. Cellophane has long been used in commercial food packaging for products that require a long shelf life. Similarly, carboxymethyl cellulose, methylcellulose, ethylcellulose, cellulose acetate, hydroxyethylcellulose, and other cellulose derivatives are widely utilized.[44]

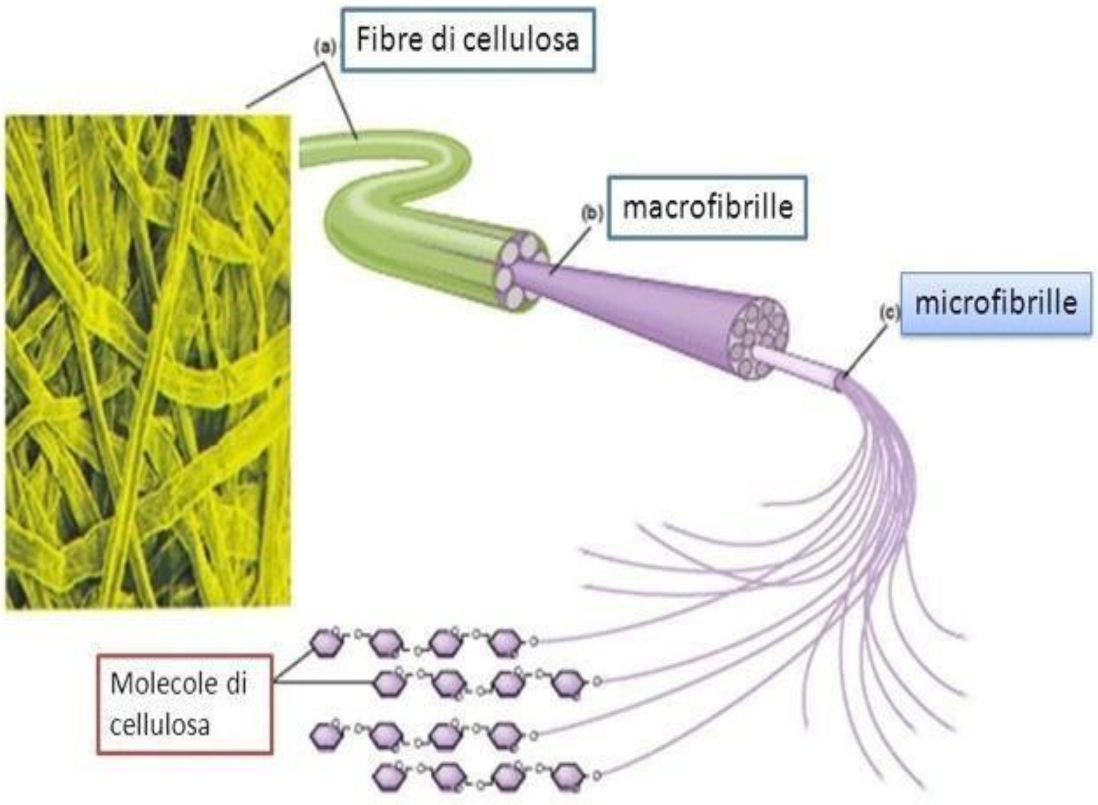


FIGURE 13 THE STRUCTURE OF CELLULOSE IN ITS INTERNAL PARTS.[44]

Cellulose is used to produce vast quantities of materials because its hydroxyl groups (-OH) can be esterified by organic or inorganic acids, thereby modifying the properties of the polysaccharide. Among its applications, it is used to create biopolymers for packaging, with applications including "filming" and addition to PVA and glycerol.[45]

Cellulose thus forms composite films with PVA and glycerol to improve its mechanical properties: glycerol is widely used in hydrocolloid film solutions, reducing their glossiness while increasing flexibility, whereas PVA is water-soluble, biodegradable, and non-toxic, allowing for a denser and more robust system by softening the film structure.[45]

In this case, the film production involves several stages. Firstly, cellulose is dissolved in a NaOH/Urea solution, followed by vigorous mixing for 1 hour. Subsequently, the cellulose is suspended and frozen at -20°C. To make the film transparent, glycerol (2.5% w/w) is added at room temperature and mixed to obtain a homogeneous solution, which is then processed for casting. The pH is then adjusted using acetic acid, and finally, the mixture is immersed in a PVA bath and poured into a container to conclude the process and obtain the final biopolymer.[45]

Chitin and chitosan

Chitin is the second most abundant natural biopolymer on the planet. [46]

Chitin is a major structural component of invertebrates' exoskeletons, insects' exoskeletons, yeast cell walls, and fungi cell walls. The physical attributes of chitin are white, rigid, and inelastic, consisting of nitrogenous polysaccharides.[46]

Chitin is composed of N-acetyl-2-amino-2-deoxy-D-glucose units that are bonded together in a linear polysaccharide (1 4) structure.[46]

The distinction between chitin and cellulose is that in chitin, the hydroxyl group of the C2 carbon is replaced with an acetamide group.

However, due to its insolubility in many solvents, the industrial applicability of chitin is limited.

The most important by-product of chitin is chitosan, whose fundamental advantage is its propensity for solubilization in diluted acidic aqueous solutions, thanks to the presence of free amino groups in its chemical structure.[9]

Chitin and chitosan are used to produce biodegradable films for packaging, marketed as edible coatings and membranes; they exhibit excellent antibacterial properties against fungi that may compromise the quality of the food contained therein. To improve the mechanical properties of the film, they can be mixed with other biopolymers such as alginate, collagen, and gelatin.

Chitosan exhibits stabilizing and emulsifying capacities, as well as antimicrobial properties against bacteria, fungi, and viruses, demonstrating biological activity against pathogenic microorganisms, protecting food from gram-negative and gram-positive bacteria.[46]

The antimicrobial activities of chitosan are linked to the presence of free chains of amino groups in glucosamine residues; the protonation of amino groups allows interaction with the microbe's cell membranes through their anionic behavior (presenting phospholipids and peptides on the surface); this bond increases the permeability of the bacterial membranes, thereby causing their destruction and death.[46]

Despite the important characteristics mentioned above, chitosan has few mechanical capabilities and poor barrier properties against water. These disadvantages can be mitigated through the production of composite films of chitosan and gelatin.

Composite films can be obtained with better mechanical properties compared to pure chitosan films, with higher torsional stress resistance values (≈43%) and a greater elongation percentage of about 40% compared to pure films.[40]

Moreover, gas barrier properties such as oxygen and vapor are improved due to the structural change of the film, which makes the passage of gases from one side of the film to the other more tortuous.

Gelatin

Gelatin is composed of 50.5% carbon, 6.8% hydrogen, 17% nitrogen, and 25.2% oxygen. [15] Chemical denaturation is used to create gelatin from the fibrous insoluble protein collagen. Gelatin is extracted from bones, skin, and connective tissues discarded during animal processing. Based on gelatin pretreatment, it can be classified into two categories.[15]

Acid-treated collagen produces.

Type A gelatin with an isotonic factor between six and nine, while alkali-treated collagen produces Type B gelatin with an isotonic factor of five.

Pig skin gelatin is referred to as Type A gelatin, while pork skin gelatin is referred to as Type B gelatin. [9]The fundamental characteristics of the source, animal age, and type of collagen have a significant impact on gelatin properties, according to Johnston-Banks. Besides the presence of lower molecular weight protein fragments, the physical properties of gelatin are influenced by amino acid composition, the relative material content of alpha, beta, or gamma chains, and better molecular weight aggregates. Due to their low cost and easy availability, they are employed for the synthesis of film packaging. Covalent bonds are interconnected in rigid molecules like collagen.

Gelatin is a good bio-based polymer due to its low melting point.[40]

The most essential physical features of gelatin are gel energy and viscosity. Gel energy, also known as the viscosity value, is a measure of gelatin's energy and stiffness, indicating the shared molecular weight of the constituents and is typically between thirty and three hundred viscosities (below 150 is considered low viscosity, 150–220 is considered medium viscosity, and 220–300 is termed excessive viscosity). With around 175 gel energy, higher Bloom values indicate more gelatin.[9]

Gelatin, whether powdered or granulated, is odorless and tasteless. It can be used as a thickening, gelling, foaming, emulsifying, and film-forming agent, among other things. Since gelatin is hygroscopic, it absorbs water depending on the relative humidity of the environment in which it is dried and stored. Gelatin films are used to extend the shelf life of some foods.[40]

Gelatin has a high oxygen retention capacity compared to other biodegradable polymers. Gelatin is widely used in the food, pharmaceutical, cosmetic, and photographic sectors due to its unique qualities. In the food industry, gelatin is used for gelling, stabilizing, texturizing, and emulsifying bread, dairy, beverages, and confectionery. Meanwhile, gelatin is used in the pharmaceutical industry to produce hard and smooth capsules, plasma expanders, ointments, wound dressings, and emulsions. Gelatin has been utilized as coating layers, emulsion layers, and non-curl layers on photographic materials in the photographic industry.[9]

Polymers processed by classical chemical fusion from biomass monomers.Polylactic acid (PLA)[8]

The building block for PLA is lactic acid, which may be obtained via the fermentation of maize or other agricultural resources. PLA is advantageous owing to its biocompatibility, processability, renewability, and minimal energy consumption. [31]

In contrast with biopolymers, including poly (hydroxy alkenoates) (PHAs) and poly (ethylene glycol) (PEG), PLA has good thermal processability. It could be made synthetic via diverse processing strategies inclusive of extrusion, film casting, and fiber spinning. PLA has been approved by the Food and Drug Administration (FDA) for use in packaging with direct contact with the food inside and has become a good option for the packaging of fresh foods or fast lifestyle goods, including overwrapping, lamination, and blister packaging, due to its biodegradable and biocompatible properties. PLA's characteristics are equally to those of polyolefins and polyethylene terephthalate (PET) therefore these compounds can be used in a wide range of applications.[47]

Thanks to its functional features, such as high transparency, good barrier properties, good salability, oil and grease resistance, and excellent organoleptic characteristics, there are potential options for food packaging.[31]

Using sustainable bio-based monomers, some polymers are created using traditional chemical processing procedures. Polylactic acid is a nice example of a chemically synthesized polymer. Polylactic acid is a biodegradable polyester composed of lactic acid monomers.[41]

PLA is biodegradable, eco-friendly, recyclable, and compostable, among other qualities. PLA is biocompatible since it has no harmful or carcinogenic properties. PLA can withstand a wide range of temperatures. PLA can be manufactured with less energy (between 25 and 55%).

PLA is made from maize starch, which is collected from plants like corn. During the growing period of maize, carbon dioxide from the environment is used for photosynthesis. As a result of the photosynthesis process, glucose is converted to starch, resulting in PLA chain formation.[31]

The fermentation of maize and agricultural biomass yields lactic acid, a building block for PLA.[47]

Polyvinyl alcohol[35,48]

Due to its enhanced film-forming capabilities, chemical resistance of artificial water-soluble polymers, superior biodegradability, and ease of production, polyvinyl alcohol (PVA) offers a wide range of benefits. [48]

Thanks to its hydroxyl groups, PVA has unique physical properties that promote hydrogen bonding. Polyvinyl alcohol can rapidly degrade biological polymers. In the last century, a wide range of products using PVA were manufactured in various industries.

Among the items were medications, food, surgical threads, resins, and varnishes. Therefore, it can be concluded that PVA can be used as a coating for specific food supplements and does not pose any safety issues.[35]

Polymers shaped directly by natural or genetically modified organisms[22,39,49]

Bacteria have the capability to produce a diverse array of biopolymers with varying biological activities and properties, which find applications across a wide range of industrial and medical fields.

Polysaccharides, polyesters, polyamides, and inorganic polyanhydrides (such as polyphosphates) represent the four main types of polymers synthesized by bacteria. Among these, polyhydroxyalkanoates (PHAs) stand out as intercellular sugars or lipids derived from plants, which are transformed into linear polyesters through microbial fermentation.[39]

While PHAs serve as promising materials, alternatives such as polystyrene (PS), polyethylene terephthalate (PET), low-density polyethylene (LDPE), and high-density polyethylene (HDPE) are also utilized.

Processing techniques for PHA films include injection molding, melt-extrusion techniques, and thermoforming. PHA-based composite films are notably utilized in the food packaging industry.

Thanks to their improved film barrier properties, thermal stability, and mechanical characteristics.

The utilization of microbial polysaccharides as packaging materials, such as xanthan, pullulan, and curdlan, has necessitated the use of biotechnological processes.[49]

Pullulan, a water-soluble, linear, exopolysaccharide, is produced by Aureo basidium pullulans from sugar-containing substrates. It finds application in various industries, including food, medicine, and cosmetics, for packaging. It is tasteless, odorless, non-toxic, and biodegradable. Pullulan-based films are typically transparent, homogeneous, heat-sealable, printable, flexible, and edible, with excellent oxygen barrier properties. However, they exhibit poor mechanical properties.

On the other hand, xanthan is produced through the aerobic fermentation of Xanthomonas campestris bacteria using sucrose or glucose as the primary carbon source.

Derived from a microbial source, xanthan is a heteropolymer composed of repeating units of glucose, mannose, and glucuronic acid in a 2:1:1 ratio, along with pyruvate and acetyl substituent groups. It exhibits viscosity, water solubility, and non-toxicity. Despite its advantageous properties, there is limited documentation regarding xanthan's application in the packaging industry, primarily due to its high production costs.[22]

Paper Mill and paper

Among the polymers mentioned earlier, cellulose is the one we'll focus on because it's the raw material used to produce paper, which in turn is crucial for producing and developing innovative and eco-sustainable food packaging.[44,50]

For the manufacture of paper, the commonly used material is cellulose pulp, obtained through chemical or mechanical treatments from plant material, usually wood.[51]

The plant and process where paper production occurs are made up of a series of elements such as:[51]

- Headbox: in this component, the mixture is distributed uniformly along the profile of the machine. The mixture flows from the headbox onto the moving wire through a lip, and to promote fiber alignment in the machine direction, the flow rate must be lower than that of the wire.[51]
- Wire section: in this area, the fibrous suspension, flowing on the wire, is hydrated to form a compact fiber mat. In this zone, over 95% of the water present in the mixture is removed using suction cylinders.[51]
- Size press: further dehydration of the paper occurs by applying pressure to the still wet paper web, passing it between steel cylinders coated with an absorbent filter.

- Drying section: in this section, the water content is further reduced using thermal energy.
- Final section: this section consists of a cooling cylinder that defines the properties of the smooth paper and the winder. The cooling cylinder cools the paper coming out of the drying section, preventing it from breaking.[51]
- The "calandred" smooths out the roughness of the sheet surface using smooth, tempered cast iron cylinders.[51]

  The machine ends with the winder, which rolls the continuous paper web onto a steel cylinder called a drum.[51]

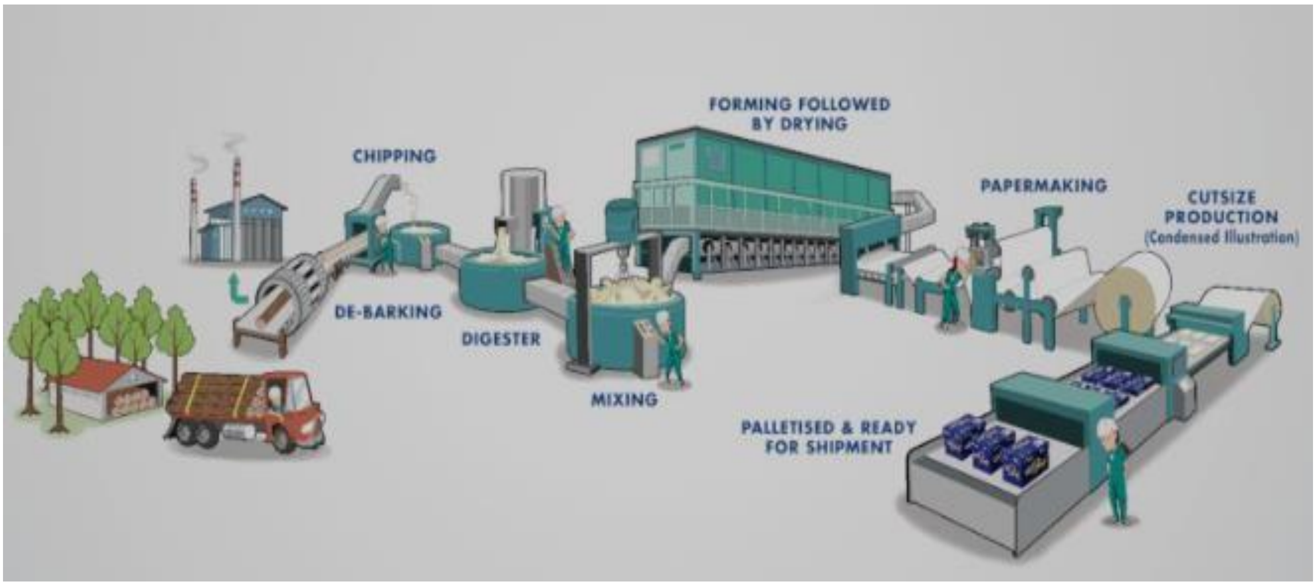


FIGURE 14 REPRESENTATION OF A PAPER MILL PRODUCTION PLANT.[44]

In the production flow of a paper mill outlined above, logs from the forest undergo initial processing through cutting and shredding to obtain "chipping," small wood fragments destined for further processing. The resulting wood chips are then sent to the digester, an imposing machine that, through heat and chemicals, breaks down the cellulose fibers within the wood to extract cellulose and obtain pulp, a fundamental ingredient for paper production.

Subsequently, the cellulose pulp, through specific conduits, reaches the continuous machines of the paper production plant, where it is distributed onto a conveyor belt uniformly. Here, the pulp is drained and transferred to the drying and calendaring section, where the cellulose sheet is dried and pressed, ultimately winding into rolls.[44]

The produced paper can undergo specific treatments, such as the addition of chemicals to make it resistant to moisture or to impart other properties, using coating technologies such as air knife coating or rotogravure. Quality control plays a crucial role in this process, with paper samples taken and analyzed in the laboratory to ensure high standards of quality and safety for food packaging.

Finally, the process concludes with the packaging and palletization section, where the paper is packaged and palletized according to customer specifications, ready for shipment.[44]

During my PhD activities, I had the opportunity to visit several paper mills plants with the aim of jointly developing new potential innovative projects toward the final realization of new products made with innovative and sustainable papers for their kind, in line with the theme of environmental impact, and meeting all the necessary requirements for food-grade packaging.

Below is a representation of images of the machine elements comprising a paper production plant. The images are taken from a facility that produces paper intended for food packaging, particularly for the bakery sector.

FIGURE 15 IMAGES OF THE MAIN COMPONENTS OF A CONTINUOUS PAPER MACHINE.

In general, a standard-sized plant produces approximately 75.000 tons of paper per year, represented over an average working cycle.

The pulper is the first constituent element of the paper production plant, consisting of a large vat into which 96% water and the remaining 4% consisting of cellulose fiber and additional substances such as mineral fillers, optical brighteners, binders, and others are inserted.

The cellulose used to produce paper for such applications is composed of a mixture of different fibers in terms of length and type, with 60% being long fibers from conifers and 40% being short fibers from hardwoods.

Refining systems allow the fiber to disperse optimally in water and consequently distribute correctly on the deposition wire to interact optimally with neighboring fibers.

The material that is accepted and deposited on the wire is dried through filtration due to the vacuum effect of the wire, and the fiber volume decreases from 4% to approximately 1% concentration.

The headbox, a component that regulates the amount of pulp to be deposited on the wire, ensures the correct gsm (grams per square meter) of the paper.

The dryer section's function is to evaporate all the water contained in the cellulose mat. It consists of a series of hot cylinders that dry the paper, and the extracted moisture is evaporated and reintroduced into circulation by adding it back to the pulper.

The subsequent phases are specific to the desired final paper product and may include calendared through the size press and/or machine press, treatments with coatings, barrier products, biopolymers using air blade systems, Mayer bars, rotogravure, etc.

The produced rolls are called jumbo rolls, from which useful rolls in strips and quantities are obtained.

Quality control of paper involves various procedures to ensure that the produced paper meets specific standards and requirements.[52]

This procedure includes a series of checks performed during production and are aimed at producing the material that conforms to the customer's requirements.

Some parameters are checked directly in line ex: grammage, optical homogeneity, thickness, and surface uniformity.[52]

Other more specific parameters that require laboratory instrumentation are carried out during production in order to be able to optimize the material during production and the checks in question are chemical physical checks such as: mechanical properties, porosity properties to air, moisture to water.[44]

Each property check adheres to specific ISO/standards, highlighted accordingly based on the evaluation parameter.[52]

- Basis weight measurement ISO 536 tolerance (+/-5%): verifying the weight or grammage of the paper to ensure consistency and compliance with specifications.
- Strength testing ISO 1924:3 tolerance (+/-5%): assessing the paper's resistance to tearing, tensile strength, and other mechanical properties to ensure durability and performance.
- Thickness measurement ISO 534 tolerance (+/-5%): checking the thickness of the paper to ensure uniformity and suitability for its intended application.
- Surface smoothness analysis Special method (angle of slip): evaluating the surface smoothness of the paper to ensure printability and aesthetic appeal.
- Moisture content testing ISO 287: measuring the moisture content of the paper to ensure it falls within the acceptable range for storage and printing processes.
- Optical properties evaluation CIELAB: assessing factors such as brightness, opacity, and color to meet the desired visual characteristics.
- Dimensional stability testing: checking the paper's dimensional stability to ensure it retains its shape and size under various conditions.
- Environmental and safety compliance: ensuring that the paper production process adheres to environmental regulations and safety standards.
- These quality control measures help maintain consistency, reliability, and performance in the manufactured paper products.

Here is a TDS on a grade of paper intended for food packaging applications, specifically for the bakery sector.

As can be observed, highlighted parameters such as roughness, water porosity, slipperiness, and tear resistance must adhere to precise values as they will enable the production of specific packaging products suitable for the intended applications of the finished product.

Property	Test method	Unit	Typical value
Grammage	ISO 536	Gsm	39-50
Bulking thickness	ISO 534	µm	45-61
Brightness	ISO 2470	%	78
Barrier	SCAN P26	nm/Pa's	2.5-1.8
Tearing resistance	ISO 1974	mN	210-290 250-350
Water absorptiveness	ISO 535	Gsm	29
Angle of Slip	Special method	° degree TS ° degree WS	11 11
Roughness	ISO 8791-2	ml /min TS ml /min WS	230-330 250-370
Moisture content	ISO 287	%	7

TABLE 4 REPRESENTATIVE TABLE OF A PAPER COMMONLY USED IN THE BAKERY SECTOR FOR TYPE AND GRAMMAGE.[53]

The values above represent the common requirements that a paper must meet to be used in such applications.

Slip angle values around 10 or lower indicate a significant surface slipperiness property of the paper, essential for easy separation between stacked baking forms.

Air barrier values of approximately 2-3 nm/Pas indicate excellent air resistance, ensuring stable shape retention of the finished product when exposed to the external environment.

Tear resistance values between 210 and 300 mN indicate that the paper adapts well to the hot forming process (thermoforming), withstanding the pressure of a thermoforming mold.

Grammage and paper thickness are considered fundamental starting parameters: the former is determined by industry standards, while the latter is crucial in the production process that shapes these papers through mechanical and thermal action.

Cellulose extraction process

Cellulose extraction can by subjecting primary sources to mechanical and/or chemical treatments to obtain two different cellulose pulps: mechanical pulp and chemical pulp.

Mechanical extraction involves the pretreatment of wood, as debarking is necessary (since the bark is unsuitable for processing due to its high lignin content), cutting into chips, and screening the chips based on size.[43]

Mechanical pulp contains a high lignin content; therefore, the resulting paper will have low resistance to aging and yellowing and will mainly be used to produce corrugated cardboard or newsprint.[53]

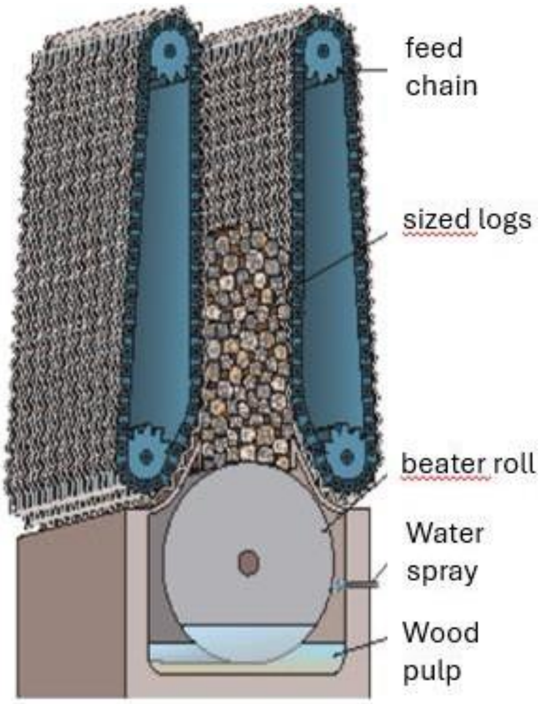


FIGURE 16 GRAPHIC REPRESENTATION OF THE MECHANICAL CELLULOSE EXTRACTION PROCESS.[44]

Above is represented the "wood pulp" process which is fundamental to extract cellulose fiber from tree logs consequently necessary to produce a wide range of cellulose-based materials such as paper and cardboard. It starts with the collection of logs, which are then transported to the paper mill to undergo a bleaching process, removing impurities and lignin. Subsequently, the logs are ground to create a fibrous mass known as pulp. This pulp, as seen in the bottom section of the image above, is then chemically or mechanically treated to separate cellulose fibers from other wood components. After thorough cleaning and washing, the pulp can undergo further treatments, such as bleaching, to improve its quality. Finally, the pulp is transformed into the desired product, such as paper or cardboard, through processes of forming, pressing, and drying.[44]

The chemical extraction process follows a different approach and process technology where the dominant factor is the reaction environment in which the raw material is subjected.

The chemical extraction process is instead divided into multiple stages and requires separation and refining of the extracted cellulose.[53]

The separation occurs by dissolving lignin in an acidic or alkaline aqueous phase to separate it from the cellulose, resulting in a solution of lignin and crude pulp, which still contains 3-10% lignin. The crude pulp is refined, yielding lignin again, and finally, refined pulp.[43]

The experimental conditions for separation vary depending on the type of chemical process applied to extract cellulose from wood. The main processes are sulfate (also known as the Kraft process), soda process, and finally, the sulfite process[43].

The pulp obtained from these processes is called high purity compared to the cellulose derived, which differs from wood pulp in that it contains a higher content of cellulose and better purity and quality.

The "soda process" was the first chemical pulping method applied on an industrial scale and was patented in 1845.[43]

Processing through this process is reserved for the source of wood material from conifers; these are known to contain a higher lignin content than hardwoods and react better with this type of wood.

Cellulose yields are about 45% by weight, and the mechanical characteristics of soda pulp over the years have been found to be less efficient compared to sulfite or Kraft processes.

The Kraft or sulfate process is currently the most popular one as it imparts the best mechanical properties to the cellulose[43]. The name of the process derives from the process chemical agent, which is sodium sulfate (Na_2SO_4), added after cooking the source to compensate for the losses of sodium and sulfur that occur during the process.[53]

The process involves combining wood chips with white liquors, which are an aqueous solution of sodium hydroxide and sodium sulfide. The liquor contained in the pulp after cooking is called black liquor and is separated from the fibrous mass by washing. After washing, the cellulose undergoes treatments such as purification, bleaching, and drying, while the chemicals are recovered and reused.[53]

The chemical extraction process of cellulose involves several steps, which may vary slightly depending on the specific method used, and generally include the following steps[43]:

- Pretreatment of lignocellulosic material: wood or other sources of cellulose are subjected to pretreatment to remove impurities and foreign materials and to make the cellulose more accessible to chemical reagents.
- Breaking down the lignin: lignin is broken down using chemicals such as sodium hydroxide (caustic soda) or chlorine dioxide. This step separates the cellulose from the lignin and other components of the lignocellulosic material.
- Cellulose extraction: after breaking down the lignin, cellulose is extracted through washing or filtration of the treated material. The extracted cellulose is then further purified to remove any residual lignin or other impurities.
- Bleaching: the extracted cellulose undergoes a bleaching process to remove any remaining impurities and to obtain a whiter and cleaner cellulose.
- Washing and neutralization: the bleached cellulose is washed and neutralized to remove any traces of chemicals used in the extraction and bleaching process.
- Drying: the cellulose is dried to remove residual moisture and obtain a finished product ready for use in paper, textile, or other materials applications.

The black liquor recovery is a fundamental part because it provides the energy needed for the pulp extraction plant and helps to limit the discharge of pollutants into the environment.

The reactions involved in the recovery process include combustion, where the black liquor is burned, to restore the original white liquor.[44]

The sulfite process generally uses an acidic solution and owes its name to the different bisulfites that form in the preparation of the cooking liquor, such as calcium bisulfite or magnesium hydrogen sulfite. It operates with aqueous sulfur dioxide and is more flexible in terms of process pH, allowing to produce various types of pulp suitable for a wide range of applications.[53]

The process phases are as follows:

1. pH 4 during the impregnation phase

2. pH 3 during cooking
3. pH 8 ÷10 at the end of cooking

Products obtained from the sulfite process are lighter and easier to bleach, but the resulting paper has lower mechanical strength.

Due to the lower fiber quality and higher energy consumption, Kraft pulp production is preferred industrially.[53]

Innovative methods of cellulose extraction - Organosolv Process

To extract and crucial issue that the world currently faces is sustainability, both in terms of primary resources and energy consumption. For this reason, research in recent years has focused on aspects of industrial processes that are less environmentally impactful through energy saving, reduction in process water consumption, and reduction in primary raw material sources.[54]

The Kraft process and the sulfite process have not yet been replaced on a large scale with more innovative process methods, as there are still too many limitations in terms of process efficiency and quality aspects of the final product. An emerging method that we delve into in this chapter concerns the extraction process using organic solvents as alternatives to recognize chemical additives, known as the "organosolv" process.[56]

In this process, the predominantly used source is lignocellulosic biomass. The cellulose extracted through the "organosolv" process can be used to produce cellulose pulp or cellulose pulp, both forms of materials intended for industrial applications in the food packaging sector because in both cases, all the requirements for compliance with the intended use would be met.[54]

The advantage of the "organosolv" process over a common chemical process is since the extraction component used to separate the different fractions constituting the starting material is an organic solvent that can be ethanol or methanol in different ratios to adjust the extraction yield of the constituent parts of the biomass of by-products, cellulose from hemicellulose and lignin.[55]

Another advantage of the organosolv process is its ability to better and more effectively valorized it represents an advanced method for extracting cellulose from lignocellulosic biomass, such as wood. This agricultural supply chains that possess high quantities of by-products. Below is a list of lignocellulosic materials and their composition in terms of fractions % of cellulose, hemicellulose, and lignin.[56]

Method involves the use of organic or aqueous solvents, such as methanol, ethanol, and ethylene glycol, which are mixed to treat the biomass. During the treatment, the solvents act on the lignin, a substance that provides rigidity to the plant, making it soluble and allowing the separation of cellulose, which constitutes the most desirable and usable part of the biomass.[57]

Once separated from the lignin, cellulose can be recovered with high efficiency. It is essential to completely remove the solvents to avoid inhibiting subsequent processes, such as enzymatic hydrolysis and fermentation. Preferred solvents are those with low boiling points and low molecular weights, as they are easier to remove after treatment.[57]

The organosolv process offers several significant advantages in terms of environmental sustainability. For example, it allows for efficient recovery of solvents through distillation, thus reducing material consumption and associated costs. Additionally, it enables the isolation of both lignin and carbohydrates, paving the way for greater valorization of biomass.

In some cases, this process can even outperform traditional cellulose extraction techniques, such as the Kraft process and the sulfite process, in terms of yield and overall performance.[54]

Paper packaging applied to the bakery sector

Food products, including those fatty and moist ones used in the pastry sector, have traditionally been packaged in paper packaging. However, the introduction of plastic has led to a gradual replacement of paper-based materials, which have lost significance in the food packaging market in favor of emerging plastic polymers.[58]

Recently, due to the negative environmental impacts of plastic, food packaging market is moving towards adopting more sustainable alternative materials. This has brought attention to the use of cellulose as a material for food packaging, with a focus on the latest research and developments in packaging materials made from renewable and easily recyclable raw materials.[59]

In the food packaging industry, the choice of material depends on the specific barrier properties needed to protect and preserve the food product. This choice is influenced by various factors such as thermo-sealing, processability, printability, strength, barrier properties (against water, oil, and gases), cost-effectiveness, environmental sustainability, and legal requirements.[58]

Currently, paper and cardboard represent approximately 31% of the global packaging market and are widely used for food packaging, ensuring prolonged preservation and consumer safety.

There are many ways in which paper and cardboard are used in food packaging to meet market demands involving major players in the food sector.[59]

However, paper alone is not sufficient to ensure the functional performance of food content preservation and safety, so it is functionalized according to the final application.

The main functionalization's include surface applications of polymer solutions and/or laminations of plastic films, always applied on the surface; thin aluminum films and solvent bath products used to impregnate the paper.[17]

In the food packaging industry, the choice of paper type is crucial and varies depending on its composition and mechanical treatment during production.

Greaseproof paper, often used in the bakery sector, is produced through a combination of chemical and mechanical processes, which synergistically provide the necessary characteristics to make it compliant and suitable for the required application. The chemical treatment occurs during the preparation of the cellulose pulp and involves the insertion of vegetable-based and/or synthetic products into the mass during the insertion of other common components such as mineral fillers, stearates, and pigments. The mechanical treatment is carried out by components of the paper mill plant and involves large steel cylinders known as "calandred," which act on the paper surface, leveling it and reducing the void spaces "bulk" between the cellulose fibers. The more aggressive the mechanical process, the higher the degree of surface gloss of the paper.[60]

Another type in the sector is "vegetable parchment," obtained through the treatment of continuous paper sheet in a bath of sulfuric acid, which serves to modify the structure of the cellulose fibers, making them more resistant and impermeable to external agents.[30]

These papers have the characteristic of biodegradability and disintegration under conditions of humidity and temperature such that they can be recognized as compostable papers according to European regulations.

For papers treated with wax-based products, the treatment involves the surface transfer of products such as beeswax and/or vegetable wax, which provide a thin surface layer on the paper. This coating provides a barrier against liquids and gases, making it ideal for packaging fatty and moist foods that are not subjected to hot applications (conventional oven/microwave).[17]

A category of papers resistant to moisture and with excellent release properties are treated with silicon-based polymers. The treatment creates a smooth and release surface on the paper, allowing cooked foods to easily detach and reducing the risk of adhesion. Additionally, the paper maintains its properties even at high temperatures and in the presence of fatty foods, ensuring safe and efficient packaging and preservation.[58]

Thanks to the protection of cellulose fibers provided by the silicon treatment, this paper can be easily recycled without losing its characteristic properties related to pulping in water, contributing to a more sustainable management of food packaging.[58]

In summary, both paper variants offer specific solutions for the needs of food packaging, ensuring safety and quality in food preservation and packaging.

The silicone system commonly used to coat paper destined for food packaging, especially for fatty products typically used in the pastry sector, generally consists of an emulsion of reactive silicone polymers combined with an organometallic catalyst. This system acts as a catalyst for silicone emulsions intended for paper coating, facilitating polyaddition-type reactions.[30]

The silicone emulsion used for this application is designed to provide excellent slip properties to the paper substrate. When applied, the silicone emulsion hardens through a polyaddition reaction in the presence of an organometallic compound, forming an elastomeric coating that fully complies with current food safety regulations in both Europe and the United States. This product complies with authorized substances in the following food safety regulations[61]:

- The German recommendation "Bundesinstitute für Risikobewertung, XXXVI and XXXVI/2 Papiere, Kartons und Pappen für den Lebensmittelkontakt (BfR)".[62]
- The Code of Federal Regulations of the United States, Food and Drugs (FDA), 21 CFR Ch. I, §176.170 and §170.180: paper and paperboard components and 175.300 Resinous and polymeric coatings.[63]
- EU Directive 1935/2004 on materials and articles intended to meet food.[64]
- All chemicals used within the EU are now subject to regulations EC 1907/2006 (REACH) and EC 1272/2008 (CLP).[64]
- Directive 10/2011 and its amendment EU 2016/1416 do not apply to this case, as it does not involve plastic materials, but rather paper with a silicone coating, which is not covered by Directive 10/2011, as are ion exchange resins and rubber.[65]

As can be seen from the image above, the silicone coating on this type of paper is applied using a size press or special blade coating equipment. This means that the continuous paper sheet passes through a pool of silicone emulsion located between two press rollers.

There is no gelification of fibers or induction treatments of silicone. This is a fundamental aspect, as cellulose fibers, without undergoing any structural changes but only protection, would be easily pulped and therefore easily recyclable as paper.[66]

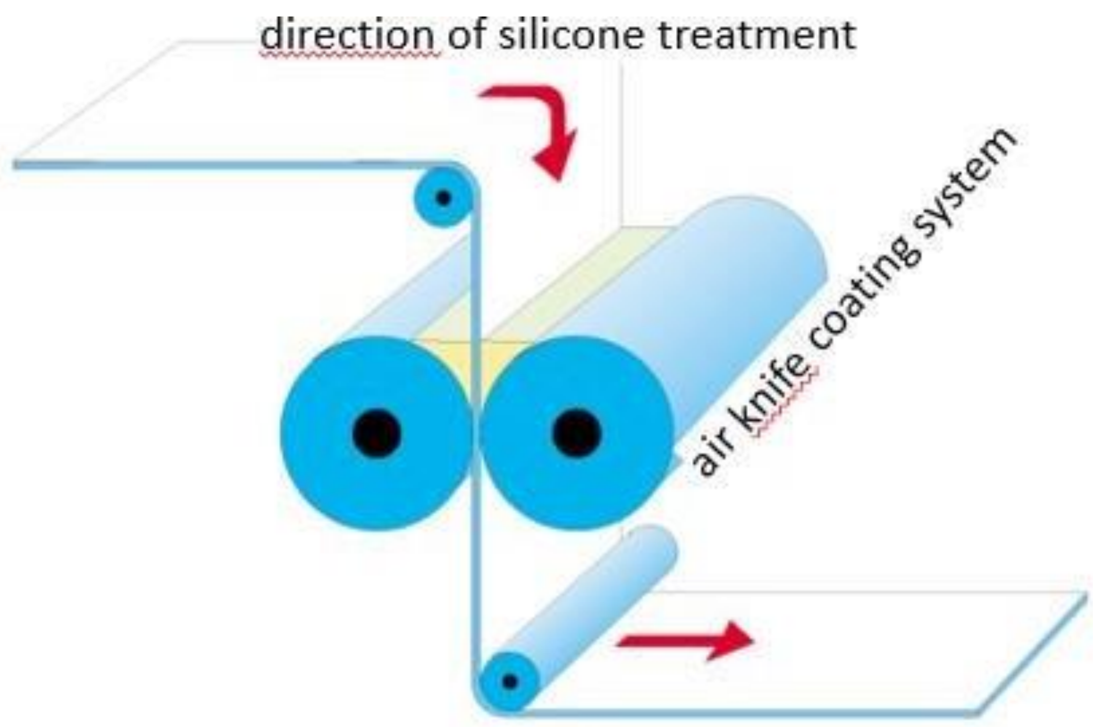


FIGURE 17 COMMON SURFACE TREATMENT PROCESS OF PAPER.[66]

Therefore, paper treatments for this specific application must be highly effective in providing the right physical and chemical protection barriers, allowing for the storage and transportation of the contents with high human health safety. Barrier products must therefore comply with all quality standards and regulations, ensuring that no substances or components migrate from the packaging to the food.[67]
Regarding compliance and safety in the specific application context, papers intended for such applications must meet suitable requirements and suitability for direct contact with various types of food: fatty, moist, and dry.[67]
A fundamental regulation that all papers destined for the bakery sector must comply with is the German Recommendation of the Bundesinstitute für Risikobewertung, known as BfR XXXVI and XXXVI/2 "Papiere, Kartons und Pappen für den Lebensmittelkontakt". BFR XXXVI/2 establishes compliance conditions for using paper in ovens at 200°C for 30 minutes.[62]

Other regulations required for paper food packaging used in bakeries include the United States Code of Federal Regulations for food and drugs (FDA), 21 CFR Ch. I, §176.170 and §170.180 for paper and cardboard components, and 175.170 for resinous and polymeric coatings that define specific requirements for paper and cardboard materials used in food packaging. These provisions establish the conditions under which paper and cardboard can be used in contact with food without compromising the safety or quality of the food itself. This includes requirements regarding chemical composition, purity, resistance to component migration, and other critical aspects to ensure that packaging materials do not contaminate food and are safe for human consumption.[63]

EU Directive 1935/2004 regulates materials and objects intended to meet food; chemicals used in the EU must comply with regulations CE 1907/2006 (REACH) and CE 1272/2008 (CLP). Paper packaging made up of plastic films applied through lamination processes, serving as functional physical barriers, are subject to EU regulation 10/2011 and its amendment UE 2016/1416, which establishes compliance of materials/plastic films for suitability in direct contact with food through migration analysis at different levels based on the final applications of the packaging material.[65]

In recent years, paper packaging for the bakery sector has played a fundamental role, particularly in terms of environmental sustainability.[24]

A significant increase in the global bakery products market is projected until 2027, with an estimated gain of $457 billion, consequently increasing the demand for paper packaging in this sector.[24]

In parallel to what has been mentioned above, paper used in packaging has been increasingly gaining market share in recent years because, by meeting safety and health criteria, it also offers significant advantages. There has been a growing importance placed on these aspects in recent years, as well as on the topic of environmental sustainability, which is significantly influencing the packaging market towards ecological solutions with minimal environmental impact according to environmental indicators that influence the final judgment in these terms. Food packaging, as it should be conceived when designed in paper, must be easy to dispose of in the paper recycling chain and/or in the organic waste disposal chain.[3]

To achieve this result, there must be a specific selection of materials and sustainable processes at the base.

In this context, my PhD project will focus on researching and experimenting with new packaging solutions through testing new materials, new production processes, and new analysis techniques to evaluate the environmental sustainability of the proposed solution.

Simultaneously, new materials and packaging products based on cellulose will be developed, incorporating treatments with various types of chemical products capable of conferring uniquefunctional characteristics to the final product in terms of both performance and environmental sustainability.[15]

The aspects mentioned above must consider a further crucial element to ensure adequate consumer safety: the freshness and quality of preserved foods. These characteristics must be ensured to guarantee continuous compliance with safety and consumer satisfaction. The factors that can influence food preservation are diverse in nature and characteristics, and for this reason, it is essential that all elements meet specific requirements based on the type of food and its required shelf life. To further explore these aspects, we have decided to present a representative image that highlights in more detail these factors and their impact on preserving the organoleptic characteristics and shelf life of the food.[17]

Figure 23 provides a detailed analysis of the main factors influencing the deterioration of bakery products. These factors are essential to understand how to ensure the freshness and quality of food throughout the entire process, from production to storage to consumption.

Firstly, the intrinsic properties of bakery products, such as nutritional composition, oxygen content, moisture, water activity (aw), pH, and ingredients used, play a fundamental role in determining their stability and quality over time. These factors directly influence the formation of microbiological and chemical contaminants during the production process.[23,30]

Processing conditions, including baking and cooling time and temperature, along with the hygiene of the processing environment, are equally crucial. Strict control of these conditions can help minimize the risk of contamination and ensure food safety.

Storage conditions, such as storage temperature, microbial content, relative humidity, and exposure to light, significantly impact the shelf life and freshness of food. Maintaining optimal storage conditions is essential to preserve quality and prolong the shelf life of products.

Lastly, packaging properties play a crucial role in maintaining food freshness. Effective packaging must provide adequate barriers against gases, UV rays, and thermal changes, along with possessing robust mechanical properties to protect food during transportation and storage. Adding antioxidant and antimicrobial activity to the packaging can further contribute to preserving food quality.

Careful consideration of all these factors is essential to ensure that bakery products maintain their freshness, quality, and safety for the end consumer.[59]

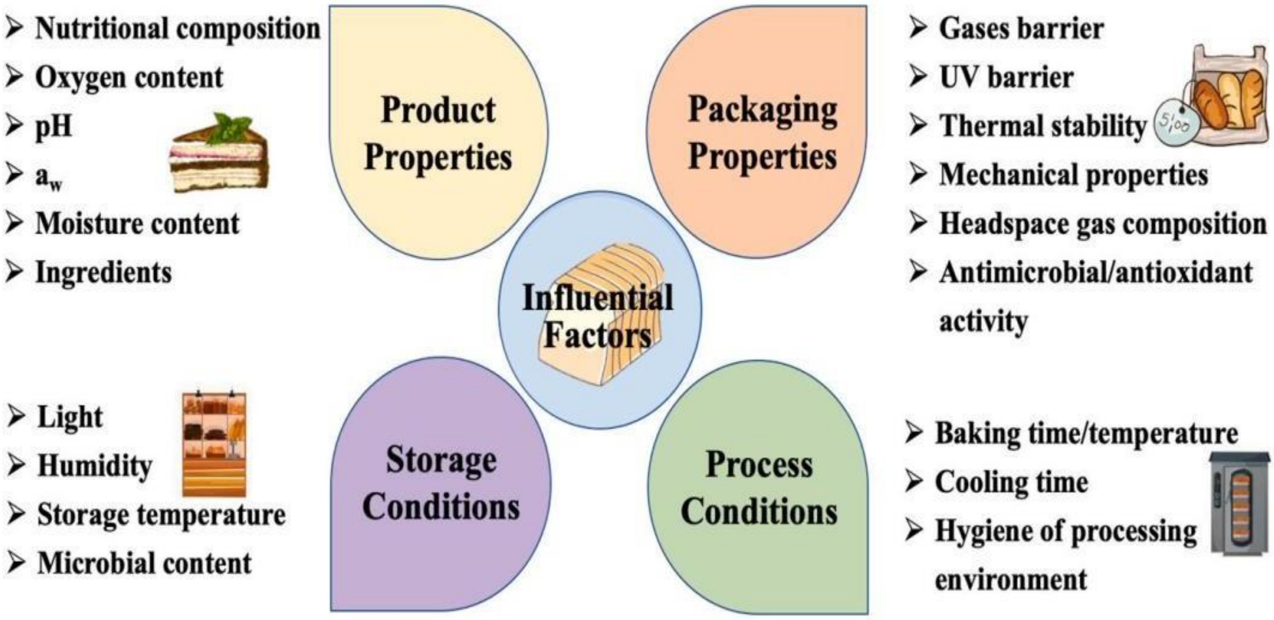


FIGURE 18 FACTORS AFFECTING BAKERY PRODUCTS SHELF-LIFE.[17,45]

In our study, we have also included a table below that analyzes the different types of packaging and their functions, along with their respective advantages and disadvantages. It is evident that in recent years, increasingly innovative coatings and films have emerged, both in terms of qualitative performance and environmental sustainability.

However, despite the multiple advantages of these new techniques and materials, most published research has focused primarily on the development and application of packaging for the preservation of fresh meat, fish, and dairy.

Regarding the preservation of bakery products, studies and alternatives used to prevent fungal deterioration and extend the shelf life of bread and other baked goods are still limited. This also includes a brief analysis of active packaging.[17]

Types of active packaging	Functions	Advantages	Disadvantages
Antimicrobial	Inhibition of microorganism growth	Reduce the use of chemical Antimicrobials added in the form of multilayer film can achieve a controlled release	Limited commercial application The use of certain antimicrobial agent Usage of pads has the risk of accidental ingestion
Antioxidant	Inhibition of unsuitable oxidation and aerobic microorganism growth	Reduce the addition of synthetic additives Show down the food metabolism	Promotion of anaerobic microorganism growth the accidental leakage of active components from a sachet deteriorates
Ethanol emitters	Inhibition of microorganism growth	Low-cost Ethanol vapor can be generated without spraying ethanol solutions directly onto products	Strong uncontrolled odor High volatility and uncontrolled release rate
Moisture absorbers	Removement of excess water	Reduce microbial growth and degradation of texture, flavor and color	Moisture absorbing sachets may change the sensory properties of the packaged food in some cases

TABLE 5 THE ADVANTAGES AND DISADVANTAGES OF ACTIVE PACKAGING IN BAKERY PRODUCTS.[17]

My PhD project research focuses precisely on studying various strategies and process technologies to improve the technical and applicative performance of packaging for bakery products.

Our research activities also aim to identify potential future trends in the bakery products packaging market, expanding the perspective, and considering possible uses of materials other than paper, but still focused on environmental sustainability and applicability in the sector.[60]

Functionalization through surface treatment of paper

For food packaging, different paper treatment methods are employed to confer the necessary functionalities for the final purpose.

Treated papers for food packaging are used to protect packaged foods from moisture, oxygen, and other external agents, while also ensuring food safety and extending their shelf life. These materials enhance the appearance of packaged products, facilitate printing, and promote sustainability using biodegradable or compostable coatings.

In summary, they offer protection, safety, convenience, and sustainability, making them an ideal choice for the food industry.[30]

One of the main strategies for improving the barrier properties of biodegradable food packaging, including cellulose-based materials, is surface coating. In principle, the application of an additional thin layer on one or both sides of bio-based films or other packaging materials (such as paper, paperboard) is defined as a coating process. There are various coating techniques, including chemical/physical vacuum deposition, solution coating (such as layer-by-layer assembly, slot-die coating, brush coating, spray coating, spin coating, and dipping), electrohydrodynamic processing (such as electrospraying, electrospinning), and other techniques like melt extrusion coating and hot pressing).[68]

Coating techniques are more versatile than lamination or coextrusion in terms of manipulating or developing multilayer structures with different thicknesses, ranging from nanometers to millimeters.

Below are the different techniques applicable for cellulose substrate barrier: they can involve a single layer or multiple layers.[68]

In the context of food packaging, electrohydrodynamic processing (A), including electrospinning, can be used to produce innovative materials with specific characteristics suitable for food preservation and protection. These materials can be employed to create membranes or coatings with barrier properties to shield food from moisture, gases, unwanted odors, or external contaminants. Additionally, electrospinning enables the production of thin surface treatments for cellulose substrates, which can enhance the mechanical strength and flexibility of food packaging without compromising its barrier properties.[30]

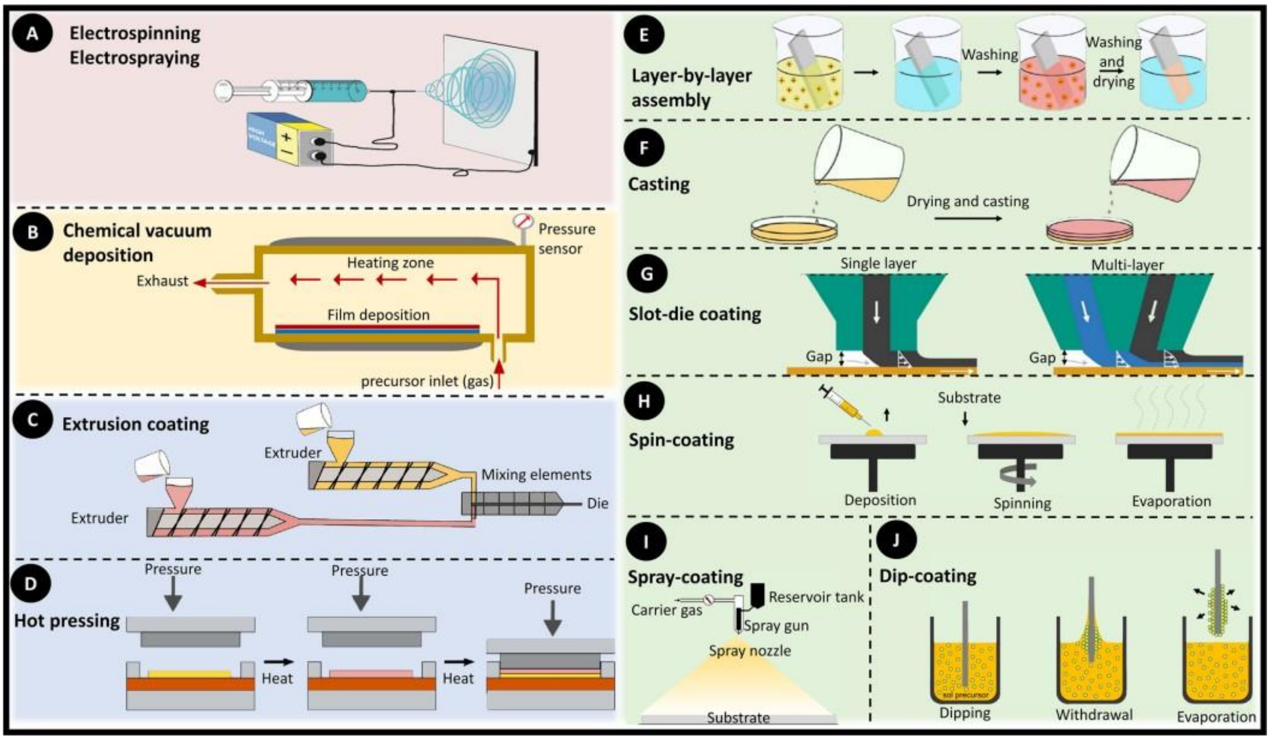


FIGURE 19 DIFFERENT COATING TECHNIQUES FOR THE PREPARATION OF SINGLE-LAYER OR MULTILAYER COATINGS[68]

Chemical vacuum deposition (B) for food packaging is a technique used to apply thin protective coatings to the surfaces of food packaging materials. This technology involves the use of a vacuum deposition process, which allows for the creation of a uniform and adherent layer on the packaging surface. During the process, the coating material is vaporized and deposited onto the packaging surface under controlled vacuum conditions.[68]

This technique offers several advantages in the realm of food packaging. Firstly, coatings applied through chemical vacuum deposition can provide an effective barrier against moisture, oxygen, and other gases, thus helping to preserve the freshness and quality of packaged foods. Additionally, these coatings can improve the mechanical strength of the packaging, protecting it from physical damage during transportation and handling.[65]

The downside is that this technique requires high application costs and specialized equipment to be effectively executed. Furthermore, it is important to ensure that the materials used in the coating are safe for food contact and do not transfer harmful substances to the food itself.

Nevertheless, chemical vacuum deposition remains a promising technology for enhancing the performance of food packaging, offering a combination of protection, strength, and safety for packaged foods.[69]

Extrusion (C) is a commonly used technique to apply coatings on substrates such as paper, cardboard, plastic, or other materials used in food packaging. During the extrusion process, the coating material is melted and pressed through an extrusion slot, forming a uniform layer on the substrate's surface.

In the specific context of extrusion coating for food packaging, the coating material can consist of thermoplastic polymers or other materials suitable for food contact. These materials are typically chosen for their barrier properties against moisture, oxygen, and other external agents that could compromise the freshness and quality of packaged foods.[69]

During the extrusion process, the coating material is melted and rendered fluid through heat and pressure applied by an extrusion screw. Once melted, the material is pushed through an extrusion slot onto a moving substrate, such as paper or cardboard. The pressure and extrusion speed are precisely controlled to ensure a uniform thickness of the coating on the substrate's surface.

After application, the coating is rapidly cooled and solidified to form a cohesive and adherent layer on the substrate. The result is food packaging with improved barrier properties and increased mechanical strength, which helps protect the food during transportation and storage.

Extrusion coating for food packaging is a highly efficient and versatile technique that allows for the application of uniform and adherent coatings on a variety of substrates, contributing to enhanced protection and quality of packaged foods.[69]

Hot pressing (D) for coating paper for food application is a process used to enhance paper substrates for food packaging purposes. In this method, a coating material is applied to the surfaces of the paper by applying heat and pressure.

The coating material is prepared, typically consisting of polymers or other substances safe for food use. This material can be in various forms, such as granules, powder, or a liquid solution. The paper substrate, often in the form of sheets or rolls, is then placed between two heated plates or rollers, along with the coating material.[69]

As the plates or rollers reach the desired temperature, they exert pressure on the combination of paper and coating material. This pressure causes the coating material to melt and distribute evenly over the surface of the paper. The heat facilitates the bonding process, ensuring proper adhesion of the coating to the paper substrate.

Once the pressing process is complete, the coated paper is cooled and solidified. This forms a protective layer that enhances the barrier properties of the paper for food packaging applications. The thickness and composition of the coating can be adjusted to meet specific packaging requirements, such as moisture resistance, oxygen barrier, or grease resistance.

This technique offers several advantages for food packaging, such as allowing for uniform distribution of the coating, ensuring consistent performance on the paper surface, and providing good adhesion between the coating and the paper substrate, resulting in durable packaging materials. However, this method may require specialized equipment and precise control of process parameters to achieve optimal results and comply with food safety regulations.

Layer-by-layer assembly (E) is a technique used in food packaging to create multi-layer coatings on substrates such as paper, plastic, or other materials. This technique involves the alternating deposition of thin layers of different materials to achieve specific desired properties in the final coating.[69]

During the layer-by-layer assembly process, the coating materials are applied sequentially onto a substrate, one layer at a time. These materials can be polymers, nanoparticles, or other substances suitable for food contact. Each layer is often only a few nanometers or micrometers thick.

The transfer of properties is closely linked to the selection of materials and their sequence. Layers of polymers with oxygen and moisture barrier properties can be alternated with antimicrobial layers to prevent bacterial growth, and adhesive layers to ensure good adhesion to the substrate.

Layer-by-layer assembly offers several advantages in food packaging because it allows for customization of the coating according to the specific protection and preservation needs of the product. Additionally, this technique can improve barrier properties, mechanical strength, and other characteristics of the packaging.[69]

Due to the complexity of the process, the costs of this technology are high, and it is adopted only if the packaging offers a high added value compared to market standards and for specific food packaging applications.

The casting process (F) for food packaging involves creating thin solid films by pouring a liquid or molten material onto a flat surface and evenly distributing it. These films, composed of materials safe for food contact, provide properties such as transparency, mechanical strength, and barrier capabilities against moisture and gases. The resulting films can be used to wrap food or form bags, ensuring flexibility and uniformity in the production of high-quality food packaging.

The slot die coating (G) is a technique used in the packaging industry to evenly apply liquid coatings on substrates made of cellulose, as well as materials like plastic or flexible films. During this process, the coating material is pumped through a thin slot called "slot die" and then uniformly distributed onto the surface of the moving substrate.[29]

This method offers several advantages in the food packaging sector, including the ability to achieve precise and uniform coating thickness, ensuring effective food protection. It also allows for precise control over the amount of coating material applied, enabling efficient resource management and waste reduction.

Due to its ability to work with a wide range of materials and thicknesses, slot die coating is suitable for producing a variety of food packaging, from thin films to thicker bags. This flexibility makes it a popular choice in the food industry for large-scale coating applications.

Spin coating (H) is based on the principle where liquid coating material is poured onto the center of a rotating substrate, which is spun at high speed.

Due to the centrifugal force generated by the rotation, the liquid quickly spreads over the surface of the cellulose substrate, forming a thin and uniform layer. It's possible to achieve very thin coatings with controlled thickness, which is important to ensure that foods are protected without adding excessive bulk or weight to the packaging. It's highly efficient and can be used to produce large volumes of packaging in relatively short times but for small packaging configurations. Additionally, the rotation speed and other process parameters must be carefully tuned to ensure uniform coating thickness and avoid material waste.[15]

Spray coating (H) and dip coating (I) are both techniques used for such applications. In the former, the liquid coating material is sprayed onto the substrate surface using a specialized spray gun, allowing for the uniform distribution of the coating over the substrate surface quickly and efficiently. It can be considered a versatile technique that enables easy coverage of large areas and is suitable for applications on substrates of various shapes and sizes.

Dip coating, on the other hand, involves immersing the substrate directly into a tank containing the liquid coating material. The substrate is dipped and then slowly lifted from the tank, allowing the coating to adhere uniformly to the surface. This technique is particularly effective for coating three-dimensional objects or achieving uniform coatings on complex surfaces.[68]

Both techniques allow for the attainment of uniform and adherent coatings that provide protection against external agents and improve the shelf life and quality of packaged foods, and the choice between the two techniques will depend on the specific requirements of the application and the characteristics of the substrate to be coated.[68]

The application technologies for barrier and protective coatings on cellulose substrates, as we have seen, are numerous. Therefore, the choice in selecting the specific technology to use depends on several factors, including the characteristics of the substrate in terms of chemical and physical properties and its degree of purity, which determine the selection of the product to be applied to achieve better functional results in terms of properties and performance. Additionally, a determining factor in selecting the substrate treatment technology involves the type of product due to its chemicalnature and the chemical-physical and rheological properties of the barrier product.

Thesis Objective

The eco-design of packaging products follows a sustainable design approach, incorporating environmental criteria to minimize environmental impact throughout the entire process, from raw materials to end-of-life disposal.

Alongside the focus on packaging, ensuring its safety is paramount to guarantee its proper use without compromising human health and safety. Packaging developed for this category falls under MOCA (Materials and Objects in Contact with Food), necessitating compliance with safety standards and good manufacturing practices to meet national, European, and international regulations for market entry.[32]
Collaborative efforts with various companies and the Chemistry Department - MOF - have led to the creation of several eco-friendly and innovative alternatives. Material and product developments have utilized paper as a base material with innovative barrier treatments, resulting in eco-friendly packaging for specific applications, targeted towards the food packaging market for large confectionery industries rather than for retail sale by paper packaging distributors.

The acquired know-how has been overseen by Ecopack spa, ensuring the verification of technical and research activities to test the properties of the new material, initially through laboratory-scale testing followed by prototyping phases.[60]

Attention has been paid to the exploration of new environmentally compatible materials, such as biobased polymers like starches, microfibrillated cellulose, and others. Simultaneously, academic research has been conducted to identify new energy solutions, utilizing innovative polymer coatings for the functionalization of both coated and untreated papers.[6,40]

With the direct support of Professor Claudia Barolo and Dr. Matteo Bonomo, two significant academic works have been finalized, leading to their official publication.

To finalize and submit the two articles, the contributions of the following individuals and research institutions listed below were fundamental:

The first article titled "Engineered surface for high performance electrodes on paper" published in the journal Applied Surface Science, Volume 608, 15 January 2023, 155117, proposes a low environmental impact and scalable method for producing PEDOT: PSS electrodes on standard copy paper through surface modification, aiming to offer more sustainable solutions for flexible electronics devices in terms of material usage and fabrication methods.

In particular, the paper substrate is treated (via blade-coating technique) with a cellulose-based polymeric coating to close its porosity and homogenize its surface before the deposition of the conductive material. This cellulose-based interface allows for the subsequent effective deposition of the PEDOT: PSS conductive layer, resulting in an improved electrode in terms of both conductive stability and electromechanical performance. The stability of the electrode was monitored over a six-month period, and the electrodes did not experience any aging effects, showing stable resistance values (within experimental error). The electrodes fabricated on modified paper exhibited lower electrical resistance (-80%) and an increased breaking point during strain tests (17 ± 1% vs 9 ± 1%) with a slight increase in resistance after 1000 bending cycles (4% vs 9%). Therefore, their enhanced performance, stability, and reproducibility open new possibilities for wearable electronic devices.

The second article titled "Enhancing Packaging Sustainability: Cellulose-Based Coatings with Improved Barrier Properties and Oxidative Stability" presents a comprehensive research study on the use of cellulose derivatives-based coatings to enhance the functionality of paper as a packaging material, addressing the demand for environmentally sustainable packaging solutions. Derived from renewable sources, cellulose derivatives emerge as a promising avenue for improving the mechanical strength and barrier properties of paper.[70]

Looking towards innovation, we introduced an inventive approach by modifying the paper surface coating with detonation nanodiamonds (DND). The working approach involved a complete characterization of the modified cellulose-based coatings, providing insights into their structural and surface properties, and determining how those properties are altered with the addition of small quantities of DND to the system.[70]

This integration allowed for the identification of unique properties, including enhanced mechanical strength, barrier capabilities, and thermal stability. Significantly, we achieved a 70% increase in water vapor diffusion resistance on the paper, a sixfold improvement in mechanical resistance, and increased chemical stability in strongly oxidizing and acidic environments.

The results underscore the vast potential of cellulose-based coatings, particularly when customized with DND, offering sustainable solutions with superior performance characteristics. This research addressed the topic of eco-friendly packaging materials development and concurrently contributed to advancing the potential efficacy of cellulose derivatives applicable in the packaging industry.[70]

During my doctoral studies within Ecopack, I had the opportunity to conduct assessments of the environmental impact of packaging products using accredited software to carefully evaluate the life cycle (LCA) of these products. This allowed us to design and implement targeted actions to improve the environmental sustainability of the company's packaging. Through these analyses, we were able to achieve satisfactory results regarding the sustainability profile of paper packaging, identifying potential areas for improvement in the design, development, and production processes.

The use of life cycle assessment (LCA) as a key methodology also allowed us to evaluate the most relevant environmental indicators, such as carbon footprint, water consumption, and energy usage throughout the entire product life cycle. The results obtained provided a solid foundation for making more informed decisions regarding the design and development of more sustainable food packaging.

This work promoted greater environmental awareness and responsibility within Ecopack, paving the way for new practices and strategies aimed at reducing the environmental impact of packaging products.

Analyses of the environmental sustainability of the company's paper packaging have been conducted to assess the sustainability profile and adopt improvement actions during the redesign or enhancement of the packaging. The LCA analysis conducted focused on key environmental indicators such as carbon footprint, water consumption, and energy usage in the various stages of the process from raw materials to the end-of-life of the analyzed products.

In the next section, all project activities related to cellulose-based food packaging will be presented, providing a chronological overview of the various initiatives undertaken within the Ecopack company, where I had the opportunity to conduct my doctoral research. The various projects tackled will be described using diversified approaches, including the selection of innovative and sustainable raw materials, the adoption of more efficient and innovative production processes, and a range of other relevant factors and indicators. The aim is to propose new environmentally sustainable packaging products that adhere to the principles of the circular economy and contribute to achieving the Sustainable Development Goals (SDGs), the 17 interconnected objectives defined by the United Nations to create a better and more sustainable future for all.

Introduction to Thesis Projects

In the current context, characterized by a growing focus on environmental sustainability, the food packaging sector faces the challenge of balancing quality and safety with sustainability. The research and development of new renewable and/or sustainable raw materials, such as biopolymers and materials derived from agro-industrial products and food by-products, represent a significant response to this challenge. The aim is to create innovative, eco-friendly, and environmentally compatible packaging solutions. The adoption of sustainable raw materials and low-impact production processes is essential for reducing ecological impact and promoting a more sustainable future for the industry. During my doctoral studies, the focus of my research and development work was on creating packaging solutions that reduce environmental impact and enhance the sustainability of food packaging products. The applied research and development activities, aimed at developing new prototypes and subsequently new packaging products, were directed towards various development lines within the food packaging sector. These lines share the use of innovative materials and circular processes characterized by a reduced environmental impact, such as lower energy consumption, which helps to decrease greenhouse gas emissions. These practices address scopes 1, 2, and 3 of greenhouse gas emissions, providing a detailed view of environmental impact and contributing to the improvement of a company's ecological footprint. Integrating these practices is essential for achieving long-term sustainability goals. A central project aimed at promoting innovation and sustainability within the company involved the development of a new tray designed for the retail and gastronomy sectors. This tray, made from cellulosic material, meets the functional criteria of MAP (Modified Atmosphere Packaging) trays, while significantly improving environmental sustainability compared to traditional plastic, aluminum, or multilayer material solutions, which are considered less environmentally friendly. Another important area of research I explored was the life cycle assessment (LCA) of a standard product category of the company, namely cake molds. In this case, the focus was on analyzing the level of sustainability in terms of CO_2 emissions, following a "cradle to gate" approach and extending the analysis through two different end-of-life recycling pathways: product recycling or industrial composting. The goal was to evaluate the best strategy to fully value this product category, from the raw materials used to the optimal disposal method. In this context, it was crucial to demonstrate how eco-friendly design choices can significantly reduce environmental impact.

In parallel, to respond to the SUP (Single-Use Plastic) directive, an initiative from the European Union aimed at significantly reducing plastic packaging waste, a new paper stick for chupa chups/lollipops was designed and developed. This innovation is characterized using raw materials that provide the finished product with high water resistance and structural rigidity, offering a sustainable alternative to plastic sticks. The creation of a new line of bakery products, made from an innovative and 100% eco-friendly paper, represented a significant advancement in new product development. This highlighted the company's innovation and provided substantial benefits in terms of biodegradability and compostability, fully respecting the environment. Other projects, such as the search for new raw materials and processes, allowed for the development of numerous prototypes that significantly contributed to achieving the results, such as the activities in projects 2 and 3.

The work carried out over these three years has laid a solid foundation for addressing future needs or environmental sustainability issues related to food packaging. Through innovative solutions, it is possible not only to reduce environmental impact but also to improve the functionality and quality of new packaging. These projects represent a significant step towards adopting circular economy practices in the packaging sector, promoting the use of renewable, biodegradable, recyclable, and compostable materials that comply with the new packaging materials and waste regulation, which will replace the Packaging Directive 94/62/EC, with the aim of contributing to the transition towards a circular economy.

Research and development projects.

In recent years, growing concerns about the limited resources of fossil fuels and the environmental impact resulting from the use of non-biodegradable plastic packaging materials have driven research towards the development of biopolymers derived from agro-industrial products and/or food waste. This scenario has sparked significant interest in the food packaging sector, where consumer demand for safe, high-quality, and long-lasting food products is paralleled by a heightened ecological and environmental sensitivity regarding packaging waste.[15]

To address this challenge, our company project will focus on developing new eco-friendly packaging solutions with lower environmental impact. The activities will involve experimenting with barrier coating formulations to be applied to cellulose using different application methods and technologies. The goal is always to develop functional and eco-friendly packaging solutions to offer to confectionery and food manufacturers.

With the increasing interest in a circular economy and a deep concern for the environment, the development of sustainable packaging is a major focus of today's industry, with paper emerging as a key material to rely on. However, the main issue with paper lies in its high porosity and hydrophilicity, which make it less effective in food applications where barrier properties are crucial.

Over the years, effective packaging alternatives have been developed to address these shortcomings, including physical treatments to close pores and compact cellulose fibers, or chemical processes to create innovative new materials.

While the former may have limitations in water and vapor barrier properties, the latter faces environmental constraints, as composite paper-polymer materials represent a significant portion of non-recyclable waste. Consistent with the goal of eliminating non-recyclable multilateral products, the aim of today's industriesis to find natural polymers to use in designing and producing new functional and sustainable food packaging with good water and grease barrier properties.

Packaging materials based on innovative raw materials and innovative process technologies offer advantages in terms of biodegradability, compostability, and recycling compared to standard materials such as petroleum-derived synthetic polymers.[68]

This project aims to develop new materials and food packaging solutions to propose as sustainable alternatives to Ecopack SpA, either as a new packaging solution or a more competitive alternative due to its sustainability and cost-effectiveness. The activities undertaken during these doctoral years have led to the development of packaging prototypes and/or innovative products aimed at launching new products on the global market as sustainable and competitive offerings.

Often, activities were carried out in collaboration with partners, as these were complex projects involving eco-design of packaging products for the confectionery sector and large-scale distribution. In the first year, the focus was mainly on the development of a new food tray for gastronomy that met the functional criteria of a MAP tray while improving its environmental sustainability. Research and development activities were carried out on new polymer bases to be applied to cellulose to confer better barrier properties.

In the second year, attention was directed towards life cycle assessments (LCA) of the company's packaging products to identify current limitations in terms of environmental sustainability and to take improvement actions on phases that were not yet optimal in terms of sustainability criteria. In the third year, the focus shifted to the processes and products that could be developed to introducenew, more sustainable, and innovative packaging products to the market, both in terms of functional properties and raw material processing criteria.

The project activities were carried out through collaboration with industry partners, progressing towards achieving results, selecting, and scaling activities, and producing prototype products and subsequent samples in sufficient quantities to present them to stakeholders interested in proposing these innovative solutions. The commitment expressed in these three years of project activities has provided me with a fundamental foundation, allowing me to delve into the details of environmental sustainability issues related to packaging products and their industrial processes

1. **Paper Pan Project development of a new cellulose**

trayPremises

During the first year of my doctoral studies, I was involved in continuing the research and development activities within the research project titled "Ecofood," funded by the Piedmont Region in collaboration with the University of Turin, Department of Chemistry, and the MOF-Functional Organic Materials research group.

The project aimed to develop a new cellulose food-grade tray with high barrier properties to liquids and gases through the use of innovative sustainable raw materials compatible with Ecopack's production processes, in order to propose a new high-performance packaging product to the GDO market with easy end-of-life management.

Therefore, building upon the results of the project, we continued our activities by extending research not only on barrier products but also on process technologies and technological treatment of the raw materials beyond cellulose.

The Grande Distribuzione Organizzata (GDO) represents a commercial sector focused on the retail sale of a wide range of products, both food and non-food, through a network of supermarkets, hypermarkets, and other retail outlets often managed by distribution chains or large companies. The GDO has a significant impact on the economy, influencing consumption patterns and market dynamics due to its extensive reach and widespread presence.

The company's decision was to continue the project internally, building on the results achieved with the intention of improving them to achieve the final outcome. During the research and development period, a new type of container was designed and realized, significantly different from models used in the past.

This tray features a flat and weldable upper rim, making it particularly suitable for a variety of applications in the food sector. Despite significant progress in realizing this tray, a critical issue emerged regarding the barrier properties of the material used.

Although the tray demonstrated good grease resistance, there was a lack of effectiveness in protecting against water, moisture, and oxygen once the material was treated with an improved performance coating.

This limitation contradicted the specific product requirements and highlighted the need for further improvements. Despite the development and testing of barrier products, it was not possible to achieve the desired barrier properties in line with the product's application requirements.

Consequently, the project was extended and continued in the following year of the PhD, in an attempt to address this significant challenge and ensure that the container fully meets the preservation and protection requirements of the food product.

Objective

Particular attention has been devoted to the research of new raw materials such as paper, inks, and adhesives, which are essential components for designing and producing innovative and sustainable packaging. The issue of environmental sustainability of packaging has become increasingly relevant, and it is crucial to promote its growth to reduce the negative impact generated by packaging in recent years.

This first research and development project conducted for the company considered the fundamental element of creating a new packaging product with the characteristics of being recyclable, compostable, and having functional gas barrier properties. The activities conducted also aimed to maximize the productive potential of the wet cellulose pulp process technology, while ensuring sustainability aspects related to raw materials and compostable of barrier products. The wet cellulose pulp processtechnology, which utilizes cellulose as a raw material, would allow the production of products with uniform and complex geometries and shapes to achieve all the required characteristics and performanceon the tray, including excellent water resistance in line with the properties and predetermined targetsto recognize it as a tray for the preservation of food in Modified Atmosphere Packaging (MAP) conditionsto allow prolonged preservation of fatty and humid foods.

Market analysis and mapping

As a preliminary activity, market analysis was conducted to identify current demand and develop a solution that is as closely aligned and suitable to sustainability goals as possible.

The channels of the large-scale retail Trade (GDO), retail sales, and major food industries play a crucial role in the paper-based food packaging sector. They are the main distributors and producers of food, thus determining the demand for packaging. Furthermore, they influence consumer purchasing choices, being able to promote the adoption of sustainable packaging through responsible purchasing policies. Through meetings with commercial agents, it emerged that the GDO channel is the most interested and sensitive to the issue of seeking sustainable solutions to meet the Sustainable Development Goals (SDGs) established by the United Nations and to encourage concrete sustainability actions, including reducing environmental impact, to promote market strategies advantageous for reputation, consumer trust, and long-term sustainability.[71] Through meetings with commercial agents, it has emerged that the channel of Organized Retail (GDO) is the most interested and sensitive to the search for sustainable solutions to meet the Sustainable Development Goals (SDGs) established by the United Nations and to promote concrete sustainability actions, including reducing environmental impact, to foster market strategies advantageous for reputation, consumer trust, and long-term sustainability.

The Onu's 2023 agenda for sustainable development, signed on September 25, 2015, by 193 United Nations countries, including Italy, defines the 17 sustainable development goals to be achieved by 2023, articulated in 169 targets, which represent a compass for putting Italy and the world on a sustainable path. The process of changing the development model is monitored through Goals, Targets, and over 240 indicators. In this regard, each country is periodically evaluated at the UN by national and international public opinion.

The 2030 agenda brings with it a novelty regarding the idea of sustainability, which is not solely an environmental issue but an integrated vision of the various dimensions of development. The 2030 agenda is based on five key concepts:

1. People: Eliminate hunger and poverty in all forms, ensuring dignity and equality.
2. Prosperity: Ensure prosperous and fulfilling lives in harmony with nature.

3. Peace: Promote peaceful, just, and inclusive societies.

4. Partnership: Implement the agenda through strong partnerships.

5. Planet: Protect the planet's natural resources and climate for future generations.

Of the 17 goals, the focus on project activities mainly concerns goals 12 and 13.

Goal 12 concerns responsible consumption and production to ensure sustainable models of production and consumption, which are involved in our doctoral research project activities. In 2022, humanity had already consumed all the resources that the Earth could regenerate in a year. For this reason, activities related to environmental sustainability are of great importance, which through new packaging solutions should offer a contribution to reducing the depletion of non-renewable raw materials by replacing them with renewable ones that are infinitely regenerable.

Goal 13, combating climate change, involves taking urgent measures to combat climate change and its consequences related to the increasing global average concentration of CO_2 in the atmosphere, which had already reached record levels by 2022, amounting to 415 ppm. In our case, the contribution will be to improve product eco-design by proposing new packaging solutions with lower environmental impact through renewable and easily recyclable packaging solutions following the correct recycling system.

The GDO is moving towards the use of trays for ready-to-eat foods displayed in refrigerators, and it is therefore essential to ensure consumers a shelf life like that of products served at the delicatessen tray, packaged as MAP (Modified Atmosphere Packaging). Throughout the project, various activities were evaluated to reduce project uncertainties, including analysis of sustainable raw materials, selection of eco-compatible materials, research, and characterization of barrier products suitable for optimizing the finished product; study and design of molds suitable for producing prototypes to be tested on an applicative scale to verify the achievement of the result. Starting from the analysis of natural raw materials, tests were carried out for characterization and compatibility with various barrier products, followed by prototypical development activities.

In the current market, there are various trays available for the large-scale retail Trade (GDO); however, these do not fully meet all the requirements listed below:

Category	Properties barrier	ISO / TAPPI /Certification authority	Target to achieve.	Definition
1	Grease resistance	DIN TEST 53116	Level 2	Medium to high grease resistance customized to the specific application
2	Water resistance	Cobb test 1800 (second)	<3 gsm	High water barrier customized to the specific application
3	Moisture resistance	DIN 53 122-1 (Hr%50, T 25°)	Wvtr <50 gr/mq*24h	High water vapor barrier customized to the specific application
4	Oxygen resistance	ISO 15105 (Hr%50%; T23)	Otr < 5 cc/mq*24h	High oxygen barrier customized to the specific application
5	Heat sealability	Internal method	from 100 ° and up thermo sealers	Suitable for using sealable film on the surface
6	Stiffness	ISO 2493-2 (mN)	from 20% and up compared to standard pulp tray	Comparable to PET trays and must maintain over time as expected
7	Compostability	Compliance to (EN 13432)	Label "OK COMPOST INDUSTRIAL"	Sustainable packaging claim
8	Recyclability	Compliance to (EN 13430)	Label "ATICELCA" Level B	Sustainable packaging claim
9	PFAs free	in accordance with European directives	in accordance with European directives	Sustainable packaging claim

TABLE 6 THE PROPERTIES THAT THE INNOVATIVE TRAY REQUIRED BY THE LARGE-SCALE RETAIL TRADE (GDO) MARKET MUST HAVE.

The table above summarizes the 9 properties required by the Organized Retail market in terms of functional and sustainability properties for innovative cellulose trays to find application for the preservation of fresh foods for a long shelf-life. Project activities will aim to achieve the targets presented above to develop and propose a new eco-friendly and innovative packaging solution.

As you can see, achieving such low gas barrier values is very challenging with bio-based materials like cellulose in this case. The biggest challenge is precisely meeting the OTR & WVTR targets that would allow defining the innovative tray as sustainable because it is bio-based, recyclable, compostable, and MAP-compatible.

Leading cellulose pulp trays available on the market

Through in-depth research based on identifying the main global producers of cellulose trays, we have identified the key available solutions, evaluating their properties and performance. For each solution identified and selected as particularly interesting, we carefully examined its strengths and weaknesses. It has emerged that, despite technological advancements and innovations in the industry,no solution currently fully meets the desired requirements for sustainability and functionality.

Cellulose pulp trays available on the market"	Description	Weak point
Hutamaki tray https://www.huhtamaki.com/en-us/north-america/consumer-goods/molded-fiber-packaging/	A tray for fresh food, made of cellulose pulp and treated with a biopolymer ensuring excellent barrier against greasy and it moist substances; it is environmentally sustainable as can be composted, being certified as such. Developed with the support of the EU's "Bio-Based Industries Joint Undertaking" research program.	Current limitation includes poor resistance to gaseous substances necessary for extending the food preservation duration. The tray does not appear to be MAP (Modified Atmosphere Packaging).

Compac Tray https://www.compac.it/wp-content/uploads/2021/07/Contenitori-in-POLPA.pdf	The tray was developed in collaboration with the company's research center. It exhibits good liquid barrier properties, excellent resistance to high temperatures, and is suitable for sealing with closure films.	Current limitation includes poor resistance to gaseous substances necessary for extending the food preservation duration. The tray does not appear to be MAP (Modified Atmosphere Packaging).
Firplast Tray https://uk.firplast.com/3-compartment-pulp-tray-12313.html	The tray is made from cellulose pulp derived from sugar cane by-product known as bagasse. It exhibits good barrier properties against fats and water and is suitable for sealing with polymeric films. It is compostable, meeting the standards set by EN 13432 regulations	The product's drawback lies in its absence of gas barrier properties, preventing it from being categorized as a MAP container. Additionally, its utility is restricted by its limited tolerance to high temperatures, up to 130°C, suitable for cooking and heating gastronomy food.
Naturesse Tray https://www.natureko.nl/en/naturesse-en	The tray from Naturesse; the cellulose comes from sugarcane waste. The tray is suitable for storing fresh foods at the counter; suitable for microwave use but not applicable for heating and/or cooking food in the oven above 150°C.	There is no evidence of environmental sustainability, such as compostability certifications; it lacks gas barrier properties, so it cannot be recognized as a MAP tray, and resistance to high temperatures is also limited.
CCM Tray https://www.compac.it/wp-content/uploads/2021/07/Contenitori-in-POLPA.pdf	The tray, developed using barrier products that offer good barrier properties to fatty and humid substances and thermal resistance up to 200°C, is suitable for cooking gastronomy foods.	The tray lacks gas barrier characteristics, and the barrier product used to provide functional barriers is no longer in compliance with environmental sustainability criteria.

TABLE 7 INNOVATIVE TRAYS AND CURRENT LIMITATIONS

In Table n°7 above, we have listed the names of the most relevant and renowned companies in the market, known for producing innovative and environmentally sustainable trays with minimal environmental impact.

These companies offer innovative trays equipped with specific functional barrier properties aimed at ensuring food preservation and structural resistance of the trays themselves. However, as can be observed, each of these solutions has its limitations, which can be attributed to functional aspects of barriers, structural characteristics such as rigidity and capacity to support the weight of the food, or even applicative properties, such as resistance and compliance with usage conditions, such as baking at high temperatures (200°C for 25 minutes).

Although it may seem like an easy problem to solve, at present there are no renewable and bio-based polymers capable of withstanding such temperatures without compromising performance and deteriorating over time.

Our project is precisely based on these limitations, with the aim of developing a better solution that can meet the market's needs and overcome the overall limitations of these types of food packaging used to preserve various types of food.

Analysis on the types of cellulose intended to produce MOCA pulp products.

Products made from pulp can be produced using various types of cellulose, each with its own distinctive characteristics.

However, to create pulp containers suitable for food storage, the cellulose must meet specific requirements.

Below is a table summarizing the key properties of the raw materials used to produce cellulose tray.

As you can see in the table below, the main parameters considered refer to different aspects and properties such as the origin of cellulose, its density which is a property that greatly influences process workability aspects; pH, a chemical property that significantly influences the nature and bonding interactions with specific products to provide barriers and functionalize the raw material in terms of performance, and then considering porosity which depends greatly on the geometric dimensions of the fiber in relation to diameter/ longitudinal length.

Properties	Normative	Target Value
Source	ISO 5267-1	Softwood
Tensile strength ISO 1924-3	ISO 1924-3	45 ÷ 65 (Nm/gsm)
Fiber length	ISO 1974	1 ÷ 3 (mm)
Fiber thickness	ISO 1924-2	25 ÷ 35 (µm)
Fiber density ISO 534	ISO 534	0.85 ÷0.95 (gr/cm^3)
pH ISO 29681	ISO 29681	5.5 ÷ 6.5
Color ISO 2470-1	ISO 2470-1	Bleached / Unbleached
Air permeance	ISO 5636-5	30 ÷ 50 (µm/Pa*s)

TABLE 8 MAIN DATA ON THE CELLULOSE USED FOR TRAY PRODUCTION.

The selection of cellulose source for pulp containers is crucial, influencing both the characteristics of the final product and the sustainability of the production process.

While wood-derived cellulose has traditionally been preferred for its availability and versatility, it has recently raised environmental concerns related to deforestation.

Therefore, alternative sources such as bagasse, a sugar industry by-product, have been explored as an opportunity to reduce waste. The use of agricultural by-products is also a step forward in valorizing agri-food waste. The analysis of external cellulose sourcing options aims to improve production efficiency.

Thus, the project has initiated experimentation focused on researching and characterizing different types of cellulose, considering specific origin and properties.

Research and experimentation on innovative raw materials

The selection of cellulose source for pulp containers is crucial, influencing both the characteristics of the final product and the sustainability of the production process. While wood-derived cellulose has traditionally been preferred for its availability and versatility, it has recently raised environmental concerns related to deforestation. Therefore, alternative sources such as bagasse, a sugar industry by-product, have been explored as an opportunity to reduce waste. The use of agricultural by-products also represents a step forward in valorizing agri-food waste. The analysis of external cellulose sourcing options aims to improve production efficiency. As a result, the project has initiated experimentation focused on researching and characterizing different types of cellulose, considering specific origin and properties. Research and experimentation on innovative raw materials. To initiate the experimental phase, we selected types of cellulose that met the specified properties. From the initial contacts with the "Sodra Group," a Swedish company leading in the production and marketing of wood cellulose (https://www.sodra.com/en/global) , we obtained a series of FA4 format samples for our tests. We received the first raw material that met the properties listed in table #3 to ensure a good starting quality that would allow excellent compatibility with the barrier products necessary to functionalize the cellulose and achieve the required targets.

To achieve initial results within reasonable timeframes, another company was also considered as a supplier of cellulose derived from bagasse.

Additionally, we researched and identified a second cellulose raw material through an Asian company specialized in producing bagasse-derived cellulose (https://www.arpz.com/) and requested samples for preliminary testing. The properties of these two raw materials, despite being of the same origin, differ in terms of physicochemical properties such as average fiber length, fiber diameter, average fibril distribution, and branching present on the main chain of the cellulose fiber, among other factors. Regarding other properties such as color grade, air porosity, and capillarity grade to liquid substances, they depend closely on the type and treatment method during the processing and usage of the cellulose. These two types of cellulose have been prepared by incorporating various by-products from the supply chain to improve their intrinsic properties, thereby facilitating the creation of optimal barrier and mechanical properties.

Experimental scale testing

The initial phase involved the basic characterization of agricultural by-products, which were then incorporated into the primary cellulose pulp mixture to assess its potential improvement in terms of properties and performance, because of that facilitating the achievement of the set targets for the final product. The various by-products underwent preliminary treatments, including drying and grinding, to facilitate their incorporation into the starting mixture and determine the maximum percentage of insertion, by weight, ensuring effective enhancement of mechanical properties and performance of the final material. For the mixing phase, a system known as a "pulper" was employed, which allowed for controlled blending of the components, thus enabling the creation of final samples for analysis. This system played a crucial role in ensuring uniform distribution of the by-products within the mixture, thereby contributing to optimizing the properties of the final material through a well-controlled and reproducible production process.

Several formulations have been carefully developed and categorized using letters to distinguish their properties and performance during the characterization phase. To enrich the cellulose raw material with greater diversity in terms of type and percentage of components, by-products from the agricultural supply chain have been added. Each by-product possesses unique morphological characteristics and intrinsic properties that could contribute to enhancing the basic characteristics of the raw material.

In recent years, there has been a growing trend in the use of agricultural supply chain by-products in the preparation of formulations, often composed of biopolymers, which have found applications in various sectors, including food packaging.

The fundamental principle of utilizing a fraction of supply chain by-products is aimed at reducing and saving virgin raw materials without compromising the fundamental properties of the biopolymer.

This added fraction is maintained at limited percentages, serving as a filler, and enhancing aesthetic aspects.

As highlighted in the table below, 9 by-products have been selected and employed in the formulation preparation phase. Each of them has been added with the aim of achieving the maximum possible concentration in the formula. The interactions obtained with the base raw material have yielded differentiated results, as each fraction exhibits specific geometric characteristics that interact differently with the cellulose fibers of the raw material.

Overall, the properties of fillers in cellulose depend on various factors, including the type of filler used, its shape, size, and concentration in the mixture, as well as interactions with other components of the material. Following the approach adopted to formulate biopolymers enriched with plant-based additives, we have developed innovative cellulose formulations intended to offer superior properties compared to standard formulations devoid of by-products. The addition of by-products can affect the properties of the final material differently depending on the type.

They could contribute to improving not only mechanical properties, such as strength, but also thermal properties necessary to ensure the applicability of the final product; reduce the density of the material, making it lighter; enhance barrier properties by reducing permeability to gases and/or aromas.

Additionally, the use of by-products can provide economic benefits, as they are often less expensive than virgin raw materials.

Below is the table with the tested formulations and general comments.

Formulation	Secondary raw materials	particle size [µm]	basic properties	Properties and characteristics of the formulated material
A 0	cellulose fibers extracted from orange peels	>500÷1000	Medium elongation	The by-product exhibits properties that blend well with cellulose pulp, allowing for thorough mixing and effective adhesion. The added quantity, expressed as a percentage of weight, was found to be 15%.

B 0	cellulose fibers extracted from grass	1000÷2000	Medium elongation	Thanks to their structure and density like cellulose pulp, they blend well and interact effectively with it. The optimal dosage percentage was found to be 25% by weight.
C 0	cellulose fibers extracted from Sulla	1000÷1500	Limited elongation	Greater steric hindrance compared to virgin cellulose fibers due to its low density. It shows good affinity to cellulose, and the maximum acceptable dosage in the formulation is up to 10% by weight of the total.
D 0	cellulose fibers extracted from wheat	500 ÷1000	Limited elongation	The material, despite having a shape like cellulose fibers, tends to detach easily after sample preparation, indicating poor interaction with virgin cellulose. This makes it not recommended for this specific application, with the maximum quantity reached being less than 5% by weight.

E 0	cellulose fibers extracted from bamboo	>1000	High elongation	The byproduct fibers exhibit poor compatibility with cellulose fibers, as they tend to detach easily. This discourages their use for this specific application. Furthermore, it was not possible to assess the percentage of fiber added to the cellulose pulp.
F 0	Cellulose fibers extracted from rice straw	200 ÷ 500	Medium elongation	This by-product was selected based on its intrinsic characteristics, deemed useful for imparting the desired barrier properties. A targeted analysis was conducted to achieve optimal dispersion within the cellulose pulp and determine the optimal insertion percentage. The most promising result was achieved with the addition of 10% by weight relative to the total.

TABLE 9 PROPERTIES AND CHARACTERISTICS OF THE SAMPLES FORMULATED USING DIFFERENT PERCENTAGES OF BY-PRODUCTS.

Here we present an example of a prototype sample obtained by formulating the new material using recipe F0 and enriching it with a percentage of rice straw, which, thanks to its nature and composition, imparts significantly improved barrier properties to the resulting compound.

Rice straw is composed of 38.3% cellulose, 31.6% hemicellulose, 11.8% lignin, and 18.3% silica. It is the presence of silica that makes this new material particularly hydrophobic.

The amount of rice straw added is equivalent to 10% of the total cellulose used.

This integration could potentially confer a series of significant improvements to the resulting material, including increased resistance to external agents such as fatty and aqueous substances, and increased structural strength. However, during the analysis of the sample, some significant considerations emerged. The preparation process followed the path of wet pulp, leading to the formation of demonstrative prototypes, essential for analyzing and characterizing them in terms of functional properties. A first observation revealed a lack of surface uniformity, resulting in increased porosity of the developed material. This non-uniformity could also cause more pronounced surface abrasion, resulting in issues both during the industrial-scale production process and in the application of the finished product. Therefore, although the addition of rice straw may offer significant performance advantages in terms of barrier properties, with a 25% reduction in moisture porosity and a 15% increase in rigidity compared to the standard recipe, it is essential to consider the potential disadvantages associated with surface non-uniformity.

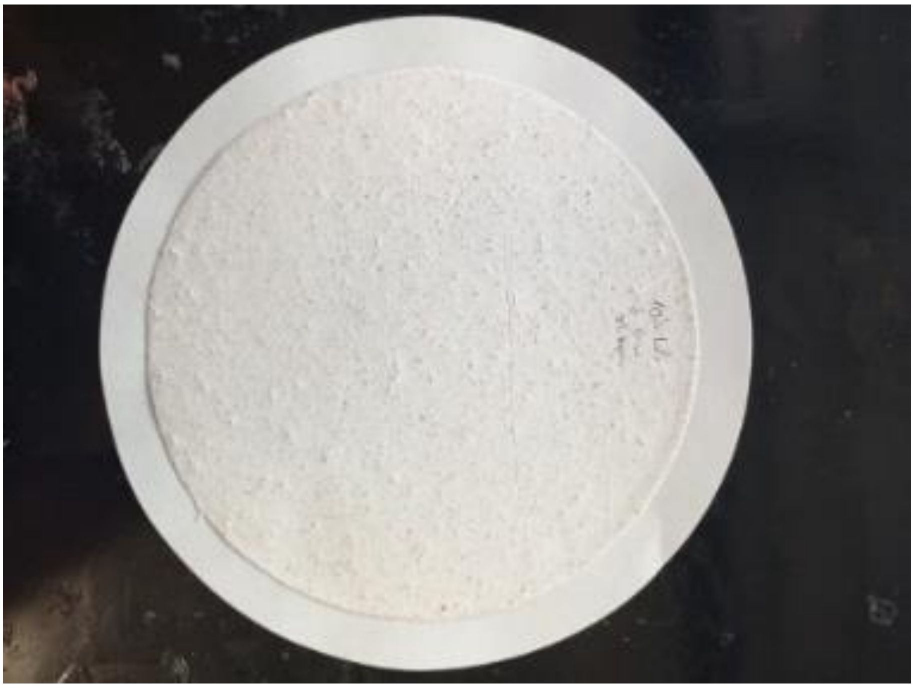


FIGURE 20 PROTOTYPE OF THE SAMPLE OBTAINED WITH RICE STRAW.

Barrier properties and applicative evaluations

Here is a summary of the results obtained from the formulations of samples with the addition of agricultural by-products to virgin cellulose pulp, aimed at developing more functional and innovative materials for prototype creation.

In all the formulations tested, a tendency of the by-product to release particles and dust from the surface was observed, thus limiting the use of the material as a base for producing trays intended for food contact.

This issue persists due to the processing technology used and will occur whenever a percentage, even optimized, of by-product is added.

However, the outcome of the formulations could be improved by considering a surface treatment to prevent the detachment of part of the innovative material and complete its functionalization.

Formulation	Targets achieved
A0	Properties 1 and 2 have not been achieved; there is no point in continuing with the other tests.
B0	Properties 1 and 2 have not been achieved; there is no point in continuing with the other tests.
C0	Properties 1 and 2 have not been achieved; there is no point in continuing with the other tests.
D0	Properties 1 and 2 have not been achieved; there is no point in continuing with the other tests.
E0	Properties 1 and 2 have not been achieved; there is no point in continuing with the other tests.
F0	Properties 1 and 2 have been achieved, but due to the detachment factor ofthe superficial particles, it is unnecessary to continue with the other tests.

TABLE 10 PRELIMINARY SCREENING OF THE ACHIEVED CHARACTERISTICS ON THE SAMPLES.

Although the treatment of cellulose with various by-products has shown positive results in terms of workability, the main challenges emerged during the analysis of the produced sheets, (see table 5).

From the initial tests conducted, it emerged that the targets related to properties 1 and 2 were not achieved as the results obtained were below the required values.

These targets concern the properties of grease and water barrier, both crucial parameters to determine the suitability of the material as a food tray.

Furthermore, the stiffness property did not perform optimally as expected, except for sample F0 where an improvement was noted.

This suggests that the composition of the by-product, particularly the percentage of silica content, plays a significant role.

During the experimental activities using a pilot mold to produce a final prototype, it was observed that the component characterizing the by-product tended to separate from the rest of the material during deposition, regardless of the type of by-product or mold parameters.

Despite the positive results in terms of workability, the main difficulties were highlighted in laboratory applicative tests, where values outside the imposed targets were observed.

Based on these results, improvement actions have been implemented to achieve the predetermined objectives. Future activities will therefore focus on describing the considerations addressed and the strategies adopted to enhance and achieve the targets related to the product under development.

Preliminary tests on bagasse cellulose and treatment with barrier products

In the second phase of the project, we proceeded with the use of bagasse cellulose, maintaining the same approach adopted previously. However, in this phase, we introduced another category of by-product during the cellulose pulp mixing phase, always keeping a focus on the properties of the raw material and following the approach of environmental sustainability.

This approach aims to save virgin raw materials and ensure compatibility with them, to achieve a more efficient process in line with the principles of the circular economy.

The by-products used are derived from the internal production of the company Ecopack and consist of paper based on virgin cellulose fiber, which undergoes no chemical modifications during the process and is considered a by-product by legal definition. These by-products have similar initial characteristics and can contribute to achieving the barrier goals set by the project.

Furthermore, within the experimental approach of this phase, we used bio-based products and/or barrier additives that interact with cellulose to impart specific properties. These products can be dispersed in water, are compatible with cellulose, and have been specifically developed to make cellulose repellent to liquid and gaseous substances.

Regarding the production of samples, prototypes were made using the most suitable by-products to allow better dispersion and improve the functionalization of bagasse cellulose pulp. To prepare the new formulations compared to the previous ones, we followed the same preparatory process, integrating the phase of treating paper trimmings. These trimmings were mechanically processed using a rotating blade mill to cut them into smaller sizes and triturate them, making them easily mixable with the bagasse cellulose raw material and promoting better interaction between the components.

Below are the prototypes made with the different formulations of Ecopack by-products, along with a description of the main aspects of each, following the same experimental approach as before.

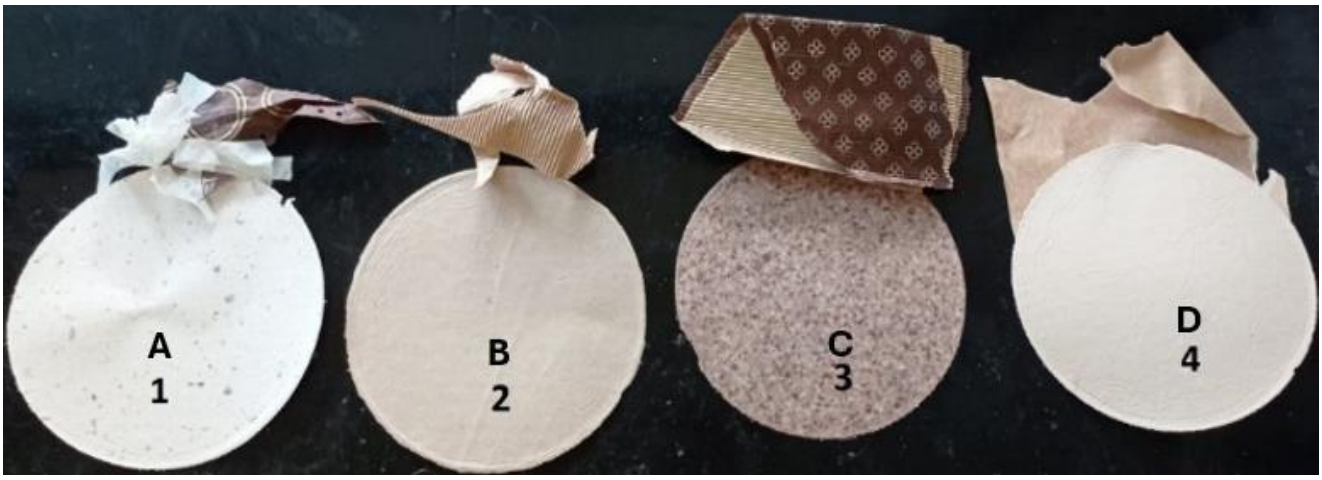


FIGURE 21 SAMPLES OBTAINED BY PROCESS BY-PRODUCTS.

The samples listed above were made using different fractions of production by-products, and they were inserted in better and more suitable percentages to achieve better results in terms of raw material savings and basic properties that can facilitate the achievement of barrier targets defined for the innovative product.

Sample A1 & B2 was obtained by incorporating a percentage of secondary raw material from white paper trimmings used in the production of products such as cupcake liners, tulips, lotus, all part of the Ecopack family sold to large bakery industries.

This formulation allowed obtaining a prototype feasible with the same production process used for standard materials of this type, without including fractions of processing by-products. On sample A1, made with this type of by-product, some dark-colored spots are visible; this is because, in line with the approach aimed at creating new innovative materials aligned with the circular economy, it was decided to also attempt to process the brown paper by-product, thus showing the result in the photo.

For sample C3, the selected percentage of secondary raw material has different properties and types of paper compared to the previous case. It consists of corrugated cardboard composed of two different types of paper bonded together with starch-based glue. In this case, the addition of this material fraction to the pure cellulose raw material caused a significant aesthetic variation in the obtained prototype, highlighting a good chemical-physical interaction between the components.

For formulation D4, a fraction of production by-product composed of papers of different thicknesses and types than the other cases above was considered. The by-product has a grammage ranging from 75 to 125 gsm and presents a surface treatment with properties such as detachment and reduced friction, slight barrier to fats and water, meeting all the requirements to be used in direct food packaging.

As in the previous cases, this by-product fraction interacts satisfactorily with the virgin cellulose fraction, allowing good results in optical homogeneity and surface uniformity on the new material.

Among the four samples produced, the best in terms of preliminary properties following basic characterization was found to be sample 1, with 10% of process by-product as the fraction and quantity added to the raw material.

In terms of barrier properties and mechanical performance, sample A1 proved to be the best. The addition of up to 10% by weight of white waste improved the values of water resistance and water vapor barrier.

Consequently, experimental material functionalization tests were conducted based on these results.

The table below shows the tests conducted by category up to number 4, which relates to oxygen gas barrier properties. Since the target value was not achieved, we proceeded with the next experimental phase, which involved testing the best barrier products for performance and compatibility with the innovative material developed.

Category	Target achieved	Definition
1	Yes, achieved	Medium to high grease proofness
2	Result almost achieved. 50%	High water barrier
3	Result almost achieved. 60%	High water vapor barrier
4	No achieved	High oxygen barrier

TABLE 11 LEVELS OF PERFORMANCE ACHIEVED FOR THE SAMPLE A1.

After examining the promising results, we have decided to proceed with experimental activities aimed at chemically functionalizing the selected material type A1 in the development phase, through interaction of bonding between the functional parts of the components, integrating into the formulation the experimentation of barrier products aimed at providing the additional functional properties necessary for the development of the new innovative MAP tray product.

Preliminary compatibility and performance tests of barrier additives

To optimize the properties of type A1 material, targeted research was conducted to identify new functional products compatible with the technological process used in the development of the new finished product, adaptable to the raw material as well. The identified additives are primarily based on biodegradable polymers and, optionally, renewable raw materials, all in the form of aqueous dispersion to facilitate application and functionalization with the base raw material.

The research was divided by barrier product category based on their initial functional properties, to verify if the interaction with the base material could lead to significant enhancements, starting from category 4, and then addressing the remaining ones.

The two categories of sought-after products are:

- Additives applicable in bulk and compatible with the raw material in the form of cellulose pulp mixed in water.
- Additives applicable on the surface through different process technologies (dip coating) and/or spray coating, the latter being the most feasible method in terms of cost-effectiveness and time efficiency in the production flow, where productivity value is crucial.

Additives developed for bulk application can be inserted at different stages of the process, and their function is to functionalize according to the interaction principle between the functional groups of the materials, generating bonds that reinforce and improve the limiting properties of the main material, in this case cellulose, particularly in formulation A1.

Among the various polymers tested, we decided to select the main ones, and two of them allowed us to achieve satisfactory results on categories that still need to be reached compared to the project target. In one case, the barrier product comes from a multinational company that designs, produces, and markets chemicals for various applications, including food packaging. The second product developed to functionalize cellulose in the form of fluff cellulose fibers, rather than paper material, comes from a start-up with which we actively collaborated to optimize the functional properties of the product and achieve the set targets.

To treat the innovative material in the wet pulp phase, barrier products are applied through various coating processes, such as rotogravure (direct application on the material to be functionalized) and flex gravure (indirect application). These are processes that allow for a surface coating application in a continuous and uniform manner.

From the numerous experiments conducted in terms of research and application tests on products of this function, the difficulty emerged in achieving all the properties per category listed previously with a single barrier product. Being functional products suitable for protecting the underlying material, it is challenging to achieve functional barriers that can provide results like those of physical barriers given by plastic films. However, satisfactory results have been achieved, allowing us to address the subsequent phases of the project. Before describing them, it is necessary to present the results obtained by barrier products listed in the table, which are protected by confidentiality agreements with the companies and start-ups with which they were formulated and optimized in terms of properties and performance.

Innovative barrier product to be performance							
Type	Category 1	Category 2	Category 3	Category 4	Category 5	Category 6	Category 7 /8/9
PB 1	achieved	achieved	achieved	achieved	No achieved	No achieved	achieved
PB 2	achieved	No achieved	No achieved	No achieved	achieved	achieved	achieved
PB3	achieved	No achieved	No achieved	achieved	achieved	achieved	achieved
PB4	achieved	achieved	achieved	achieved	achieved	achieved	achieved

TABLE 12 RESULTS ACHIEVED COMPARED TO THE PRESET TARGETS.

For the properties of categories 7, 8, and 9, since they are parameters closely dependent on the finished product that still needs to be tested, we have nonetheless included the achievement of results. This is because all the products show excellent potential to contribute to the sustainability of the finished product, thanks to the raw materials with which they are formulated and produced.

Parallel to the previously examined additives, we conducted tests on an innovative barrier product developed in collaboration with a start-up that has since become a small, limited liability company (SRL).

After numerous preliminary tests, we developed a formulation with the desired properties to overcome the limitations observed in the previous formulations.

Following numerous adjustments, the best formulation was selected, which proved to be such for the achieved targets.

Prototypical Development

After completing the experimental phases, we proceeded with the prototypical development of the tray, following the flow of the production process considered best in combination with the raw materials to develop the cellulose MAP tray, aiming to propose it as an innovative and highly functional product.

The technological approaches to realize the first demos were different, and the most innovative one was chosen with the perspective of processing the new innovative material on an industrial scale and proposing the best product also in terms of sustainability and production process efficiency.

The innovative material is initially processed to reduce it to cellulose "fluff," which is then deposited on a conveyor belt following the "airlaid" technology. This technology allows for an oriented deposition of the innovative material, enabling better interaction between the material parts that can be treated more efficiently to achieve the required functionality's uniformity and quantity of barrier product.

The process technology employed is comparable to that of the company Pulpac, specialized in developing innovative packaging solutions in cellulose material, particularly focused on producing specific packaging where high barrier properties are not required, such as caps, cutlery, spoons, etc.

Regarding the treatment of the innovative material, the best mode was found to be "spray coating" technology, which allows for controlling the quantities and distribution of the barrier product continuously and consistently through automatic process systems.

Once the innovative and functional material is prepared, it will be passed to the pre-pressing and forming station to obtain the desired product.

Below are some images of the material manufacturing process.

FIGURE 22 MATERIAL PRE-FORMING PROCESS FOR THERMOFORMING.

These aspects considered are the right premises to highlight the examined process as more effective in providing better barriers to the final product. Below is a configuration of the barrier additive application station.

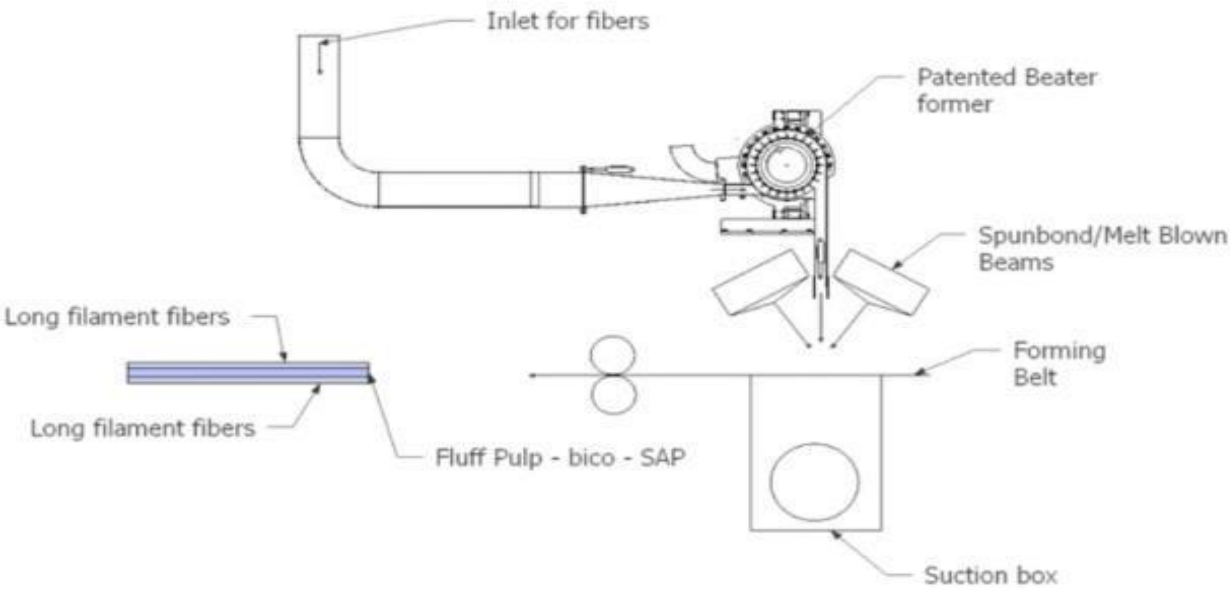


FIGURE 23 ADDITIVE PRODUCT APPLICATION PROCESS TO FUNCTIONALIZE INNOVATIVE AIRLAID MATERIAL.

As can be observed, the spray coating application system allows the barrier product to uniformly treat the entire area of the material, and furthermore, the dosage quantity can be adjusted according to the outgoing pressure flow and the diameter of the nozzles.

Experimental Prototyping Phase

Before proceeding with the forming of the samples, mechanical tests were carried out to verify machine sizing and determine the material thickness required to thermoform it during the process. Below are the results of the initial tests conducted at the company's prototype facility, which allowed us to define the most suitable thickness for the material to be processed.

The material will need to be thermoformed with a minimum thickness of 200 mm as it proves to be more compatible with the process and could potentially ensure better results even on the final product. Following these considerations, through the described process and in combination with the barrier additive from the start-up, we have produced some tray prototypes. The samples have been characterized and they show slight improvements compared to the results obtained the previous year, but unfortunately, they are still not sufficient to be recognized as MAP trays. The oxygen barrier limit remains a challenge, falling short of the imposed target. Additionally, the produced prototypes have been stacked with a view to inline production, and a tendency for the samples to stick together once stacked has been observed. To properly size the mold press and ensure the stretching of the material to achieve the desired shape, in-depth studies have been conducted that have led to important fundamental conclusions for the realization of prototypes. Given the considerable thickness of the material, the press load should be around 50 tons on average for products of small to medium sizes, such as trays with dimensions of 50*30H or 125*45H mm^2.

FigurE 24 Prototype tray produced at the end of the experimental activities.

The mold must be designed to allow the correct flow of the material during the forming and baking phases. At the same time, it must be longitudinally anchored to prevent the material from breaking under high stresses, which could cause significant deformations. Following all these considerations, the phase of preparing the first demos was initiated, and below we can present them.

Conclusions

After the experimental activities, we focused on identifying the most suitable process for working with the innovative material, thus bringing the project to completion, and producing demonstrative prototypes. These prototypes will be tested on an application scale to evaluate if they meet the desired requirements and can be considered viable eco-friendly alternatives to current MAP trays, maintaining the same functionalities but with improved sustainability criteria.

The initial application tests seem to indicate that the trays could meet market needs, which is a positive sign. Although there are still many activities to complete before the market launch, the prototypes show great potential. We have managed to achieve the functional and application property targets necessary to seriously consider the new product for the next phases.

Prospects see a growing interest from the Organized Retail market for this type of packaging, especially considering the increasingly impactful action of the European Community in replacing single-use plastic packaging with more environmentally sustainable solutions. What we have developed seems to perfectly meet these requirements and could be a valuable resource for the sector.

2. The life cycle analysis of cellulose pulp

traysIntroduction

A second activity carried out throughout the PhD. involved the LCA study conducted on different types of food trays available on the market, compared to a representative type of tray developed during the first project case study: a cellulose pulp tray with barrier properties necessary for the containment and preservation of commonly found fresh food items in GDO (large-scale retail trade) tray.[72]

The objective of a Life Cycle Assessment (LCA) of a product such as a food tray is to quantify all potential environmental impacts attributed to that specific product because of the material flows involved in and out of nature. It represents a tool for measuring the environmental performance of different products and processes. The measurement is made by evaluating the resources extracted and used in the production processes (inputs) and the emissions released into the environment (outputs) in the various processing and distribution phases, up to product disposal.[72]

Environmental inputs and outputs refer to (i) demand for natural resources and emissions/production of solid waste both during raw material extraction and production, (ii) inbound and outbound transportation, (iii) use, and (iv) end-of-life (waste management or recycling).[55] LCA analysis also allows for the comparison of different products with the same applicative function and to identify the most relevant impacts on which to focus efforts to minimize them.[72] LCA calculations often rely on a functional unit, which is the function provided by the product system. For example, when discussing food packaging, the functional unit commonly used is the kilogram of material.[72,73]

Objective

The objective of the work was to assess the environmental impact of the new packaging solution developed during the previous year, which could be a viable alternative proposal if the environmental sustainability of this container proves to be greater compared to commonly produced containers made of different materials and displayed on the shelves of retail stores.[25,26,55]

The study was conducted in accordance with the ISO 14044:2006 standard; primary data used were collected and utilized by us to address the analysis of the previously developed container. Secondary data were used for other types of containers used for comparison with the innovative solution.[26]

The adopted system model is also in accordance with the standard, as well as the research data, choice of method, and system for calculation and interpretation of results. The system boundary adopted is cradle-to-grave, and the method used for the study is "Environmental Footprint," using all impact categories included in the Product Environmental Footprint (PEF) [55,74]guidelines during the screening phase, and selecting the main categories of interest such as:

- "Carbon footprint"

The carbon footprint is an indicator of the greenhouse gas emissions generated to produce a good or service. The total amount of these emissions is expressed in terms of CO_2 eq (carbon dioxide equivalent).[75]

- "Water consumption"

Water consumption is the volume of freh water withdrawn by individuals and businesses. This includes both water consumption is linked to the concept of the water footprint, developed to improve understanding of how our production and consumption choices affect the use of globa water sources.[71]

- "Resource utilization"

Resources that are continuously replenished at a rate equal to or greater than of depletion. For example: cotton, hemp, corn, wood, wool, leather, agricultural byproducts, nitrogen, carbon dioxide, and sea salt. These materials must be produced using regenerative manufacturing practices to fit intoa circular economy.[75]

In the study, various scenarios were also analyzed regarding the production of the innovative container to assess the different impact footprints, especially from the perspective of carbon footprint, such as:

A. Mixed scenario -> production of the solution partly in the East (China) and partly in the West (Italy)
B. On-site production scenario -> entirely Italian production.

Further investigation was conducted by expanding the system and attributing environmental benefits to the disposal phase of the packaging once used; this was done following a model that allows for evaluation of potential benefits at different levels and types of disposals.

Defining the objectives to be achieved made it possible to assess the most significant environmental impacts and apply the necessary corrections to propose a reliable and true eco-friendly solution.

To address the study, we considered the functional unit of the tray to be analyzed and compared with others made of different materials, taking as a reference a volume capacity of 500 ml and, for logistics and transportation aspects, 500 km. As mentioned earlier, the study was conducted in accordance with ISO 14040 & 15044:2006, and the approach involves the various stages of the production process to arrive at the realization of the finished product and the assessment of its environmental impact once its function and purpose for which it was generated are completed (extraction and production of raw materials; transformation processes, transportation of finished products, and final disposal).

The product life cycle assessment (LCV), also known as Life Cycle Assessment (LCA), is a method designed to measure, interpret, and evaluate the environmental impacts of a specific product or service throughout its entire life cycle.

This cycle encompasses all stages, from production to disposal, including raw material extraction and treatment, manufacturing, transportation, distribution, use, reuse, recycling, and disposal.

The ISO standards of the 14040 series, such as UNI EN ISO 14040:2006 and UNI EN ISO 14044:2006, provide the international regulatory framework for conducting LCA studies. These documents establish principles, requirements, and guidelines for environmental management and life cycle assessment.

A direct outcome of life cycle assessment is the Environmental Product Declaration (EPD), a voluntary certification system born in Sweden but with international applications. This scheme, in accordance with UNI ISO 14025:2006, defines the requirements and procedures for type III environmental declarations, specifying the methods and contents of the EPD system.[26] Regarding the raw material bagasse used to produce the innovative container, the system considered begins by considering the use of bagasse for cellulose production, without considering the impacts of production to obtain bagasse.

Analytical approach

Below is a series of representations of system boundaries for each type of reference considered.

The types of trays analyzed are:

Cellulose pulp

Innovative tray aiming to be developed while meeting environmental and functional requirements to be proposed as an alternative to current MAP trays.

Paperboard + PLA

Tray made with coextruded PLA cardboard, with a weight and thickness ranging from 220 to 300 gsm.

Paperboard + PET

Tray made with coextruded PET cardboard, with a weight and thickness ranging from 220 to 300 gsm.

PET

Tray produced with PET pellets extruded into sheets with a thickness ranging from 100 microns and above, which is then processed using thermoforming technology.

Aluminum

The tray is produced starting from aluminum foil, which is folded and cut to be formed into the correct dimensions. Aluminum foil in rolls can have a thickness ranging from 20 microns and up.

Below are the system boundaries applied for the LCA study on food trays for the retail sector. The profiles presented below encompass all versions of trays currently available on the market and have been used to enable an effective comparison among them and evaluate the advantages of one over the others.

Cellulose pulp tray

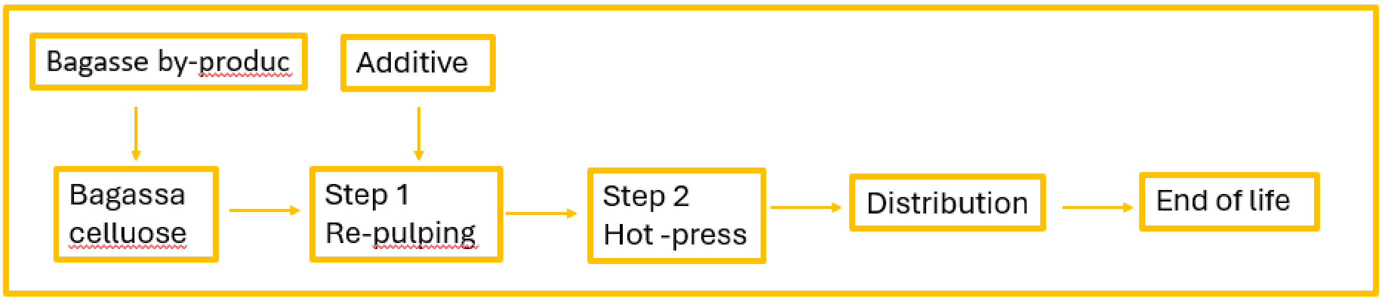


FIGURE 25 CELLULOSE PULP TRAY ANALYSIS FLOW SYSTEM.

As can be seen above, we have defined the boundary for the LCA study from raw materials to the end of life of the product.

Paperboard + PLA tray

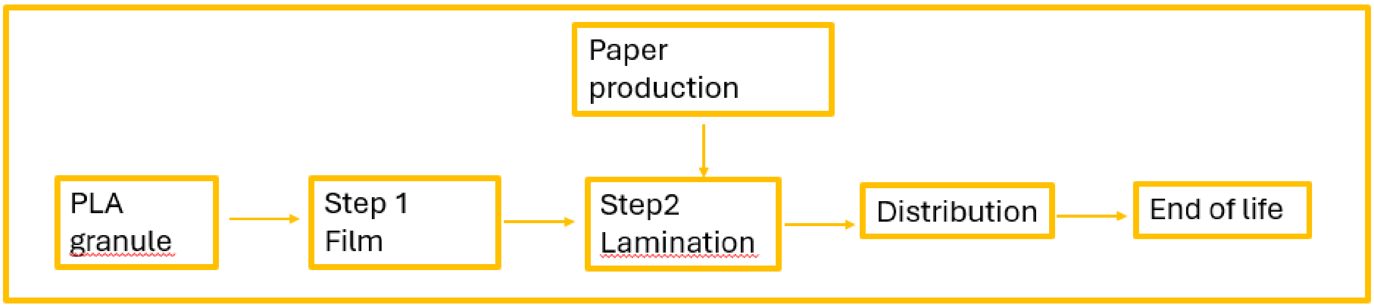


FIGURE 26 PAPERBOARD + PLA TRAY ANALYSIS FLOW SYSTEM.

Here too, the system boundary concerns the raw materials up to the end-of-life of the product. We wanted to highlight the processing stages of the raw materials used to produce the material to be processed because they differ from the previous case, and we do not know the impact they have on the finished product. It is important to consider them to allow for a more accurate product LCA study and comparison in terms of environmental benefits.

Paperboard + PET tray

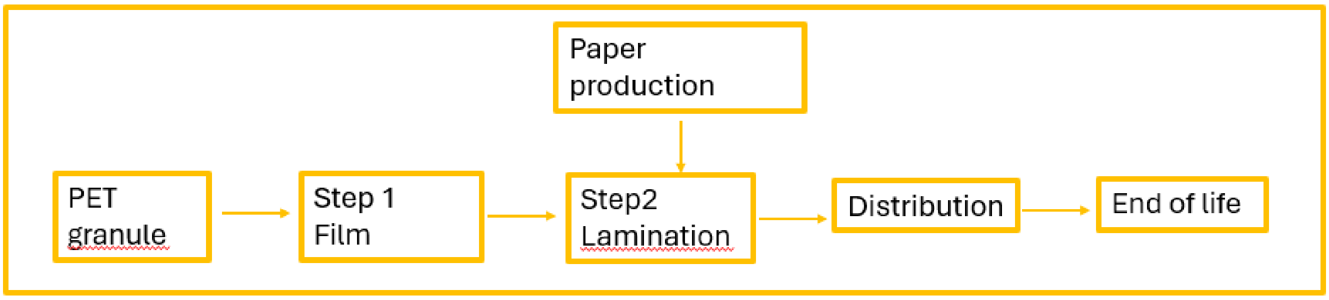


FIGURE 27 PAPERBOARD + PET TRAY ANALYSIS FLOW SYSTEM.

As in the previous case, the system boundary considers raw materials as initial data up to the end-of-life of the product, and as can be noted, the difference compared to the previous case concerns the surface material used to provide barrier properties to the cardboard and enable its application as a tray for the retail sector.

100% PET tray

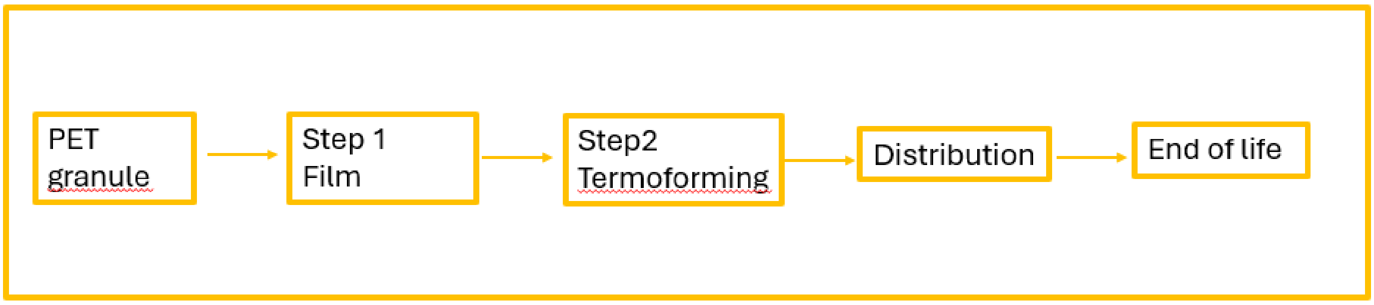


FIGURE 28 VIRGIN PET 100% TRAY ANALYSIS SYSTEM.

To assess the LCA of the PET tray, we considered the raw material as 100% without any additional components and based on virgin material.

100% Aluminum tray

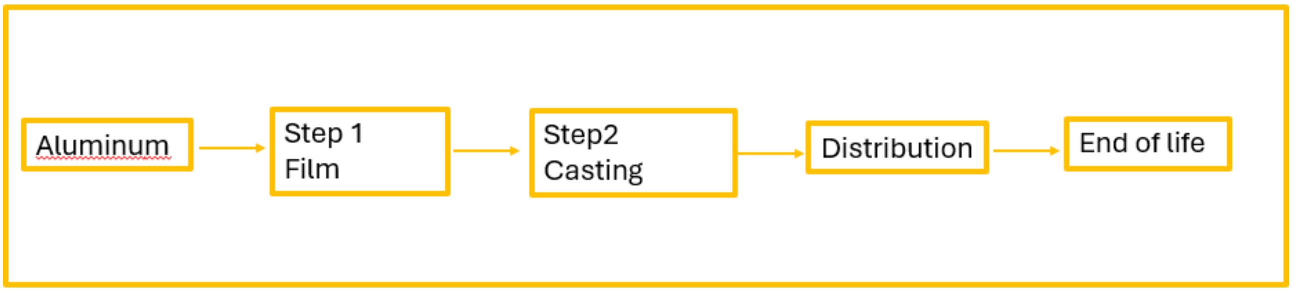


FIGURE 29 100% ALUMINUM TRAY ANALYSIS SYSTEM.

The system boundary is the same as in the previous cases; since the material differs in origin and production process to obtain it, it is also important here to consider all the stages that will lead to the LCA analysis up to the end-of-life of the product.

Data Collection and Modeling

For the modeling phase, we utilized LCA SimaPro version 9.3 software and the Ecoinvent version 3.8 database, proceeding in two distinct ways:

- Trays made from bagasse: we employed primary data regarding materials and energy provided by us, based on information gathered from our suppliers. Additionally, it was communicated that the energy consumption of the machinery used in the production and forming processes of the trays utilizes a portion of electricity derived from renewable sources.

- For the other types of trays being compared, we calculated the weights of the materials composing the trays and obtained inventory data for the products preceding the tray-forming process (such as granule production) from the Ecoinvent database.

SimaPro was developed with the goal of making sustainability a fact-based endeavor. PRé has been a leading voice in sustainability metrics and life cycle thinking development for nearly 30 years, pioneering the field of environmental and social impact assessment. We develop tools that help you create value and drive sustainable change. SimaPro is distributed through a Global Partner Network. A partner in your country will act as your local SimaPro sales and support representative and can show you a personal demo or provide more information. Find your local partner:

Where possible, we selected secondary data that best fit the substances used in the processes.

Ecoinvent 3.8, on the other hand, has been employed as a database to provide detailed and up-to-date Life Cycle Inventory (LCI) datasets for a wide range of materials, processes, and products. This database is widely used in Life Cycle Assessment (LCA) to assess the environmental impact of products and processes in various industrial sectors, such as energy, agriculture, material production, and more. Ecoinvent 3.8 contains data on resource consumption, greenhouse gas emissions, air pollutants, water consumption, and other environmental impacts associated with the different life stages of a product or process. It is an essential tool for conducting comprehensive and empirically based environmental analyses.

Analysis of tray production

Regarding the innovative tray, we examined the same production process described in the first project case. For both production processes, we used data on energy and water consumption. In the first case, the tray production was in China, while in the second case, it was in Italy. We referred to the energy and water mixes from the ecoinvent dataset, considering the geographically closest market.

For the other types of trays, differing in material and production process, used for comparison, we considered the average data provided by ecoinvent, as precise information on production sites was not available. Specifically, the following data were considered:

<u>Paperboard-PLA Tray</u>

Paperboard: Production of solid bleached and unbleached pulp

PLA: Production of PLA granules associated with the extrusion process to obtain the plastic film

Lamination process of the cardboard and subsequent thermoforming phase

Paperboard-PET Tray

Carton: Solid bleached and unbleached pulp

PET: Production of PET granules associated with the extrusion process to obtain the plastic film

Lamination process of the cardboard and subsequent thermoforming phase

100% virgin PET Tray for food contact application

PET: Production of PET granules associated with the extrusion process to obtain the plastic film

Thermoforming process

100% Aluminum Tray food contact application

Aluminum: Production of $AlMg_3$ ingot

Formation process of aluminum sheet and metal processing. In the following image, examples of the trays studied are shown. The shape of the trays does not correspond to the actual sample; in the study, the comparison was conducted using only the weight of the trays.

FIGURE 30 THE SAMPLES CONSIDERED TO ADDRESS THE LCA STUDY.

The table indicates the composition and unit weights of the trays studied.

Material	Composition (%)	Weight (gr)	Data Quality
Innovation tray	Cellulose 97.7 Additive 2.3	23.55	Primary data
CA-PLA tray	Paperboard: 89.4 PLA: 10.6	14.76	Primary data
CA-PET tray	Paperboard: 82.4 PLA: 10.6	9.34	Primary data
PET tray	PET: 100	17.32	Primary data
Aluminum tray	Aluminum: 100	8.83	Primary data

TABLE 13 QUALITATIVE DATA OF THE DIFFERENT TRAY.

As can be observed, the substantial difference between the trays is determined by the weight of each one, which is closely related to the specific weight of the materials and the geometric shape. Weight will be the factor influencing the environmental sustainability values, as it is significant for material consumption and logistic phases. For the innovative tray, we used primary data such as weight and composition. For the other trays, we relied on basic data from the ecoinvent 3.8 database.

Here is a table showing the materials and weights, as well as energy and water consumption for the various production technologies. Below are instead expressed the energy and water consumption of the processes to produce the imagined innovative trays, considering manufacturing in China, and the values are expressed per kg of material produced.

Process	Electricity (kW*h/Kg)	Natural gas (mq*h/kg)	Faucet water (kg*h/kg)	Data quality
Step 1 Pulp preparation	0.1144 (0.0343 from Renewable sources)	0.008	0.004	Primary
Step 2 Process production	0.4536 (0.1361 from renewable source)	-	0.0016	primary

TABLE 14 STAGES OF THE CELLULOSE PULP TRAY PRODUCTION PROCESS

Transport Analysis

To include the logistical aspects related to the innovative tray of project 1, we adopted two distinct approaches:

- We assessed the approximate distance between China and Italy for the delivery of the trays.
- We considered the Italian market for the distribution of the trays.

For all road transports, we used data related to freight transport with trucks weighing from 16 to 34 tons, compliant with Euro 4 regulations.

Regarding the weight of the trays, we considered the total of 6.000.000 pieces produced.

Tray	Weight (Kg)
Innovative tray	141.300
CA-PLA	88.560
CA-PET	56.040
PET	103.920
Aluminum	52.980

TABLE 15 WEIGHT OF THE TRAY.

The higher weight is found in the innovative tray as it is composed of more raw materials, but with the advantage that these are renewable and biodegradable materials.

End-of-Life Product Analysis

For end-of-life modeling, we used the latest average Italian data based on the technical reports from consortia responsible for collecting various materials (COMIECO, COREPLA, CIAL, and ISPRA).

Below are the consortium data.[76,77]

Focusing on the innovative container, we notice that about half of them, composed mainly of this type of material, have their destination in organic disposal at industrial composting facilities. There is no mention of the recycling chain in this case, as the consortium tasked with receiving these products for recycling in the paper recycling stream was unable to do so, and therefore significant data in this regard are not available.

Sample	Consortium (%)	Disposal (%)
Innovation tray	Cellulose 97.7 Additive 2.3	Compost: 51 Incineration: 48
CA-PLA tray	Paperboard: 89.4 PLA: 10.6	Recycling Incineration Landfill
CA-PET tray	Paperboard: 82.4 PLA: 10.6	Recycling Incineration Landfill
PET tray	PET: 100	Recycling Incineration Landfill
Aluminum tray	Aluminum: 100	Recycling Landfill Incineration

TABLE 16 DATI COLLECTED FROM THE TECHNICAL REPORTS OF THE CONSORTIA RESPONSIBLE FOR DISPOSING AND RECYCLING CONTAINERS MADE OF VARIOUS MATERIALS.

Environmental impact categories

There are numerous categories of environmental impact; the environmental footprint method provided in the SimaPro software, EF 3.0, has allowed us to identify these categories within the EF method. Below is a simplified list of impact categories.[55]

The SimaPro software, along with the EF (Environmental Footprint) method, allows for the assessment and analysis of the environmental impact of products, processes, or services throughout their entire life cycle, considering various environmental impact categories. The EF method provides a framework and methodology for measuring and assessing the impact on various environmental aspects, such as greenhouse gas emissions, use of natural resources, air and water pollution, and more. SimaPro is a software tool that applies this method to conduct environmental footprint analyses efficiently and accurately.

Experimental Phase and Environmental Characterizations

The results to be presented are primarily comparative and pertain to trays of the same volume. The data are expressed as relative percentages across different categories, in absolute values, and as normalized values. Normalization involves multiplying the results of the life cycle impact assessment by normalization factors to calculate and compare their contribution to the impact categories of the EF relative to a reference unit.

The results to be presented are dimensionless and normalized, expressed as scores, representing the burdens attributed to a product relative to its reference unit, such as the environmental impact generated by an average European citizen over a representative one-year period.

It is important to note that the normalized results of the environmental footprint do not necessarily indicate the severity or relevance of the impacts considered and can be interpreted differently depending on what is being reported.

We conducted an initial screening analysis using all impact categories of the EF method to gain a comprehensive overview of the environmental impacts.

Subsequently, we focused on three categories of particular interest:

- Climate Change (CO_2 equivalent emissions)

This category focuses on the greenhouse gas emissions that contribute to climate change.

It typically measures the amount of carbon dioxide (CO_2) equivalent emitted during the life cycle of a product, process, or service.

Greenhouse gases trap heat in the Earth's atmosphere, leading to global warming and climate change. Besides CO_2, other greenhouse gases like methane (CH_4) and nitrous oxide (N_2O) are also often included in this category and converted into their CO_2 equivalent for comparison.

- Water Use (consumption and deprivation)

This category instead considers both the consumption and deprivation aspects of water. Consumption refers to the amount of water withdrawn and used during the life cycle, including water used for irrigation, manufacturing processes, and consumption by humans and animals. Deprivation, on the other hand, accounts for the reduction of available water resources in each area due to the water consumption associated with the product, process, or service.

This category helps evaluate the impact on freshwater resources and can include assessments of water scarcity and water stress.

- Fossil Resource Use

Fossil resource use measures the consumption of non-renewable resources such as coal, oil, and natural gas throughout the life cycle. These resources are finite and contribute to environmental degradation and climate change when extracted and burned for energy or used as raw materials in various industries. Assessing fossil resource use helps understand the depletion of these resources and their impact on ecosystems, biodiversity, and the climate.

These categories will allow for evaluating different types of trays in terms of environmental sustainability by analyzing through environmental indicators which aspects are more impactful in terms of sustainability and for which materials and processes the trays are manufactured.

Below are the absolute impact values for all EF categories of the innovative cellulose tray, always considering the number of trays as reference.

Impact category	Units	Total	Description
Climate change	**Kg CO_2 eq**	**199.474**	**It represents all the inputs or outputs causing greenhouse gas emissions, leading to global temperature rise and sudden regional climate changes. This climate impact has global-scale consequences.**
Ozone depletion	kg CFC-11 eq1	0.02	It represents the degradation of the ozone layer in the stratosphere caused by emissions of ozone-depleting substances, such as long-lasting chlorine and bromine gases.
Ionising radiation	KBq U-235 eq	9.848	Represents the negative effects on human health caused by radioactive emissions.
Photochemical ozone formation	kg NMVOC eq	1.423	It represents the formation of ground-level ozone in the troposphere through the photo-oxidation of volatile organic compounds, carbon

			monoxide (CO), and nitrogen oxides (NOx) under the influence of sunlight. High concentrations of tropospheric ozone at ground level damage vegetation, human respiratory pathways, and artificial materials.
Particulate matter	Disease inc	0.02	Represents the negative effects on human health caused by particulate matter (PM) emissions and its precursors (NOx, SOx, NH3).
Human toxicicty, non-cancer	CTUh	0.001	Describes adverse health effects from inhaling air, consuming food/water, and skin contact with non-carcinogenic toxic substances, excluding those caused by particulate/smog from inorganic emissions or ionizing radiation.
Human toxicicty, cancer	CTUh	0.001	Represents the adverse health effects on humans caused by the intake of toxic substances through air inhalation, ingestion of contaminated food/water, and skin absorption, particularly concerning carcinogenic effects.
Acidification	Mol H^+ eq	1.817	Emissions of NOx, NH3, and SOx release hydrogen ions (H+), contributing to soil and water acidification, damaging forests, and acidifying lakes in areas with low buffering capacity.
Eutrophication, freshwater	Kg Peq	44	The lack of oxygen, sometimes leading to fish kills, can result from this phenomenon. Eutrophication, measured by the oxygen needed to decompose dead matter, is assessed in three impact categories: terrestrial, freshwater, and marine.
Eutrophication, marine	Kg N eq	479	
Eutrophication, terrestrial	Mol N eq	5.077	
Ecotoxicity	CTUe	5.179475	Toxic effects on an ecosystem harm individual species and alter its structure and function. Ecotoxicity results from various toxicological mechanisms triggered by the emission of substances directly impacting ecosystem health.

Land use	Pt	22.926094	Land use involves activities like agriculture, forestry, road construction, housing, mining, etc., affecting land quality and duration of occupation. Land use change considers the extent of soil property alterations and affected area.
Water use	**mq depriv**	**233.912**	**This refers to the remaining amount of water in a hydrographic basin once the needs of humans and aquatic ecosystems are met. The risk of water deprivation for other uses is assessed considering that a scarcer water availability increases this risk.**
Resource use, fossils	**MJ**	**2.417.418**	**Use of non-renewable fossil natural resources (such as natural gas, coal, oil).**
Resource use, minerals, and metals	Kg Sb eq	1.20	Use of non-renewable abiotic natural resources (minerals and metals).

TABLE 17 ENVIRONMENTAL INDICATORS TO ASSESS THE ENVIRONMENTAL IMPACT OF THE TRAYS

The table highlights in bold the environmental indicators on which we have focused the most in terms of evaluations and overall observations among the case studies under review.

As a preliminary analysis, we considered the innovative cellulose tray and examined the three impact categories over its entire life cycle (the three impacts are distinctively highlighted in bold). From the data obtained, the following relevant results were identified, interpreted, and reported in the table below:

The production process of extracting sulfate pulp cellulose appears to be the most impactful phase across all three categories, particularly concerning “water use”, where it stands out significantly at around 87%.

Regarding the "Climate change" category, the preparation of cellulose pulp has a significant impact, accounting for 32%, followed by the impact from tray production, representing 24.46% of the total, while the pulping process generates an impact of 6.31%.

Additionally, a notable impact contribution related to logistics has been observed, accounting for 21.17% of the total for this category. This contribution is primarily attributed to the transportation of trays from China to Italy.

In the "Resource use, fossils" category, the percentages relating to the process phases are quite uniform. However, a significant data point emerges concerning the additive, which accounts for 26.6% of the total impact. This figure is primarily attributed to the transportation of the additive to the production site, where it is used in the manufacturing process. Interestingly, product end-of-life does not appear to have a significant impact compared to other phases for this impact category.

We wanted to summarize in a table 13 the main environmental impacts that would occur in the case of production of the innovative tray. The most impactful process on all three indicators is the extractionof the raw material used to formulate the innovative material for producing the finished tray product.

Category	R.M 1	R.M. 2	Transport	Process1	Process2	Process3	Distribution	End oflife
Climate change	8.69	-0.15	21.17	32.02	6.31	24.46	5.84	1.35
Water use	8.88	-0.24	0.82	87.13	0.6	2.34	0.23	0.44
Resource e use,fossil	13.13	-0.40	26.60	32.03	4.66	17.75	7.34	0.70

TABLE 18 ENVIRONMENTAL IMPACT DATA FOR THE INDIVIDUAL PROCESS STAGES RELATED TO THE INNOVATIVE CELLULOSE TRAY.

Comparison between different types of trays and the innovative solution.

To compare the 5 tray solutions, we considered the same environmental impact categories, and the reported data are expressed in absolute terms.

Category	Units	CA-PLA tray	CA-PET tray	100% PET tray	100% Aluminum tray
Climate change	Kg CO_2 eq	+75.477	+110.815	-300.151	-439.580
Water use	mq depriv	+151.513	+192.685	-15.735	+108.932
Resource use, fossil	MJ	-190.986	+329.952	-7.033.830	-4.597.111

TABLE 19 COMPARISON OF THE ENVIRONMENTAL IMPACT FOR THE CONSIDERED INDICATORS OF THE INNOVATIVE TRAY COMPARED TO OTHERS.

Positive values, highlighted in red, indicate an increase in impact from the new innovative tray solution made of cellulose pulp compared to the alternatives currently on the market, used as references for comparison.

Negative values indicate the advantages offered by the new innovative solution, which proves to be very competitive in terms of environmental sustainability compared to the 100% aluminum tray and 100% PET versions tray.

Conversely, negative values, highlighted in black, represent a reduction in impact from the innovative tray, indicating an advantage in terms of sustainability.

For example, the innovative tray reduces emissions by 300.000 kg CO_2 eq compared to the 100% PET tray but has an impact of 110.000 kg CO_2 when compared to the CA-PET tray. The obtained values indicate that even the innovative cellulose tray causes an environmental impact and that configured as imagined so far, it would no longer be more sustainable if compared overall (for the three impact categories selected as the most interesting for us to analyze) to a cardboard + PET tray.

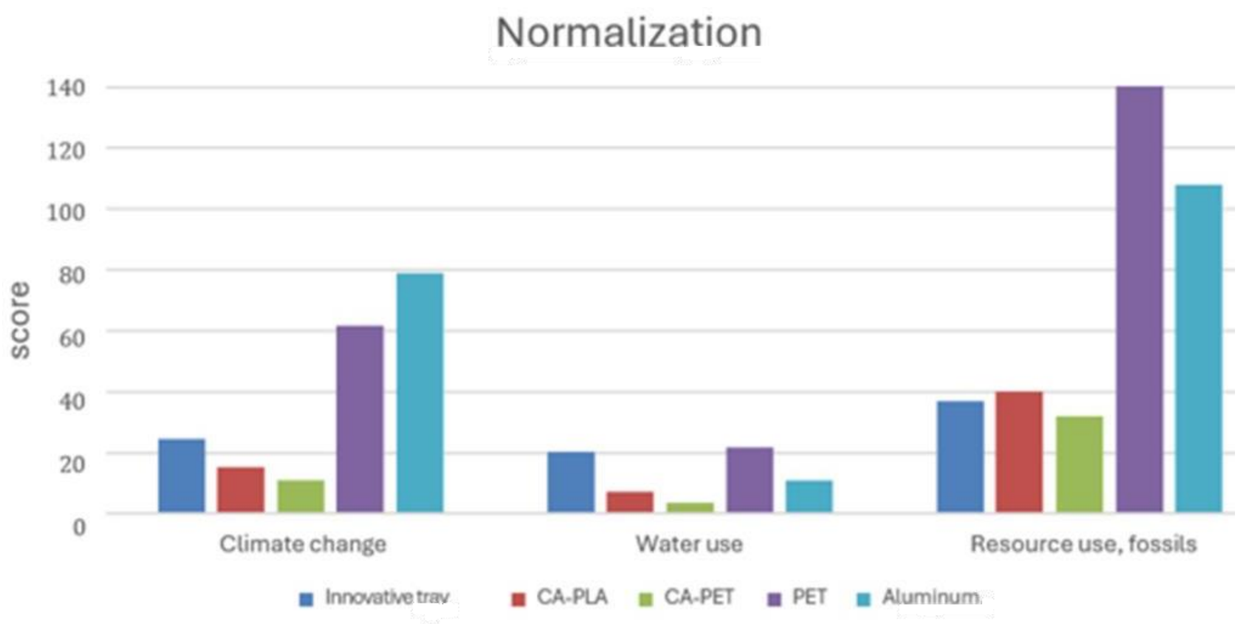


FIGURE 31 REPRESENTATIVE AND NORMALIZED CHART FOR THE CONSIDERED VOLUME UNIT

The overall impact score obtained considering the three analyzed categories shows that cellulose pulp trays have a lower total impact compared to PET and aluminum trays but are less favorable compared to paper trays coupled with PLA and PET in general considering the reference impact categories. From the data, it emerges that the "Climate change" and "Water use" categories are the ones that cause the greatest impact compared to the values resulting from paper and plastic trays. The main cause of these impacts is related to the extraction and additive process of the barrier material to produce the innovative material to be processed.

Instead, the "Resource use, fossils" category is rather similar when comparing the innovative tray to the one made of PET cardboard and PLA cardboard. It is important to emphasize that the comparison based on single scores is subject to normalization and weighting factors that are partly subjective and depend on the calculation method used.

Sensitivity Analysis

Sensitivity analysis involves modifying certain parameters to assess their effect on the overall impact of the system under consideration. Using two variables, sensitivity analysis was applied. Variation in the type of database was considered by considering the APOS (allocation at point of substitution) typeand considering both the innovative container and the cardboard-PLA container, both compostable.

To examine the scenarios, we focus on the experimental strategies associated with the first project case, aiming to further optimize the production of raw materials for the new innovative material.

In the first scenario (1.1), it is assumed that 85% of the cellulose, imported from China, while the remaining 15% is obtained from high-quality agricultural by-products sourced locally.

In the second scenario (1.2), it is imagined instead that all the cellulose raw material comes exclusively from local agricultural by-products in Italy.

As for the barrier additive, its production remains unchanged both in terms of composition and place of production. Regarding the logistical aspect, which includes the preceding phases and entry into the production process flow, we have estimated about 20.000 km. This estimate considers the transportation from China to Italy for bagasse cellulose via ship and then from the production site of the containers.

Below is the comparison of the three different scenarios to produce cellulose pulp trays from bagasse (absolute impact values).

Category	Units	Innovative tray	Scenario 1.1	Scenario 1.2
Climate change	Kg CO_2 eq	199.475	154.831	94.679
Water use	mq depriv	233.912	243.131	270.932
Resource use, fossil	MJ	2.417.418	2.163.091	1.451.541

TABLE 20 DIFFERENT IMPACT SCENARIO FOR THE PRODUCTION AND DISTRIBUTION OF THE INNOVATIVE CELLULOSE TRAYS.

From the table above, it is evident that transitioning from the preliminary approach of developing and imagining the production of innovative trays under the previously defined conditions to the approach followed with scenarios 1.1 and 1.2 would result in significant improvements regarding the climate change impact category, which would be greatly improved (from 199.475 to 94.679 kg CO_2 eq), as well as for fossil resources, where there would be an equally significant reduction (from 2,417,418 to 1.451.541 MJ). However, the impact category that could create a slightly more negative effect could be water use, as optimizing raw material consumption using fractions of by-products from the Italian agricultural chain might require increased water consumption to treat and incorporate these by-product fractions into the innovative raw material formulation.

In the table 16, the absolute values of scenarios 1.1 and 1.2 are reported. Negative figures indicate that these scenarios are environmentally advantageous for the three impact categories, while positive values indicate that further optimization is required to achieve a significantly less impactful approach compared to the preliminary configuration of the innovative trays. For example, it may be necessary to optimize production processes that consume a larger amount of water to reduce the environmental impact of this category and thus propose a truly innovative solution in terms of Life Cycle Analysis to tray.

Category	Units	Scenario 1.1	Scenario 1.2
Climate change	Kg CO_2 eq	-44.644	-104.769
Water use	mq depriv	+9.218	+37.020
Resource use, fossil	MJ	-254.327	-965.878

TABLE 21 ABSOLUTE VALUES ON ENVIRONMENTAL INDICATORS COMPARING THE INNOVATIVE TRAY BEFORE AND WITH THE TWO SCENARIOS.

Below is presented the comparison between the trays using the normalization factors of the EF method. The EF approach simplifies LCA analysis and provides results that are easier to interpret compared to more detailed methods like the Life Cycle Impact Assessment (LCIA) method, which evaluates intermediate environmental impacts.

Comparing scenario 1.1-1.2 with the innovative product, only one indicator shows a positive value, representing a negative contribution in terms of impact. All other negative values in green indicate the benefits that could be obtained if the innovative product were realized considering the assumptions made for scenario 1.2.

However, it is important to note that the EF approach may result in a loss of detail and precision, as it aggregates many impacts into a few final indicators.

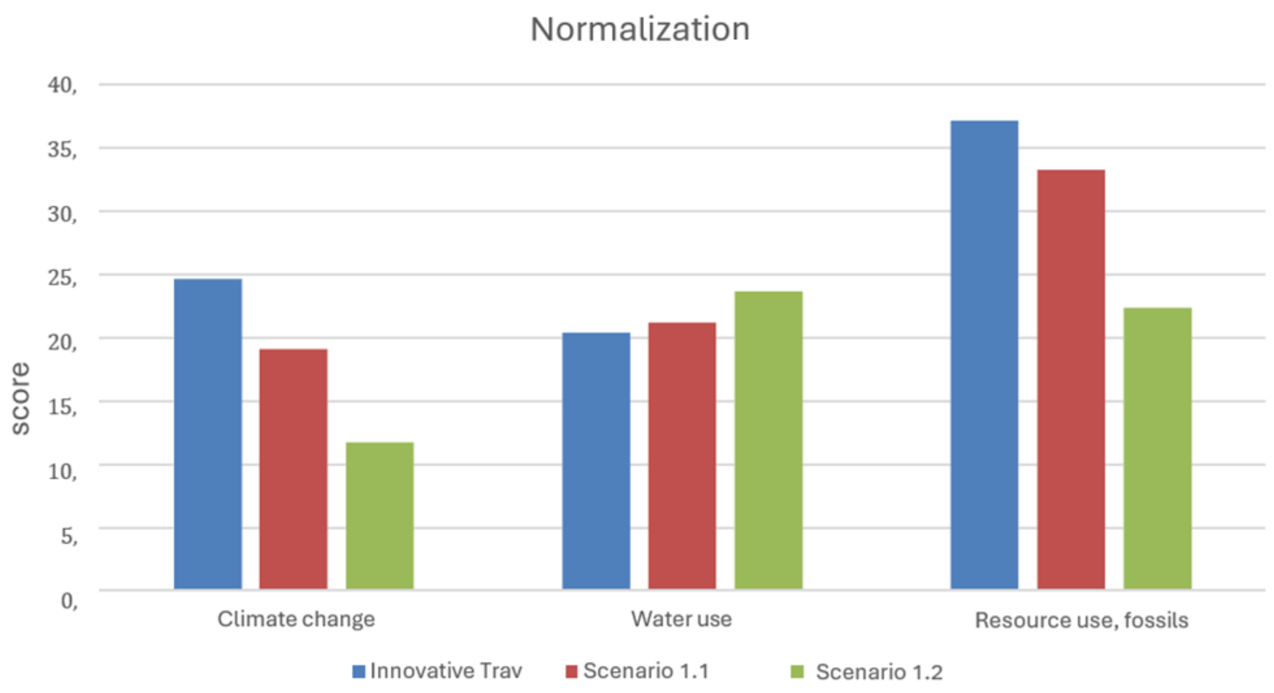


FIGURE 32 REPRESENTATIVE AND NORMALIZED CHART FOR THE CONSIDERED THE TWO SCENARIOS.

The cut-off database used in the previous study is based on a model of extended producer responsibility, where the primary production of materials is attributed to the main user. In this model, recycled materials do not give any credit to the primary producer, who does not bear the costs associated with recycling processes.

In contrast, in the APOS database, an attributional model is adopted, which shares burdens and benefits between the producer and the waste user. This methodological difference makes direct comparison of results between the two databases difficult. Therefore, the results obtained must be interpreted with caution and confirmed through specific studies.

It emerges that the differences in the environmental impact of materials are significant and influenced by the calculation methodology used.

Using the APOS system, environmental sustainability aspects will be better compared to CA-PLA and CA-PET tray versions as it is assumed that the tray is 100% recycled in composting plants (innovation cellulose tray).

The same principle has been applied to the CA-PLA product, called CA-PLA-R. However, there is still a greater reduction in CO_2 equivalent emissions with bagasse, likely due to the different disposal methods anticipated for the CA-PLA product in the base scenario.

A greater reduction would occur if we were to consider the "water use" category, where water is not depleted because the use of compost on the soil would lead to a reduction in water consumption for irrigation. Similarly, in the "Resource use, fossil" category, there would be a more or less evident reduction in both scenarios since evaluating the benefits of recycling and/or composting would result in environmental improvements.

Conclusions

The conclusions drawn from these analyses indicate that the innovative tray offers several environmental advantages compared to the alternatives examined, especially those made of PET and aluminum.

However, comparisons with tray made of cardboard laminated with PLA or PET show overall disadvantages, except in the "fossil resources" category where the innovative container exhibits a slight reduction in impact.

Sensitivity analyses conducted on different production scenarios highlight significant reductions in CO_2 emissions and energy resource use, with a positive effect on environmental sustainability.

However, it is important to note that an increase in water consumption is predicted in both scenarios.

The uncertainty analysis indicates that only the results related to the "climate change" category are statistically reliable. Therefore, while it is reasonable to assert that the CA-PET container has lower CO_2 emissions than the bagasse container, the uncertainty in the data used for the other two categories prevents definitive conclusions on this front.

Through the product LCA study, we have been able to better understand the impacts generated by different types of tray products, including the innovative cellulose tray, which has shown many advantageous aspects in terms of sustainability. However, there are also aspects that need improvement to achieve decisive results that can enable the replacement of even those tray solutions that are also sustainable because they are easily recyclable in organic and/or paper waste streams. Starting from the results obtained from the LCA study, the actions for improving the environmental sustainability of the packaging will be more specific and targeted. The approach adopted has proven to be effective in addressing the upcoming challenges of innovation, development, and continuous improvement for these categories of packaging products. It will be crucial to consider the results of the LCA study to better understand where to improve the tray and propose it on the market as an innovative solution with high value in terms of LCA.

3. Innovative and sustainable solution for manufacturing compostable products within the 'Pie'family

Introduction

During the second year of the Ph.D., specific market needs related to the production line of Ecopack's standard products, primarily known as "Pie, tart pans", here is the link where you can go to see the products that need to be improved in terms of environmental sustainability profile and functionality compared to standard performance https://www.ecopack.com/en/products/paper-moulds-with-rolled-edges/ . which are paper molds designed for containing and baking sweet foods, were highlighted. The main requirements primarily revolved around the environmental sustainability aspects of the packaging and the performance improvements achievable on the finished product through the research and development of raw materials with such characteristics, aimed at ensuring them on the finished product, consequently leading to increased market demand due to its innovative features.

Objective

Packaging products in this category present some technical limitations if they were to be used for slightly different applications than the standard ones.

The component that exhibits these limitations is the adhesive used in the paper lamination process, which induces:

- Difficulty in passing compostability tests in compliance with European regulation EN 13432.

The European standard EN 13432 provides a self-assessment structure to determine whether the standard requirements are met. However, verifying the biodegradation and compostability of packaging involves laboratory tests that require specific skills and equipment, usually not available to companies introducing packaging to the market. For this reason, our preliminary approach is to search for and select the best and most suitable raw materials to meet compostability requirements. The characteristics to be evaluated through laboratory tests are as follows:

Absence of toxic substances: Packaging material must be virtually free of heavy metals and other elements specified in the standard.

Biodegradability, meaning the tendency of packaging material to be converted into CO2 by microorganisms, like natural waste.

Disintegrability, i.e., fragmentation and loss of visibility in the final compost (absence of visual contamination).

Absence of negative effects in the final compost: Packaging material must not contaminate the final compost with eco-toxic substances and must not diminish its quality.[78]

Each of these points is necessary for defining compostability, but alone, it is not sufficient. For example, a biodegradable material is not necessarily compostable because it also needs to disintegrate during a composting cycle. On the other hand, a material that breaks down into microscopic pieces during composting but is not fully biodegradable afterward is not compostable.[78]

- Low resistance to "delamination" in the oven, as repeatedly reported by our customers.

These performance requirements are requested by the customer as evidence of the product packaging's performance limits during the production flow. The project activities should also aim to improve the structural resistance aspects of the mold throughout its usage time in the production line, preventing product components from collapsing and losing their original structure and shape.

Through research and selection of new compatible entities capable of contributing to the development of a new and innovative adhesive product alternative to the one currently in use, an Italian entity was identified for collaboration. This collaboration aimed to develop a new water-dispersed adhesive formulation that would meet the requirements to address the challenges on the finished product and adapt it to the standard lamination production process.

The collaboration with this entity was initiated through a confidentiality agreement to facilitate the exchange of sensitive data and ensure efficient management during experimental activities.

The fundamental role in achieving the project targets is closely linked to the adhesive used in the lamination phase of the papers together. Research and development will focus heavily on optimizing this raw material to meet performance and sustainability targets.

The adhesive to be developed should contain predominantly biodegradable components; it should be highly functional to allow proper functionalization and adhesion strength between the paper parts and resistance to the conditions and process parameters of the production line it will be destined for.

Description of the development plan

The experimental phases of this project involved testing on raw materials; developing different formulations for composition and properties, and once the best one was identified, transitioning to prototype development, which involved producing finished sample products for internal testing through and application tests, and external testing through interested customers.

Below is an outline of the flow of activities performed, highlighting the objectives to be achieved for each phase.

Phase	Design solution Phase 1	Approval products Phase 2
Objective	Formulation of water-based adhesive. Testing of the new formulation	Verify the performance tests onthe prototypes and products produced during the sampling phase
Price (Economic sustainability)	-0.07 euro/kg step 1 -0.09 euro/kg Step2	Provide cost-effective solution, aiming at least a 3,5% saving on the new glue to compare the standard
Timeline	Seven months	Two months

TABLE 22 PROJECT ACTIVITY PHASE.

Below, in the table 18, the theoretical process parameters hypothesized are reported and compared with the actual production to obtain the economic savings data resulting from the switch from standard glue to innovative glue. The industrial cost analysis was used to verify the economic feasibility of the project and thus proceed with the experimental and subsequent phases. As can be observed, the targeted development for the replacement of the raw material "glue" would lead to a final economic saving of approximately 16%.

This data allowed us to proceed with the gate of the subsequent activities.

Finished product cost	Cost €/mq	dry glue (%)	Machine speed (mt/min)	wet of glue (Gsm)	Gsm dry of glue (Gsm)	Overall saving
A RIF.	2.11	32	50	30	0.05	Rif.
B NEW SOLUTION	2.04	30	20	15	0.10	-16%

TABLE 23 ECONOMIC FEASIBILITY IN FORECASTED TARGETS FOR BOTH THE GLUE AND THE PROCESS.

Phase 1

During the initial experimental phase, there are several aspects to consider. The first is ensuring that the formulated raw material complies with current regulations and laws. This means verifying that it does not contain prohibited chemicals and that it is produced in accordance with ethical and environmental standards established by law.

Regarding quality, it is important that the formulation meets the required standards for the final product. This involves evaluating its physical, chemical, and biological properties to ensure consistency and reliability in use.

Compatibility is additional crucial aspect to consider. It is essential that the formulation fits well with the materials it meets and the application process used. Additionally, reliability is essential to preserve the physical and chemical characteristics of the raw material and, consequently, of the finished product over time.

Finally, the sustainability of the formulated raw material is essential to meet the required standards, including compostability for products in this category. Ensuring that the raw material is sustainable contributes to ensuring a positive impact on the environment and society.

The table 19 specifies the physical, chemical, and applicative properties based on the formulations that will be developed as improvements compared to the standard.

Physical properties	Chemical properties	Application test of the formulation
Viscosity (sec) (TF4 instrument)	Composition	Mechanicals rigidity Bending test
Density (gr/cm^3) (pycnometer instrument)	Percentage of components	Tear resistance Dynamometer
weight of solids content (%) Gravimetric analysis	Properties of components	Wettability properties Contact angle of drop

TABLE 24 CHEMICAL; PHYSICAL AND APPLICATION PROPERTIES TO ANALYZE IN THE NEW ADHESIVE FORMULATIONS.

Initially, we received several existing formulations from our partner company, which served as a starting point for our project to develop and improve the new product.

These formulations, provided by the operational reality, represented an important reference point for evaluating current performance and identifying areas where modifications were needed.

By carefully examining these formulations, we analyzed their characteristics, including physical, chemical, and functional properties, to fully understand their behavior and limitations.

This approach provided us with a solid starting point for our optimization process, allowing us to formulate effective strategies to enhance the product in line with established objectives.

Here is the list of measured results on the various formulations and internal evaluations.

Initially, we proceeded to develop several basic formulations, made possible through collaboration with a company operating in the adhesive sector and with support from the Chemistry Department where I pursued my doctoral studies. The initial formulations were created as an improved reproduction of the standard aqueous base ethylene vinyl polymer glue. Through these formulations, we conducted initial tests to evaluate performance and identify areas for improvement. By characterizing the formulations, we were able to appreciate and recognize the best characteristics and rheological properties compared to the standard.

This was observed as we transitioned from synthetic base formulations to those based on various biopolymers (some protein-based and others vegetable biopolymer).

Of all the formulations developed, we decided to present those that yielded promising results. From these, the best one was selected based on the predetermined project targets.

Physical properties	Formulation1	Formulation 2	Formulation 3	Formulation 4
Viscosity (sec)	>120″	>120″	>120″	>120″
Density (g/ml)	1.02	1.02	1.05	1.05
Dry glue (%)	30	30	35	35
Bending resistanc e (mN)	90	70	85	120
Coating	uniformity	uniformity	uniformity	uniformity
Tear test	Compliance with target	Compliance with target	Compliance with target	Compliance with target
Test delamination	negative	negative	negative	Compliance with target
Wettability (°)	50	45	60	40

TABLE 25 LIST OF FORMULATIONS AND PERFORMANCE RESULTS FOR EACH ONE.

Formulation 4 has enabled us to achieve the desired results through a specific combination that considers the compatibility of the formulations with the production process in the machine.

Compared to previous formulations, in this case, we optimized the performance of the adhesive components by reducing the content of mineral fillers and replacing them with a plant-based component, microfibrillated cellulose.

This strengthened the polymer structure of the adhesive, consequently improving its adhesion to paper.

Formulation	Properties	Observation
1	Compliance with Physical properties and mechanical properties on final product	The adhesive exhibits good physicochemical properties: from an application standpoint, it is suitable for the Ecopack production process. The observed limitation is due to the delamination test result. The formulation components meet excellent requirements for achieving compostability in the final product
2	Compliance with Physical properties and mechanical properties on final product	The above issues persist. The chemical properties continue to be confirmed
3	Compliance with Physical properties and mechanical properties on final product	The issue of delamination is recurring, like to the previous occurrences; additionally, accelerated degradation has been observed in the formulation. The chemical properties continue to be confirmed.
4	Compliance with Physical properties and mechanical properties on final product	With this latest formulation, successful resistance to delamination of the coupled paper has been achieved, as confirmed by preliminary oven testing at previously established high temperatures.

TABLE 26 CHARACTERIZATIONS AND OBTAINED RESULTS.

With Formulation 4, excellent resistance to delamination has been achieved, along with the other evaluated parameters, allowing for the optimization of laboratory-scale activities and the development of the first scalability test with the aim of obtaining prototypes for the validation step.

For sampling activities, it was necessary to recognize and define the process targets to be applied. One of the fundamental parameters to consider for technical and economic aspects is the maximum and minimum quantity of product to be applied; in this case, the dry weight of the product should be approximately 20 gsm, as it would allow the finished product to have good resistance in properties and performance, further facilitating the achievement of product sustainability, as it should guarantee obtaining compostability certification.

Also, the interaction and bonding time between the two papers should be respected and be approximately 7 seconds to allow for good applicability in production.

Finally, the parameters that will affect the final performance also include the residual moisture content of the semi-finished product, which should meet the target of 20% for good workability of the paper, and for good process efficiency, the target of 50 meters per minute should not be exceeded.

Phase 2

The test in the machine using the process technology recognized as semi-flexographic system at the laminating machine for papers represents a crucial phase in the production cycle of printed materials and packaging. This technology, which relies on the use of rubber or steel rollers allowing the transfer of the product to be applied on cellulose substrates and thus on paper, offers high performance in terms of versatility and precision. To effectively manage the sampling flow during the process, the company leverages the "Salesforce" tool, a platform that provides a solid infrastructure for the traceability of business activities, allowing for detailed and accurate recording and monitoring of each phase of the production process.

During the initial machine test, conducted in accordance with the specific sampling case (case number 6713), crucial data were collected to evaluate the process performance and the performance on the semi-finished product. These data are essential for understanding the current state of the standard production process and for identifying any areas for improvement.

Among the results of the process test, several fundamental metrics stand out.

The recorded process speed of 22 meters per minute is a direct indicator of the relationship between process efficiency and the quality of the semi-finished product ensured by the optimized formulation.

Additionally, the residual humidity after lamination was recorded at 40% relative humidity, crucial for assessing the quality of the final product and its resistance to moisture, particularly important for specific applications in food packaging.

The applied grammage of the optimized formulation was approximately 15 - 20 grams per square meter, necessary to ensure good bonding between parts and stability on the semi-finished product. Moreover, the formulation exhibited a slower bonding reaction time compared to the standard glue. Based on initial process measurements, a more in-depth analysis was deemed necessary to identify the causes of discrepancies and implement any necessary corrections or optimizations to ensure the achievement of predefined quality standards.

The second process test was conducted following the case described, applying initialimplementations throughout the process, including modifications to the spreading system and prolonged heating of the semi-finished product before it was wound into a roll.

The speed during the semiflexo, technology coating, where the component to be used is applied by a cylinder recognized as anilox. This method allows for a uniform doctoring and offers greater flexibility in adjusting the thickness and quantity of adhesive applied to the printing surface. coupling phase wasrecorded at 15 m per minute. The residual humidity after coupling remained unchanged at 40% relativehumidity despite extending the drying path of the semi-finished product. The grammage of the dry gluewas measured at 17 gsm. The required bonding time was found to be over 10 seconds per 100 °C. To reduce the percentage of moisture retained in the laminated paper, the installation of an infraredlamp was proposed and implemented to lower the moisture content on the finished product. However,the result was not confirmed as the moisture retained by the material remained elevated, causing aloss of performance on the prototype produced.

Despite the efforts made, most of the objectives set for the paper coupling process were not achieved, like those of test 2.1.

Compared to previous cases, further modifications have been made to the plant by implementing a heating device for irradiation using IR lamps with a frequency suitable for the vibrational modes of the water molecule.

The relevant data revealed that allowed achieving a good result on the semi-finished product was the moisture content retained by the semi-finished product, which decreased by approximately 50%, from 40% to 20%.

The other parameters have not changed and remain the same as those recorded in previous phases.

The semi-finished product was processed in a controlled flow, and it was possible to produce finished products while keeping the process parameters unchanged during the thermoforming process.

Below is the representation of the finished product that was created and then tested on an applicative scale to evaluate its actual performance and replicate the process flow adopted by industrial bakery entities.

FIGURE 33 PHOTO OF THE FINISHED PROTOTYPE PRODUCT PRODUCED.

Conclusion

From a process perspective, the necessary lamination speed to transition from the testing phase to production and accurately assess compatibility with the production flow was not achieved. The attained process speed of approximately 17 meters per minute was insufficient to ensure adequate production efficiency for the laminated product. To overcome this technological limitation, it will be necessary to configure the production plant for the laminated product in a more innovative and flexible manner. Two improvements will be required for both production efficiency and the performance of the finished product: the adhesive doser must be designed with the appropriate hardness (shore) to allow for uniform distribution and the correct quantity, and the drying method must follow the principle of initial drying of the adhesive spread on the paper, extending the drying time compared to the standard to allow for increased water evaporation and enhanced bonding strength.

Regarding prospects, in addition to technological improvement, it will be necessary to identify the worst-case scenario within this product family to conduct compostability tests and achieve compliance with EN 13432 standards, if the test results are positive, within the imposed limits. This will further enhance the value of this product line, aligning with the principles of circular economy packaging.

Lastly, it is important to highlight significant technical successes achieved with the finished product, ensuring qualitative compliance. In the future, this innovative recipe can be applied to the bakery packaging market, meeting food safety regulations.

4. Life cycle assessment on the product case study of the paper baking mold categoryPremise

Focusing on the project activities from the previous case, a Life Cycle Assessment (LCA) was conducted on the finished product, evaluating it in its standard configuration. The study was undertaken with the support of the University, and through the provision of accurate data and information by Ecopack, it was possible to proceed with the study on the standard product. The aim was to demonstrate the product's sustainability and identify the limitations that needed improvement through actions and leverage points to achieve better environmental indicators.

In this scenario, one of the raw materials used to produce the standard product might not contribute to improved product sustainability due to its chemical nature and the quantity used in the process.

It is precisely for this reason that the LCA study was conducted to understand the sustainability factors to be applied in proposing a more sustainable alternative product solution, adopting the applicative approach related to the previous case study.

Nowadays, market choices are increasingly driven by the environmental performance of products. To this end, LCA is one of the most important analytical tools to provide the scientific support necessary for engineering solutions to sustainability.

The LCA study was conducted by the research group of the "Department of Science and Technological Innovation" (AL) in accordance with the ILCD (International Reference Life Cycle Data System) guidelines. It consists of a report and a computer model, created using the "open CA v.1.11.0" software and based on both primary data and the "ecoinvent v3.8" database.

Objective

The LCA study conducted from cradle to gate on the worst-case product was focused on improving aspects such as: reducing the use of raw materials; disposal of the product at its end of life, leading to improvements in disposal in terms of compostability and recyclability, and minimizing the percentage of industrial waste through engineered systems. The same actions were taken at the experimental applicative level to meet the design needs of the previous case study.

The LCA analysis was performed on a reference product representing the Pie product category; the product code is 8018030B. It is a round cake mold produced at the Ecopack plant in Italy, TO, by laminating two pieces of grease-resistant brown paper, treated with silicone on the inner side.

Two different end-of-life scenarios are hypothesized: waste disposal in industrial composters and waste valorization through standard paper recycling processes. Waste disposal in industrial composters, defined as scenario A, involves a controlled decomposition and humification process of biodegradable materials in an aerobic regime. Scenario B assumes that the end-of-life product is 100% recyclable, and the recycled material becomes part of the recycled paper for graphic paper production.

For the evaluation of the two EoL scenarios, the finished products are considered as input flows since the use phase is not considered in the present study.

The study aimed to promote and integrate sustainability in the operations and value chain of the company Ecopack, aspiring to become a model for other entities in this sector to follow. In 2021, the company received the prestigious "Gold" recognition from Ecovadis, an award given to companies evaluated for their CSR activities and sustainable purchases. This demonstrates the ongoing commitment to improving internal aspects to be increasingly sustainable. In the coming years, attention will be focused on reducing the use of raw materials, sustainable disposal of end-of-life products (compostability/biodegradabily) and minimizing industrial waste through engineered systems.

Given the overall objective and purpose of the study, the following functional unit was chosen as the single product unit, with a weight of 8.26 grams (+/- 5%).

This report focuses on one of the company's products and has been carried out in accordance with the ILCD guidelines (European Commission. Joint Research Centre. Institute for Environment and Sustainability, 2010).

Below is the table with the complete description of the product specifications.

Item Code Producer	8018030B
TOP diameter(mm) ±5%	Inner 189; outer 206
Botton diameter (mm) ±5%	180
Height (mm) ±5%	30
Weight (gr) ±5%	8.26
Capacity (ml) ±5%	789.9

TABLE 27 SPECIFICS OF THE PRODUCT 8018030B

The system boundaries may vary in relation to the different parts of the analysis, as can be observed below; furthermore, the use-phase is not considered in the actual study but only as a subsequent scenario.

Boundary analysis of the system

To trace the product profile, the entire production chain has been analyzed, from raw materials for producing product components to packaging the finished product.

The starting point of the system is the manufacturing of the raw materials necessary for product production.

The paper used has natural grease-resistant properties, is colored, and treated with silicone on one side, manufactured from virgin fiber with high chemical purity.

The glue used to laminate two sheets of paper together is a water-based adhesive where an ethylene vinyl acetate copolymer is dispersed without the addition of any plasticizers.

For the paper production process, primary data were used, while for the glue, we used secondary data from the production inventory. Transport to the Ecopack plant was also considered, along with emissions and energy consumption for both materials based on the database.

Regarding the product assembly phases such as lamination, cutting, thermoforming, and repackaging, primary data on energy consumption and raw materials were used.

Emissions were considered based on secondary data.

Analysis flow of item

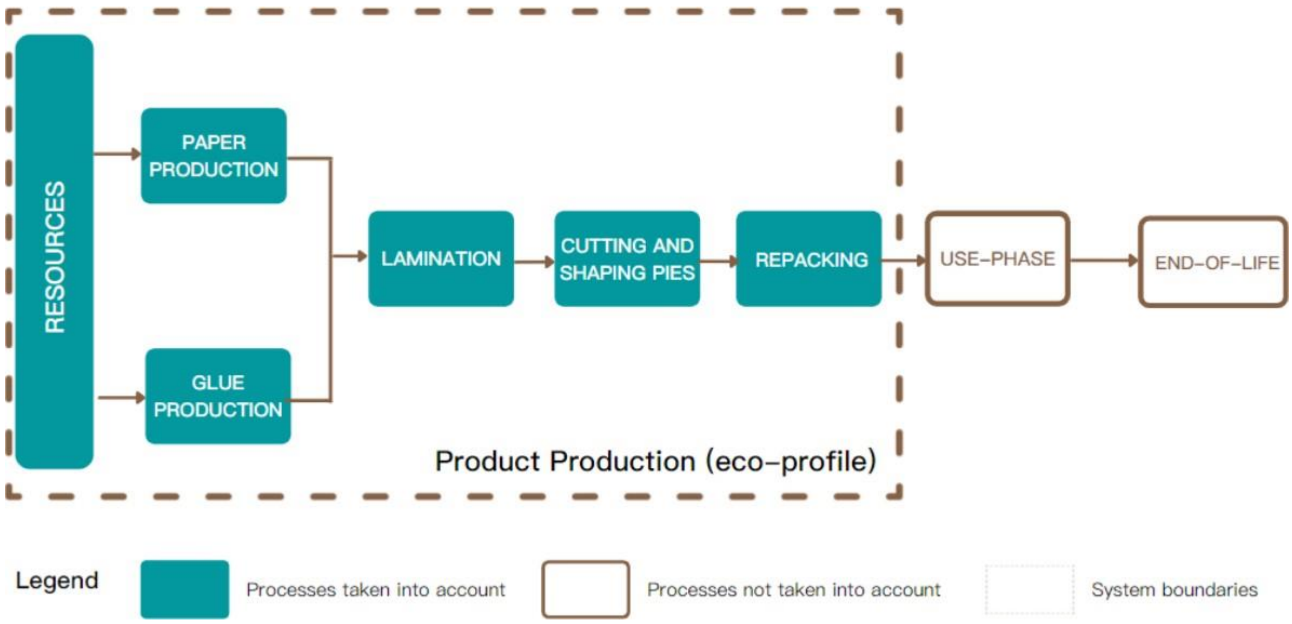


FIGURE 34 PRODUCT LIFE CYCLE DIAGRAM AND RELATED STEPS OF ANALYSIS, CASE ECO-PROFILE.

To draw the environmental profile of the pie mold 8018030B, the entire production chain has been analyzed, from raw materials production to the packaging of products at Ecopack S.p.A. Therefore, the system boundaries related to this part include all the life cycle steps from cradle to the outlet factory gate. Figure 3.1 represents the graphical diagram of the product life cycle and related analysis steps. Above is depicted the analysis flow involving the various process stages, delineating the system boundary up to the production of the finished product.

Flowchart

Based on the data and some assumptions described above, the environmental impacts related to the production of the product will be presented in this section. The study of the environmental profile also highlights any critical points that could be addressed by the company in planning future activities. Below Fig. 41 is the graphical representation of the modeled product system related to the functional unit of 1 round-shaped pie mold, type 8018030B.

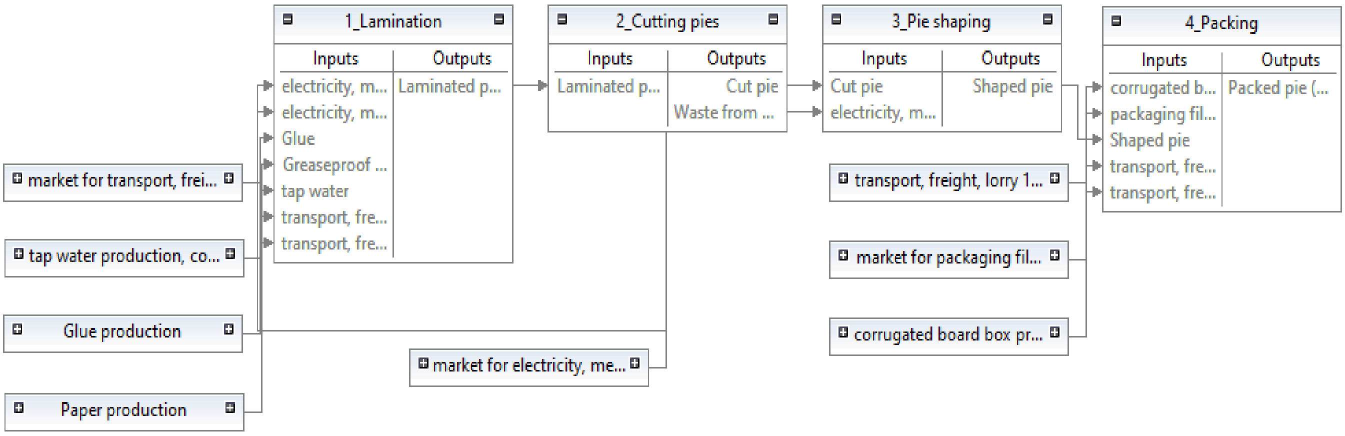


FIGURE 35 MODEL GRAPH OF THE SYSTEM MODELLED FOR IMPACTS' ANALYSIS, CASE ECO-PROFILE.

The system flows to assess the product were correlated with the amount of material required for product manufacturing and are therefore mass flows.

The finished product was then used as input data for the product systems of the two scenarios that will be presented below, considering two different approaches following the use of the packaging in question: Consideration 1, destination of the code in the paper recycling stream, and Consideration 2, destination of the code in the industrial composting stream. The finished product was then used as input data for the product systems of the two scenarios for recycling the finished product once used. For both scenarios, the system boundaries include transportation of the products to the waste treatment plant and related energy consumption and emissions. Comparing these scenarios aims to provide an approximate estimate of their respective environmental impacts and actions for improvement to achieve a more virtuous finished product.

Life cycle analysis of case A & B

The finished product was then used as input data for the product systems of the two scenarios for recycling the finished product once used. For both scenarios, the system boundaries include transportation of the products to the waste treatment plant and related energy consumption and emissions.

Comparing these scenarios aims to provide an approximate estimate of their respective environmental impacts and actions for improvement to achieve a more virtuous finished product.

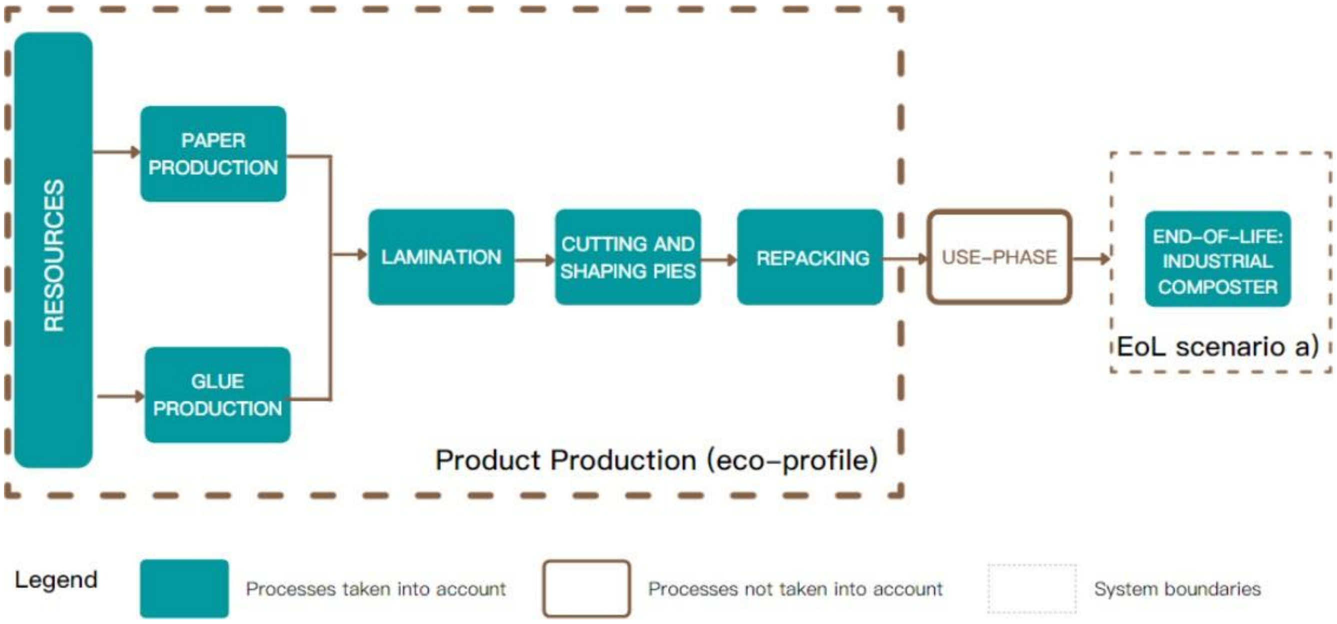


FIGURE 36 PRODUCT LIFE CYCLE DIAGRAM AND RELATED STEPS OF ANALYSIS, CASE EOL SCENARIO A)

In this case, EoL scenario A aims to analyze the product's benefit if it were destined for recycling in the composting stream.

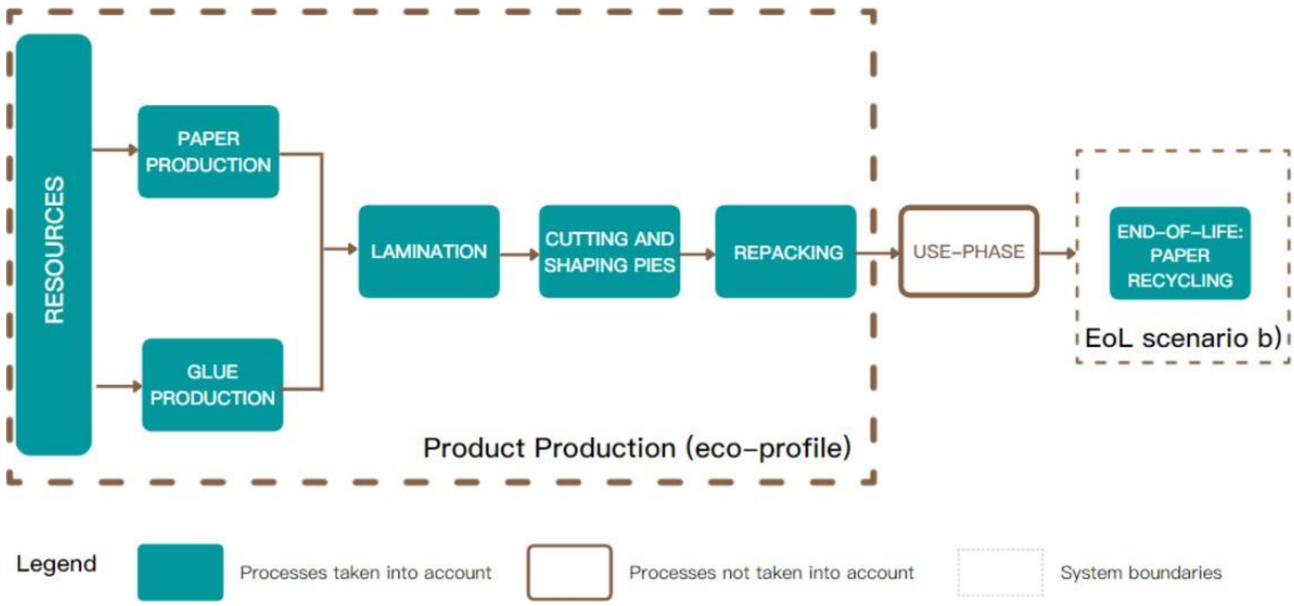


FIGURE 37 PRODUCT LIFE CYCLE DIAGRAM AND RELATED STEPS OF ANALYSIS, CASE EOL SCENARIO B)

In this case, however, the benefit was evaluated considering the end-of-life disposal of the finished product once its use is completed, in the paper recycling stream. The end-of-life scenarios were analyzed using the ecoinvent method. For both of these scenarios, the system boundaries include the transportation of products to the waste-treating facility and related energy consumption and emissions. Only secondary data were considered, sourced from the ecoinvent database and related to activities located in Europe. The transportation contribution was derived by assuming a mean distance of 40 km by lorry. Indeed, since the product is sold to private customers, it is likely to enter waste-treating facilities that are usually located close to municipalities. As stated before, the investigation and the subsequent comparison of these scenarios are intended to provide a rough estimation of related environmental impacts.

Life cycle inventory analysis

The primary data considered in the various process stages were critically evaluated in relation to their ability to meet the objectives and scope of the study. They were then input into the open-source software "open CA v.1.11.0". The LCI database used as a reference is "ecoinvent v3.8", which is based on the so-called "cut-off" approach. This means that the primary (first) production of materials is always allocated to the primary user of a material. Therefore, waste is the responsibility of the producer, incentivizing the use of recyclable products, which are available without burdens ("cut-off"). This choice allows for the tracking of the product's environmental profile based on the "polluter pays" principle.

The figure below represents the analyzed system and highlights the input and output flows for the environmental profile case, as will be discussed in the following paragraphs.

All the primary data related to this production come from "Nordic Paper Holding AB" and are entirely reported in the "Annexes" section of this report.

They mainly comprise:

- the ingredients list for the paper's production, as delivered to Ecopack S.p.A.
- the flow-chart for the production process, with specifics of energy consumption.

In details, the ingredients list provided by the Company is reported in Table ….and refers to the production of 1 ton of paper.

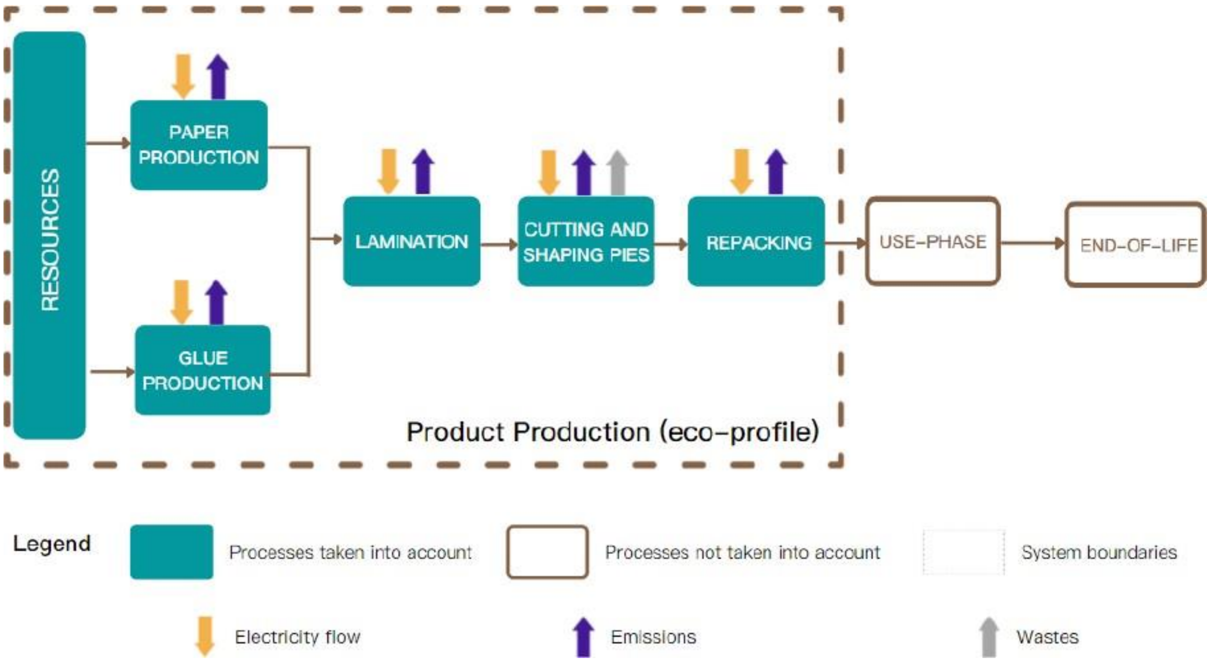


FIGURE 38 SCHEMATIZATION OF THE STUDIED SYSTEM WITH THE UNDERLINING OF THE INPUT AND OUTPUT FLOWS CONSIDERED, CASE ECO PROFILE.

Component	Amou nt
Bleached sulphate pulp (Ton)	1
CMC, 2-side treatment, dry cotton (gsm)	0.15
Color additive Blu (Kg/ton)	1.4
Color additive Red (Kg/ton)	2
Color additive Yellow (Kg/ton)	1.7
Fixative (kg/ton)	1.5
Foam, silicone (Gsm)	0.02
Foam white water trade goods (Gsm)	0.15
Magnesium sulphate heptahydrate (Kg/ton)	7.5
Silicone treatment 1-side, dry content (gsm)	0.6
Talcum (ton)	0.002

TABLE 28 PRIMARY DATA RELATED TO THE INGREDIENTS' LIST FOR PAPER PRODUCTION, AS DELIVERED TO ECOPACK S.P.A.

This list also includes the addition of three different color additives (yellow, red, blue), totaling an overall amount as low as 0.51% per ton, and "talcum" (talc) in an amount as low as 0.2% per ton. However, due to these low contributions and the absence of further details regarding the compositionof additives, these contributions were not considered. Additionally, the addition of "fixative" is not accounted for due to its limited amount (0.15% per ton), the absence of further details about its composition, and the lack of secondary data from the ecoinvent database. Generally, no details regarding suppliers were provided, so average European processes from the ecoinvent database wereused as references. Since the Producer specified that the utilized "bleached sulphate pulp" consists of long softwood fibers (from Spruce and Pine) with a smaller addition of short fibers from Spain, data related to kraft pulp obtained from northern European softwood was chosen.

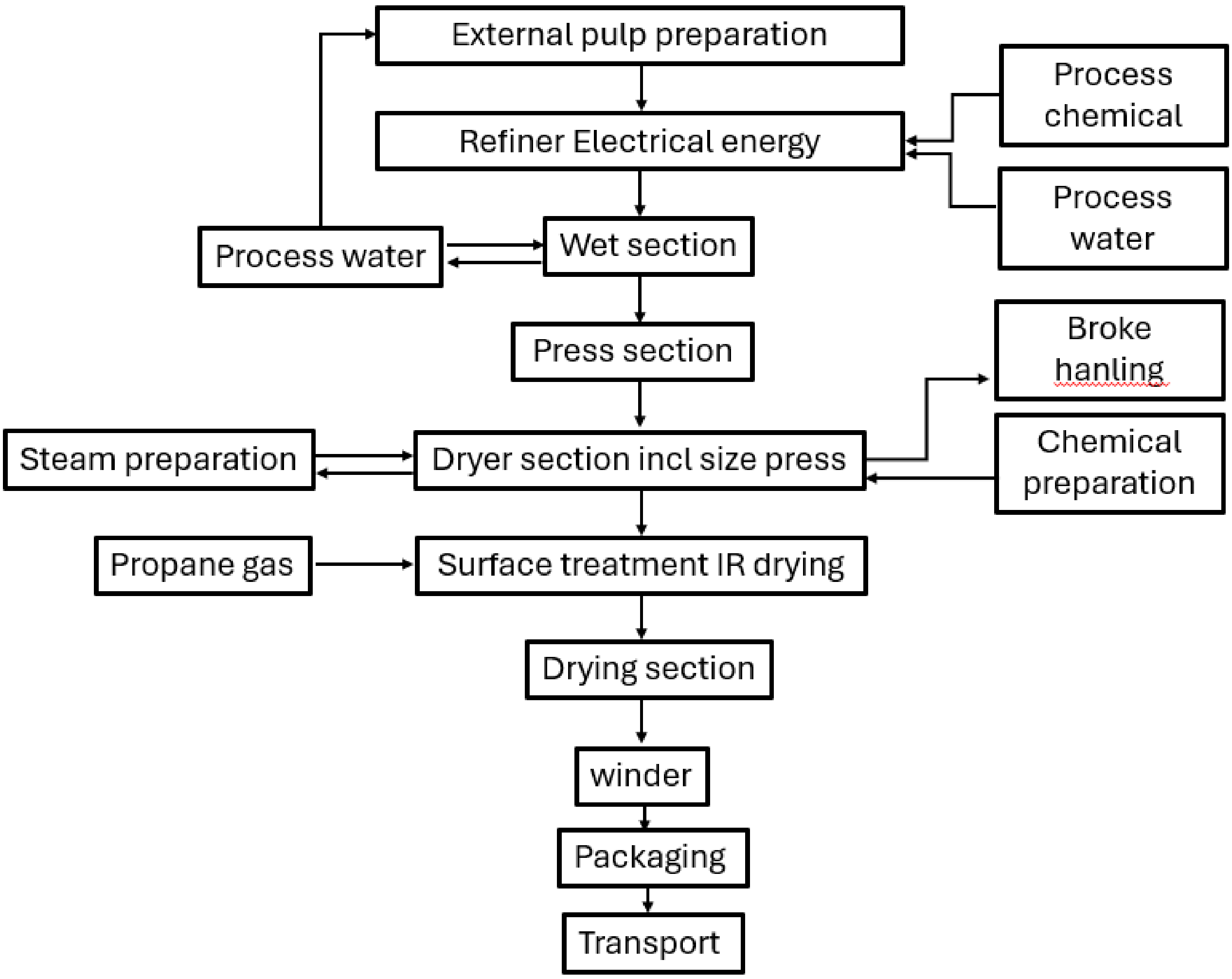


FIGURE 39 FLOW-CHART OF MANUFACTURING PROCESS FOR GREASEPROOF BROWN PAPER, ONE SIDE SILICONE-TREATED.

The manufacturing process diagram (see Figure 45) highlights additional flows to be considered, including the addition of water (process/white/tap) and propane gas. The paper's manufacturing company provided the following material consumptions per ton of paper produced:

- 77 m^3/t (both pulp and paper) of process water (white water)
- 2.4 m^3/t of tap water
- 20 kg/t of propane.

Although requested, no details of wastewater recycling processes were provided. Nordic Paper's website states that "we recycle virtually all our process water. The used process water is purified beforeit is returned to the same watercourse from which it came. We measure the quality of this water regularly to ensure that it does not cause harm to the environment." However, literature research hashighlighted that process water cannot be recycled indefinitely due to the enrichment of ion concentrations over time (Hubbe, 2007).

Therefore, to fully evaluate the environmental impacts of the paper's manufacturing process, the entire amount of white water (77 m^3/t) was included as an input flow in the model of the present LCA study. No details were provided regarding the amount of waste generated and related recycling processes. Nordic Paper's website also mentions that "we use most of the residual products that arise from our production ourselves. We extract the tall oil and sell some to buyers who refine it into new products; the remainder becomes pitch oil, which we then use to produce fossil-free energy. Ash and fiber sludge are sold for landfill." Therefore, in the absence of specific information and given the selling of wastes to third parties, the generation of industrial scraps was not considered in modeling the study.

As specified by the Swedish company, two different sources of energy are used in the paper manufacturing process: electric and thermal/steam energy. However, the information provided by the company was not suitable for implementation in the model (see Annexes), so secondary data were assessed. Data from the ecoinvent database were chosen, including data for both the Swedish market for electricity (medium voltage) and the European market for "heat production, natural gas, at industrial furnace >100kW". This allowed for the consideration of the entire related environmental impacts in the subsequent impact assessment stage, in accordance with average European processes for wood-containing paper production. The final product is coiled on a support made from paperboard.

As stated by Ecopack S.p.A., paper is delivered as reels with the characteristics listed in Table 24.

Component	Characteristics
Grammage of the paper (Gsm)	75
Weight of a single reel (Kg)	316.5
Weight of the paperboard core only (Kg)	1.5
Length of a single reel (m)	5000
Weight of a single reel (m)	0.84

TABLE 29 CHARACTERISTICS OF PAPER REELS AS DELIVERED TO ECOPACK S.P.A.

The data above indicate that the contribution of paperboard cores to the total mass is limited, approximately 0.47% per reel. Additionally, these cores are reused for subsequent processing operations by Ecopack S.p.A. for an unspecified number of cycles. Therefore, their contributions were not considered in this LCA study.

Finally, data regarding the transportation of paper from "Nordic Paper Holding AB" to Ecopack S.p.A. were estimated using an online maps provider, covering 2.200 km. Since Ecopack S.p.A. stated that there are about 20-30 reels per lorry, it was assumed that the transport could be carried out by a EURO4, 16-32 metric ton lorry. This assumption was made because the average transported weight of these vehicles is approximately 5.79 tons, and their Emission Standard category well represents the fleet of freight vehicles within the EU.

The utilized plasticizer-free dispersion adhesive, mainly based on EVA polymer, complies with the general regulations for materials and articles intended to meet food on a European level (EC No 1935/2004).

Since no primary data were provided regarding the glue's formulation, the ecoinvent dataset related to the average European production of solid ethylene vinyl acetate copolymer was utilized. This dataset includes raw materials, chemicals, transport to the manufacturing plant, estimated emissions to air and water from production, estimation of energy demands, and infrastructure of the plant.

The adhesive is supplied to Ecopack S.p.A. in 1000 kg tanks, assumed to be transported by a 16-32 metric ton lorry for an estimated distance of 230 km. Like the estimations made for the paper, the dataset related to EURO 4 vehicles was selected from the ecoinvent database. Contributions related to the manufacturing of tanks were not considered.

Lamination, cutting, shaping pies, and repacking processes are based on primary data provided by Ecopack S.p.A. and are derived from industrial records of 2021.

The primary data related to the production of the pie mold 8018030B were provided by Ecopack S.p.A.and are based on industrial records of 2021. This section will describe the available data and assumptions, in relation to each processing phase.

Component	Amount
Greaseproof brown paper	2 layers
Glue dry content	20 gsm
Tap water	20 gsm
Product	Reels of laminated paper
Waste	20 m for reel
Hourly production	2160 m
Energy requirements for production	21 kWh
Energy requirements for paper drying	28-30 kWh

TABLE 30 PRIMARY DATA RELATED TO THE LAMINATION PHASE PERFORMED BY ECOPACK S.P.A.

In detail, the input flows considered include the raw materials required, including their transportation to the factory gate, as well as the energy consumed in the process. This energy consumption consists of two components: the average consumption of production lines and the consumption of static furnaces for paper drying. Ecopack Company specified that the drying step is not always performed to facilitate following processes, so its consumption per hour could be estimated as 10% of the total energy required for paper drying. To estimate all these energy requirements without specifics of the utilized energy mix, average data related to the Italian market for electricity were considered. Due to the minimal amount of material wasted for each utilized reel of paper (0.4%), it was decided not to consider its contribution as a waste flow for this processing phase.

The "cutting pies" process begins with cutting a reel of laminated paper into three smaller reels, each with a width of 0.28 m. These reels are then further cut into squares measuring 0.29 m * 0.28 m. These squares undergo shaping in the subsequent phase. Ecopack S.p.A. reported that approximately 15% of material is generated as waste for a single pie. These industrial scraps are collected and recycled as paper-based items by external companies. Therefore, following the "cut-off" approach, their contributions were fully allocated to the primary user (Ecopack S.p.A.).

The energy requirements were evaluated based on average data related to the Italian market for electricity.

Component	Amount
Laminated greaseprood brown paper (kg weight)	Dimension 0.29 m *0.28 m0.0138 kg weight
Product	Shaped pie mould
Waste (% for FU)	15%
Hourly production (FU for hour)	1742
Energy requirements (kWh)	17

TABLE 31 PRIMARY DATA RELATED TO THE PHASE OF PIE-SHAPING PERFORMED BY ECOPACK S.P.A.

In the final production step, the input flows consist of the finished product and the materials used for packaging. Specifically, these materials include paper-based boxes with LDPE inner bags. To incorporate inventory data related to the packaging materials, average datasets from the Eco invent database were utilized, representing the European market for corrugated board boxes and the global market for LDPE inner bags. The selection of the latter dataset was due to a lack of more detailed information. Energy requirements, emissions, and wastes were already included in the selected datasets.

According to information provided by Ecopack S.p.A., to account for the contributions associated with the transportation of these materials, distances of 35 km for boxes and 80 km for LDPE bags were estimated using an online maps provider. Subsequently, data from the ecoinvent database related to EURO 4 vehicles were considered, aligning with previous estimations.

Component	Amount (One for box)
Corrugate board box (kg ca)	0.75 -1.2 kg ca
LDPE inner bag (gr)	6.92 g
Pie 8018030B	900 FU

TABLE 32 PRIMARY DATA RELATED TO THE PACKAGING OF FINISHED PRODUCTS.

Life cycle inventory analysis scenario A & B

For both scenarios, transportation contributions were considered using secondary data related to Euro 4 trucks weighing 3.5 - 7.5 tons. Transportation contributions were calculated for a theoretical distance of 40 km, utilizing secondary data related to Euro 4 trucks weighing 3.5 - 7.5 tons. Scenario A involved estimating waste disposal contributions using average data from the database. The industrial composting process used as an example is comparable to European facilities.

With secondary data, it is possible to represent the respective impacts due to industrial composting facilities.

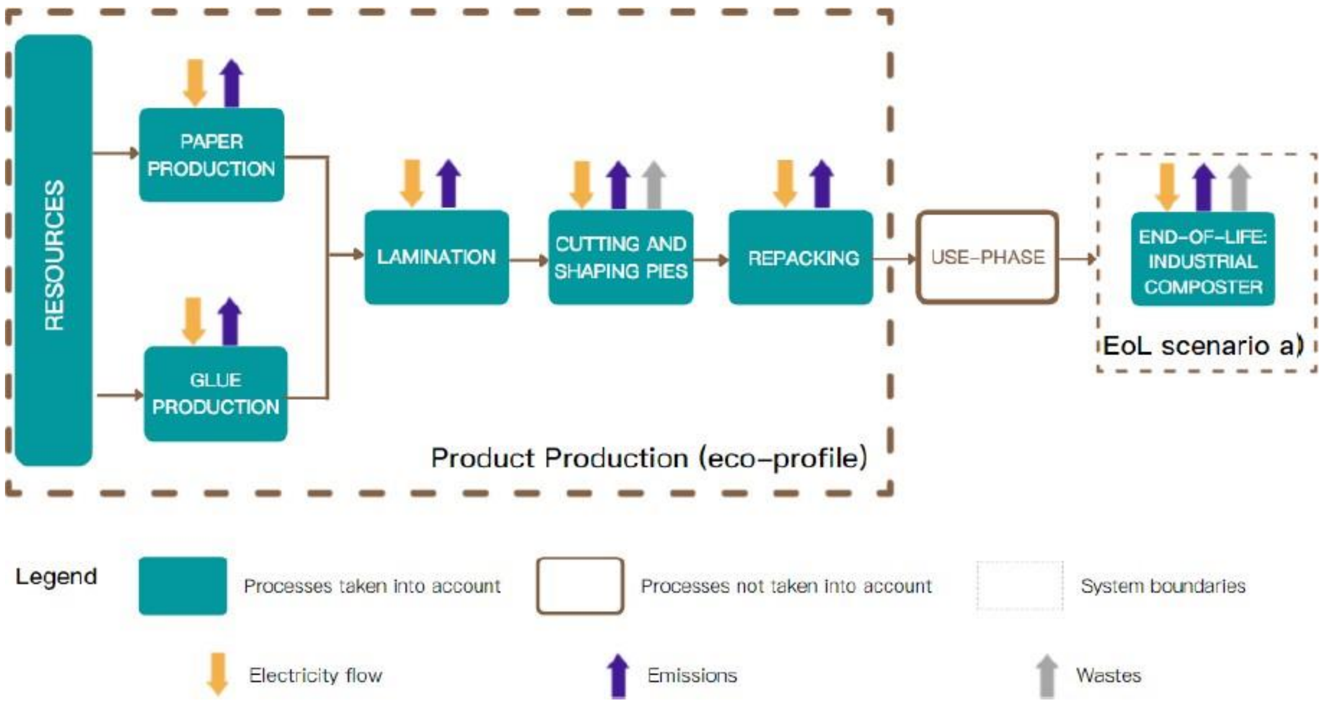


FIGURE 40 SCHEMATIZATION OF THE STUDIED SYSTEM WITH THE UNDERLINING OF THE INPUT AND OUTPUT FLOWS CONSIDERED, CASE EOL

No certified data regarding the compostability of the pie mold 8018030B were provided. Consequently, to estimate the contributions related to waste disposal in industrial composters, average data from the ecoinvent database were utilized. Specifically, the industrial composting process was chosen, with values referencing the situation in Switzerland, which is comparable to other European facilities. Although these secondary data do not explicitly pertain to biodegradable wastepaper, they adequately represent the impacts of standard industrial composting facilities. Output flows associated with this process include the production of municipal solid waste (1.85E-5 kg per 1 kg of biowaste) and average wastewater (2.25E-4 m3 per 1 kg of biowaste). Additionally, further treatments for co-produced municipal solid waste include open dumping, municipal incineration with fly ash extraction, and open burning. Wastewater undergoes additional processing in plants with varying treatment capacities.

Scenario B encompassed all input and output flows related to the paper recycling process, including contributions from cellulose washing, internal treatment of incoming and outgoing wastewater, as well as waste generated such as waste and mixtures of wood ashes. Additionally, by-products were considered following the principle of the polluter pays present in the LCA study.

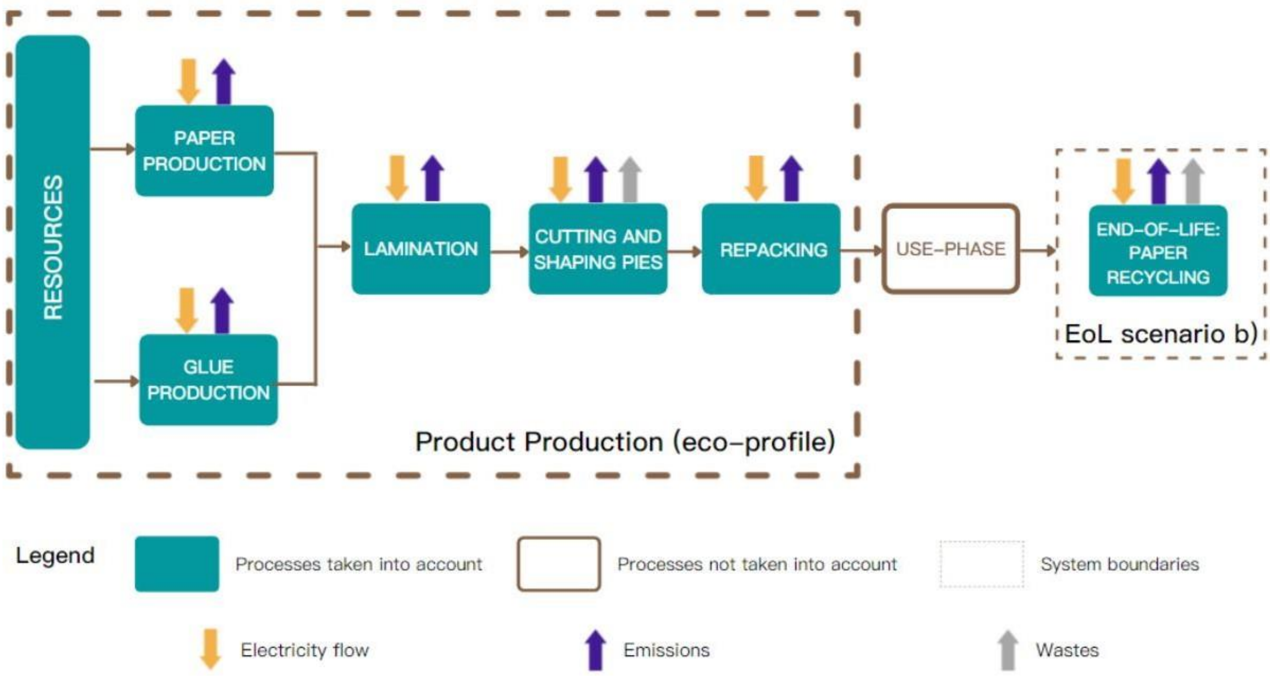


FIGURE 41 SCHEMATIZATION OF THE STUDIED SYSTEM WITH THE UNDERLINING OF THE INPUT AND OUTPUT FLOWS CONSIDERED CASE EOL

In this scenario, the product is disposed of as unsorted wastepaper and transported to urban paper mills for recycled paper production. Due to the lack of plant specifics, secondary data on the average technology used in Europe for 100% graphic paper production were considered. This module encompasses all contributions related to pulp deinking, paper production, on-site energy production, internal wastewater treatment, and auxiliary transport to the paper mill. Output flows include hazardous waste for incineration, municipal solid waste production (0.0138 kg per 1 kg of recycled paper), unpolluted wastewater (0.0106 m3 per 1 kg of recycled paper), and wood ash mixture (0.00685 kg per 1 kg of paper). Further treatments of these by-products were also accounted for, aligning with the "polluter pays" principle adopted for the LCA study.

Impact Assessment

Based on the previous data and assumptions, the corresponding impacts and subsequent analysis are presented.

The study of the eco-profile will allow us to understand which aspects need improvement, as well as the approaches related to the previously described project case.

This will enable us to propose products with a high environmental sustainability score on the market.

Below are the aspects that have been considered.

Impact category	Reference unit	Result
Acidification	Kg SO_2 eq	0.00007
Climate change	Kg CO_2 eq	0.03198
Ecotoxicity freshwater	KG 1.4 DCB eq	0.00955
Ecotoxicity marine	KG 1.4 DCB eq	25.20585
Ecotoxicity terrestrial	KG 1.4 DCB eq	0.00014
Energy resources non renewable	MJ	0.49221
eutrophication	Kg PO_4 eq	0.00004
Human toxicity	Kg 1.4 -DCB eq	0.01763
Material resources metals, minerals	Kg Sb eq	$0.122*10^{-6}$
Ozone layer depletion	Kg CFC -11 -Eq	$0.450*10^{-8}$
Photochemical oxidation	Kg ethylene Eq	$0.476*10^{-5}$

TABLE 33 ENVIRONMENTAL IMPACTS GENERATED TO PRODUCE A SINGLE FU (1 ITEM OF ROUND PIE MOLD, TYPE 8018030B)

The results indicate that the impact categories mentioned above have negligible effects compared to others and will not be further examined. As these impact categories result from the aggregation of multiple flows, additional investigations were conducted to identify hotspots in the product's manufacturing processes. Specifically, a graph illustrating the relative contribution of each processing phase to every impact category is presented Table 28.

A more detailed analysis of the contribution of each process (raw materials, energy requirements, transportation, transformation processes, etc.) for each impact category will be discussed below.

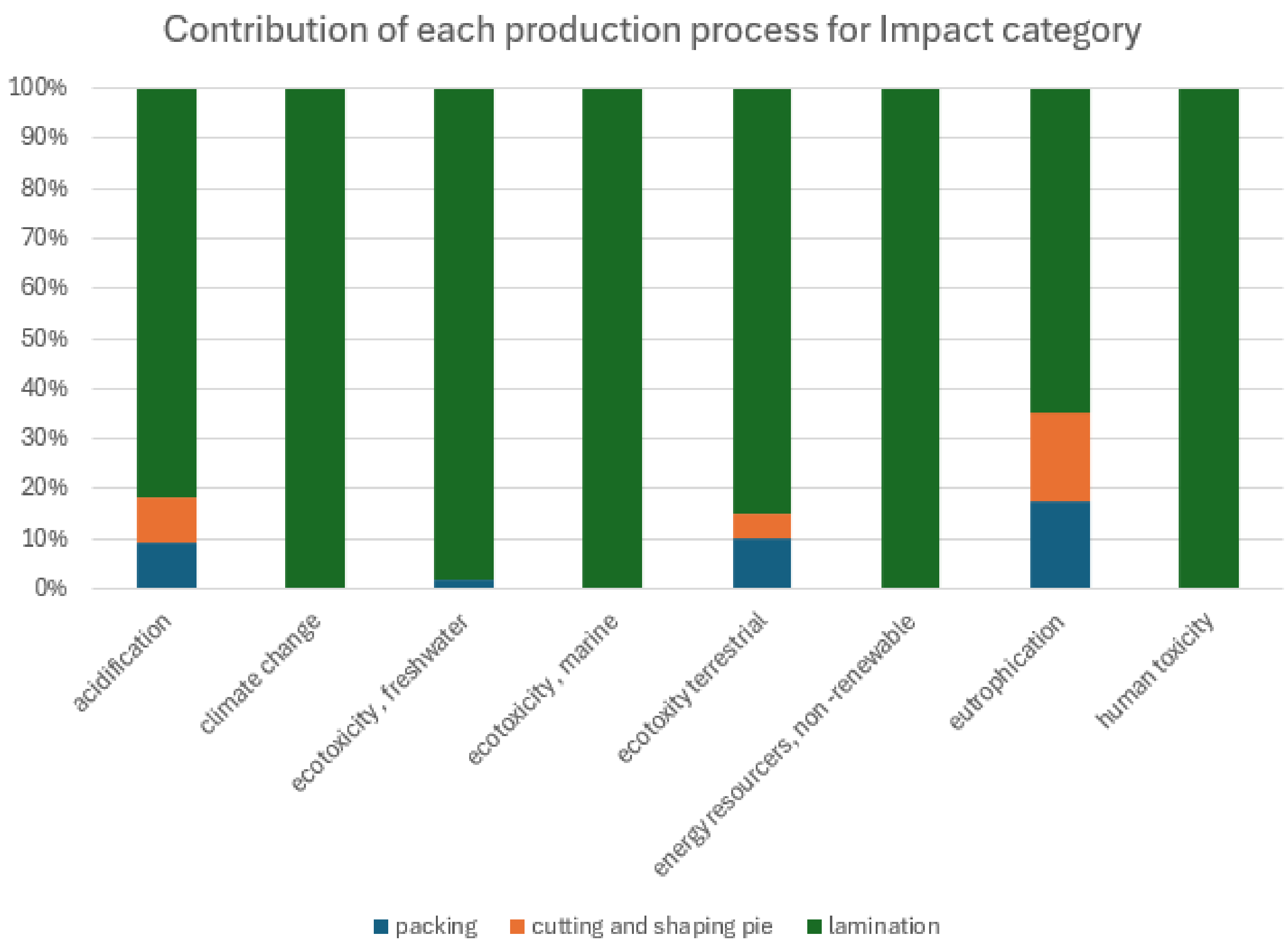


FIGURE 42 RELATIVE CONTRIBUTION OF EACH PROCESSING PHASE FOR IMPACT CATEGORY, CASE ECO-PROFILE.

The results of the last three impact categories listed above can be neglected due to their smaller magnitude compared to the others and will not be further assessed.

The highest contribution is always attributed to the lamination phase, as this process encompasses all the impacts from the production and transportation of raw materials. Paper fabrication involves numerous processes, including the production and transportation of chemicals, energy production, electricity from the grid, waste treatment, and wastewater treatment plants.

The most significant human contributions to Global Warming (GW) are the combustion of fossil fuels, which can lead to increased global average temperatures and abrupt regional climate changes. Specifically, a substance is considered a contributor to GW if it absorbs infrared radiation and remains stable in the atmosphere for decades to centuries. The characterization factors for this impact category are expressed as Global Warming Potential over different time horizons, with 100 years (GWP100) being the most common, and the reference unit is kg CO_2 equivalent.

The top 5 direct contributions to this impact category are shown in Figure 49. The highest value is attributed to the production of heat required for the paper manufacturing process. The contribution of raw material transportation to Ecopack S.p.A. is about half of the previous process but still significant. The values of the remaining three top contributors are comparable, linked to the production of electricity and steam required for paper manufacturing, as well as the sulfate pulp production itself (green bar). The highest contribution is caused by the "1_Lamination" process performed by Ecopack S.p.A. and, more precisely, by "Paper production" (61.07% of impacts). This is due to the CO_2 emissions from the thermal/electricity requirements of the process performed by the Swedish manufacturing company (44.76%), the most relevant contributions to this impact category.

Acidification can be caused by pollutant emissions into the air, water, and soil, mainly from combustion processes in electricity and heat production, and in transport systems. Specifically, these emissions consist of acidic gases (e.g., sulfur oxides, ammonia, nitrogen oxides) that react with water in the atmosphere to form "acid rain." The acidification potential is expressed using the reference unit of kg sulfur dioxide equivalent (SO_2 eq.) and accounts only for acidification caused by SO_2 and NOx. The highest value is linked to the production of sulfate pulp for paper manufacturing, followed by the transport category.

The "Lamination" process contributed the most to acidification, precisely accounting for 90.82% of the total impacts. This is due to sulfur dioxide emissions associated with paper production (58.45% of impacts) and significant impacts caused by the transportation of raw materials to Ecopack S.p.A., as previously highlighted for the top 5 direct contributions.

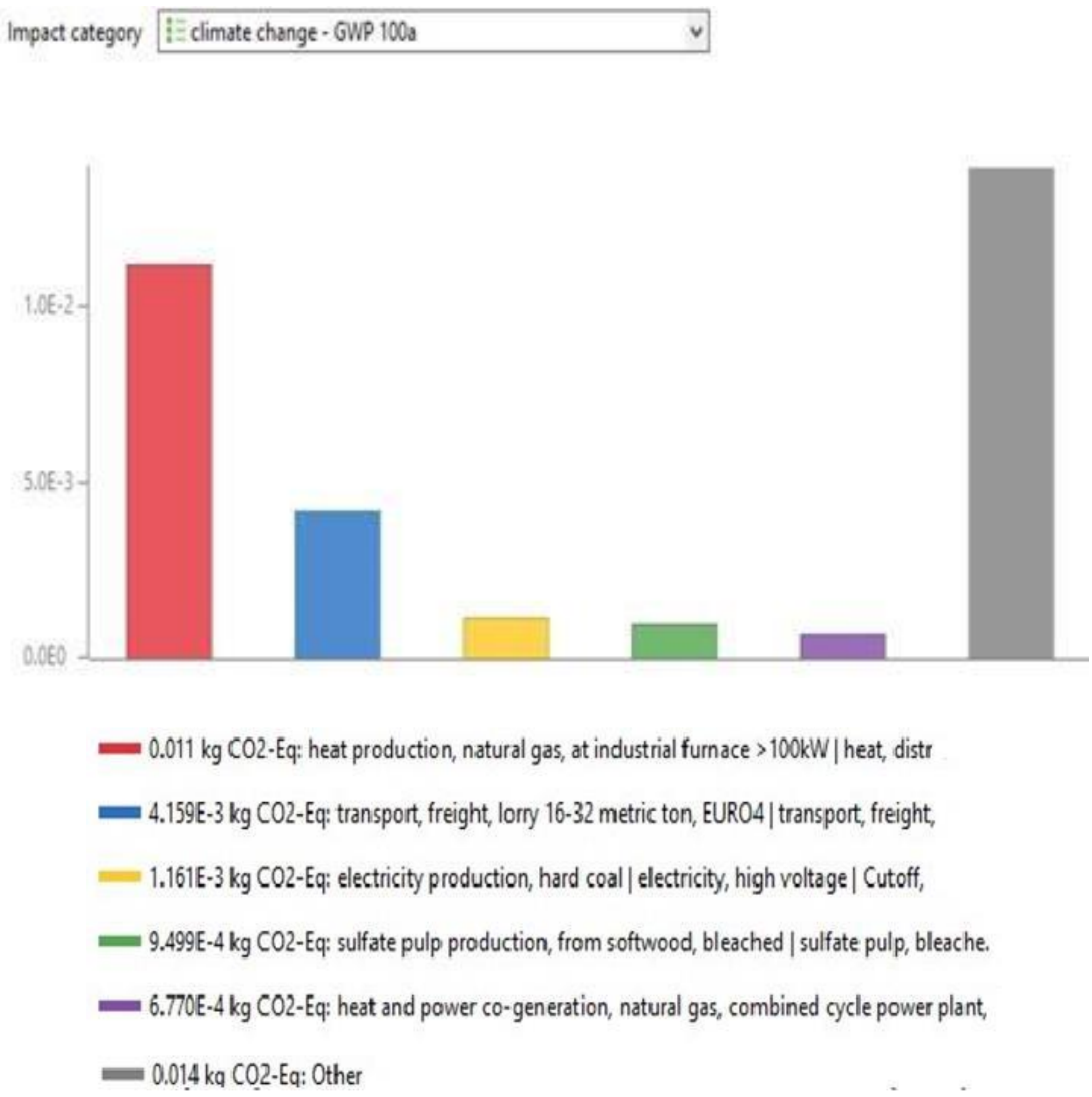


FIGURE 43 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "CLIMATE CHANGE", CASE ECO-PROFILE.

Eutrophying substances are evaluated for both aquatic and terrestrial ecosystems. Specifically, the method assesses the natural production of biological material (BOD or COD) per input of nutrient emissions transported by air and water (N and P). This enrichment of terrestrial ecosystems with the macronutrients N and P is expressed as equivalents of phosphate anions (PO_4^{-3}- eq.). Among the top processes with the highest direct contributions, those of "sulfate pulp production" (red bar) and "treatment of spoil from lignite mining" (blue bar) have comparable values. Specifically, the latter considers specific emissions from waste leachate of lignite related to electricity production processes, hence short- and long-term emissions to groundwater from rainwater infiltration leaching, with nitrates as direct impact contributions. It's interesting to note that the treatment (surface landfill) of green liquor dregs from pulp production - including base sealing and leachate collection system - has the lowest contribution (violet bar) among the processes shown in the figure 50.

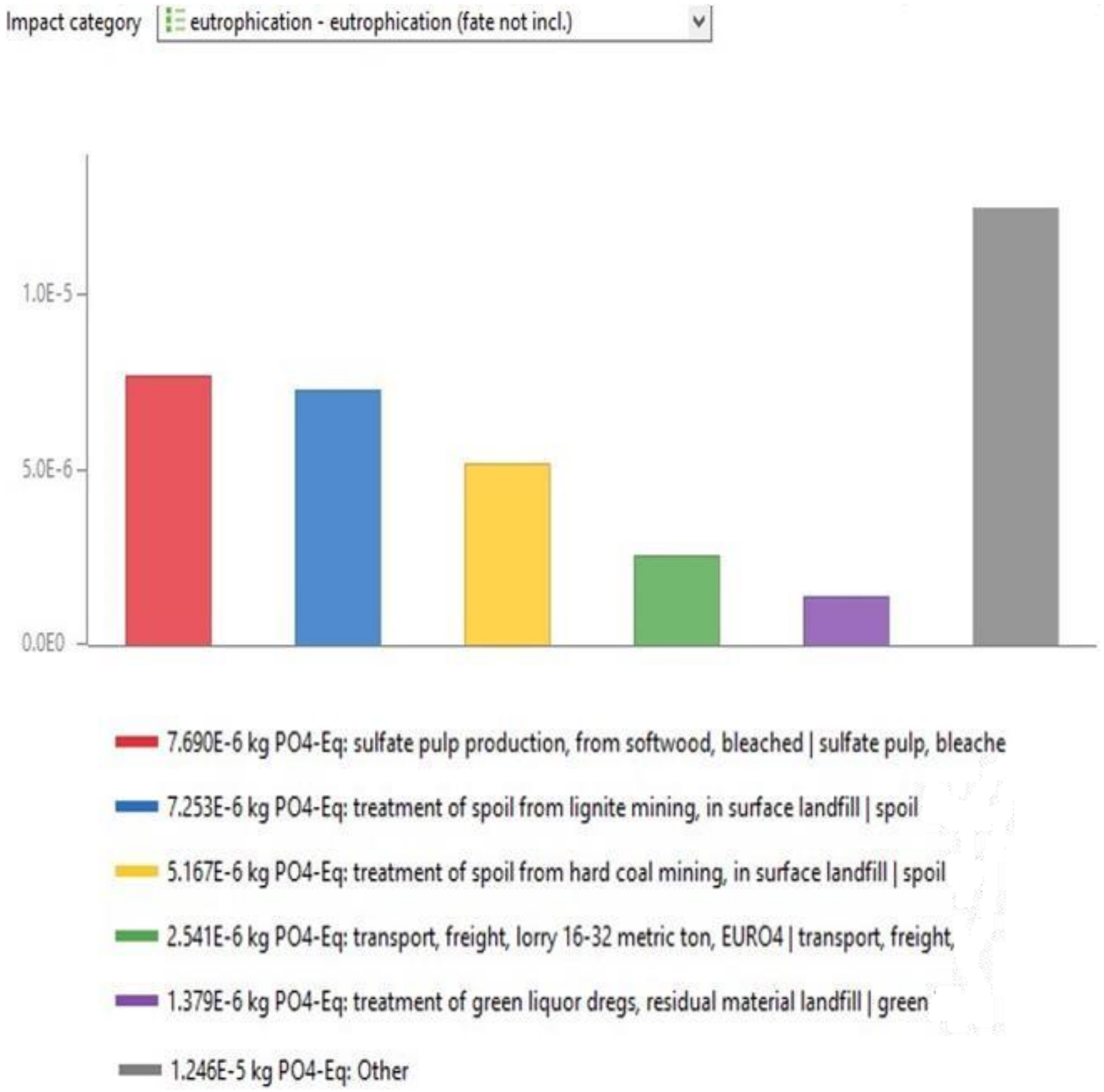


FIGURE 44 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "EUTROPHICATION", CASE ECO-PROFILE.

For the current environmental profile, the entire contribution to impacts caused by paper production represents approximately 63% of the total. This is because many upstream processes influence the environmental burdens associated with sulfate pulp production, such as chemical manufacturing, along with their respective emissions.

The category "Energy resources: non-renewable (ADP for fossil fuels)" is related to non-renewable resources of fossil fuels. Elementary flows are expressed in MJ, and the highest value is attributed to offshore production of oil and gas (see figure 51 and red bar). Data related to oil and gas production are comparable, whether it is crude oil production in the Middle East.

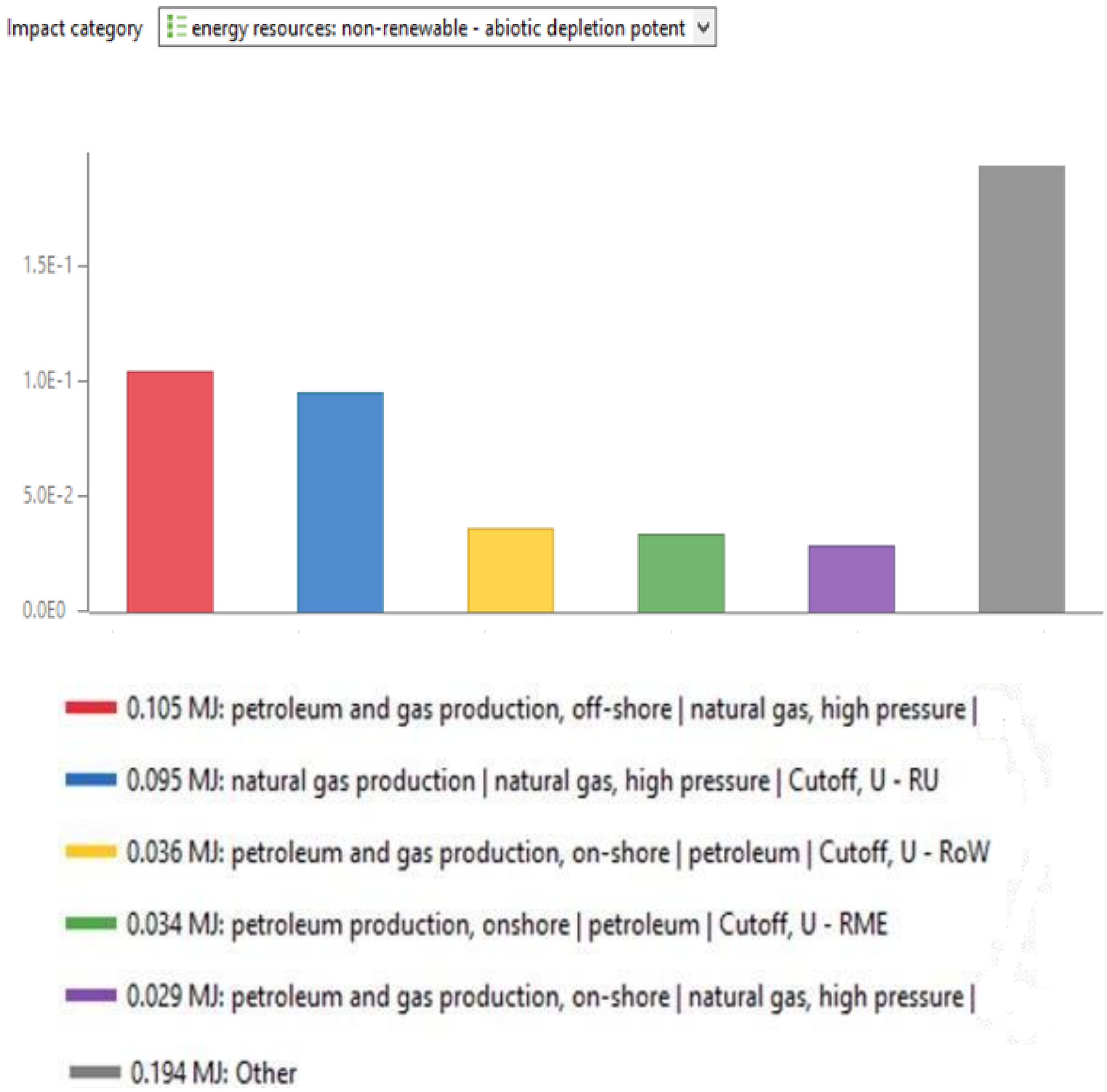


FIGURE 45 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "ENERGY RESOURCES: NONRENEWABLE", CASE ECO-PROFILE.

The process that has the greatest contribution within this Impact category is once again the upstream process of paper production (62.71%), due to its high energy requirements, mainly thermal. A contribution as high as 17.15% is attributed to raw materials transportation. Overall, the results of this Impact category are the second highest recorded in the current LCA study.

Precisely, the ecotoxicity potential for freshwater is primarily divided between two processes: chemical production (specifically H_2O_2) and the treatment of waste generated in the process.

The main contributors to both these processes are copper (31%), vanadium (25%), and nickel (18%).

The five processes that have the major direct contributions to this impact category are shown in the image below. The highest value is linked to the "treatment of scrap copper" in municipal incineration. This aligns with the calculation of impacts within this category and could also be related to the widespread use of copper in the electricity grid. Other significant contributions are attributed to the treatment of waste from processes linked to energy and raw materials production. We focused on the impact category, climate change, and its direct contributions are represented below.

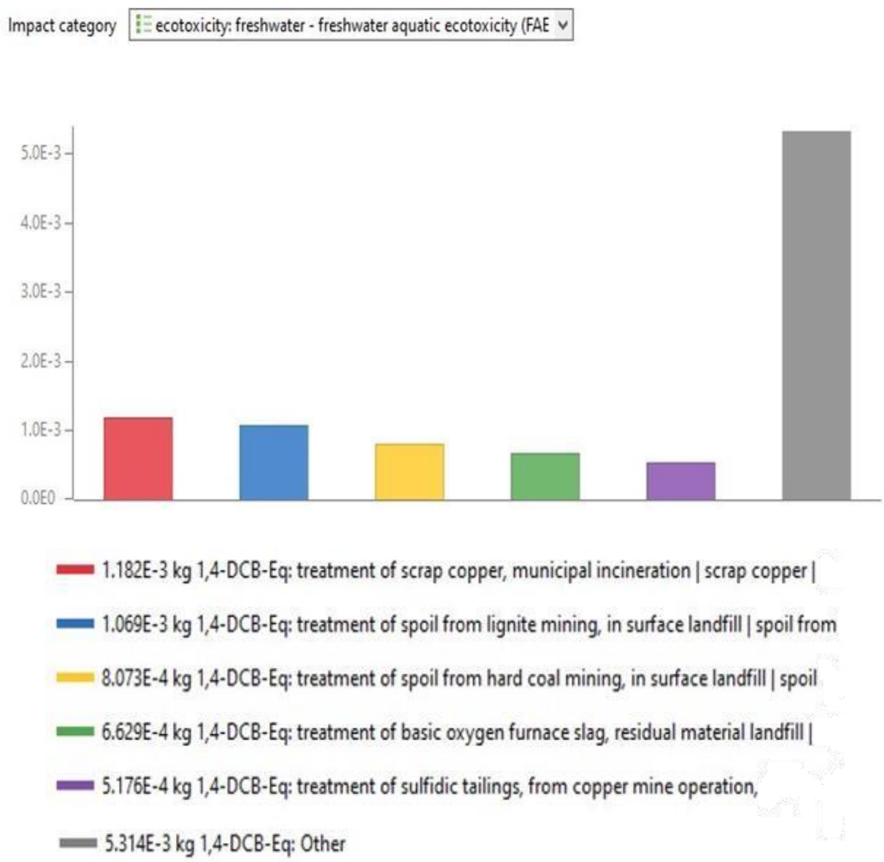


FIGURE 46 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "FRESHWATER ECOTOXICITY", CASE ECO-PROFILE.

Regarding the overall contribution to this Impact category, the highest value is attributed to the "1_Lamination" process (73.21%) due to upstream impacts related to paper production (58.51%). Specifically, the major impacts for this latter process are attributed to sulfate pulp production and the associated energy requirements. In this impact category, the "1_Lamination" process contributes the most due to the polluting emissions associated with paper production. Another significant contribution

(26.49%) is represented by the production of corrugated cardboard boxes used for packaging the final products.

Among the five processes that have the major direct contributions to Ecotoxicity: terrestrial, the construction of the electricity network and direct emissions from landfarming applications of wood ash mixture have comparable values. This stems from flows related to freight transport carried out by unspecified lorries, as it involves the production of untreated wood waste.

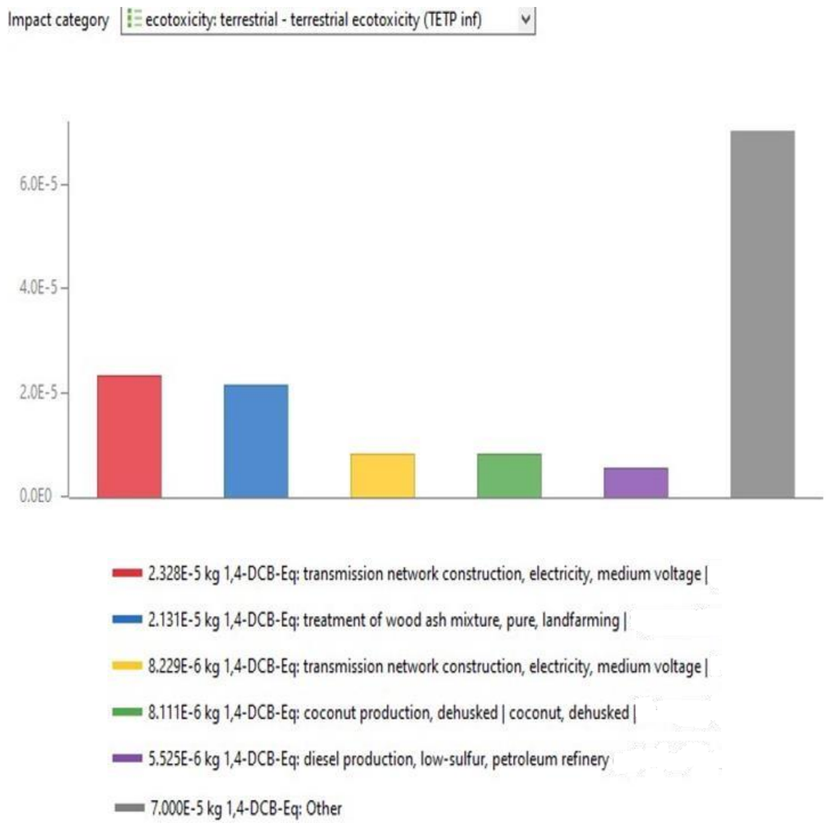


FIGURE 47 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "ECOTOXICITY: TERRESTRIAL", CASE ECO-PROFILE.

The highest contribution to terrestrial ecotoxicity is associated with paper production (66.04%). In contrast, environmental loads related to transportation contribute around 12%. Generally, the effects of chemicals on terrestrial ecosystems are lower compared to aquatic ecosystems. This could be related to the nature of pollutants emitted during the most impactful phase of product manufacturing, specifically the Lamination phase and more precisely paper production.

Among the five processes that have the major direct contributions for this impact category, the construction of the electricity network and direct emissions from landfarming applications of wood ash mixture have comparable values. This stems from flows related to freight transport carried out by unspecified lorries, as it involves the production of untreated wood waste.

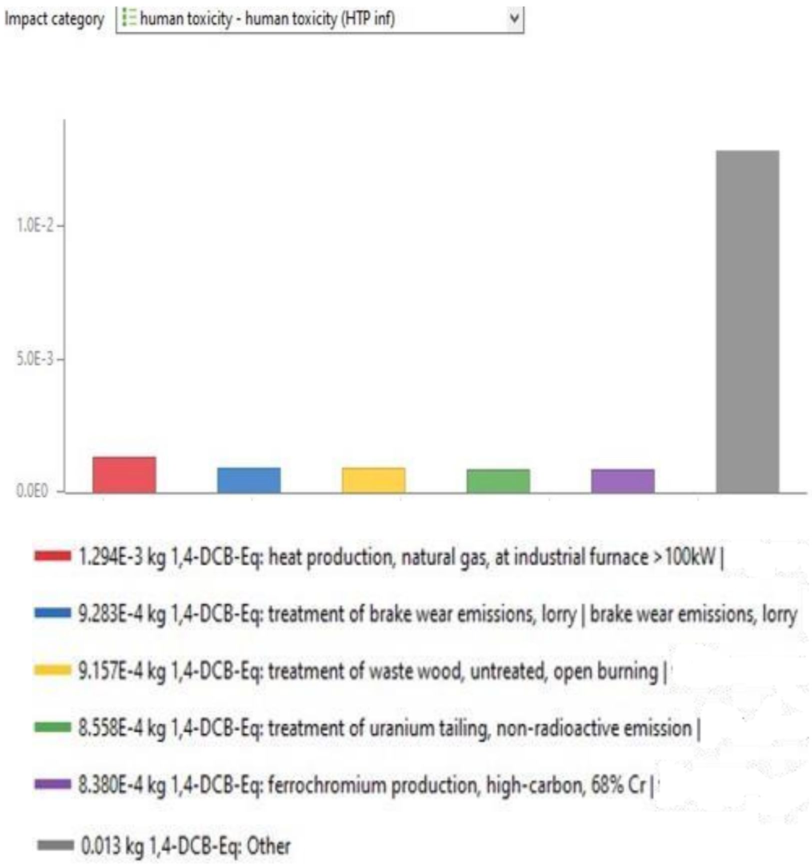


FIGURE 48 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "HUMAN TOXICITY", CASE ECO-PROFILE.

Given the characteristics of the impact factors used, the main contributions derive from heat production associated with the high thermal energy requirements of the paper manufacturing process and waste treatments related to transportation.

End of life scenario A) waste-disposal in Industrial Composters

In detail, the production of 1 kg of biowaste from 1 kg of finished product was used as a reference. In the absence of certified information regarding the product's compostability, this assumption represents an appropriate baseline for the hypothesized end-of-life (EoL) scenario. At first glance, the five highest results are associated, in decreasing order, with the impact categories of marine aquatic ecotoxicity, energy resources, climate change, human toxicity, and freshwater ecotoxicity. These results were entirely obtained based on secondary data, so they should be considered as a generic overview of impacts related to industrial composting treatment. The most relevant impact categories related to the top five results will be further analyzed below. The five highest results are associated, in decreasing order, with the impact categories of marine aquatic ecotoxicity, energy resources, climate change, human toxicity, and freshwater ecotoxicity. These results were entirely obtained based on secondary data, so they should be considered as a generic overview of impacts related to industrial composting treatment. The most relevant impact categories related to the top five results will be further analyzed.

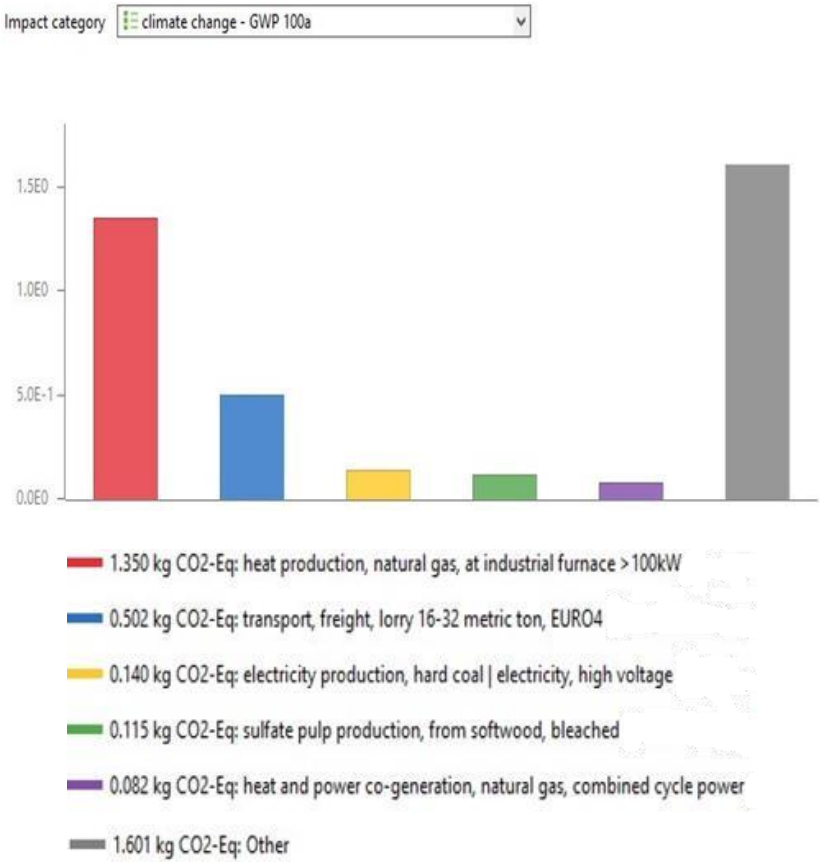


FIGURE 49 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "CLIMATE CHANGE", CASE EOL SCENARIO A)

The top 5 direct contributions to this Impact category are associated with the production phases of 1 kg of finished product to be composted. Specifically, the production of petroleum and natural gas from Norway (red bar) and high-pressure natural gas in Russia (blue bar) have comparable values and are the highest contributions. Petroleum and gas production from other locations follow with similar contributions. This means that industrial composting of pies does not impact this Impact category, like production processes.

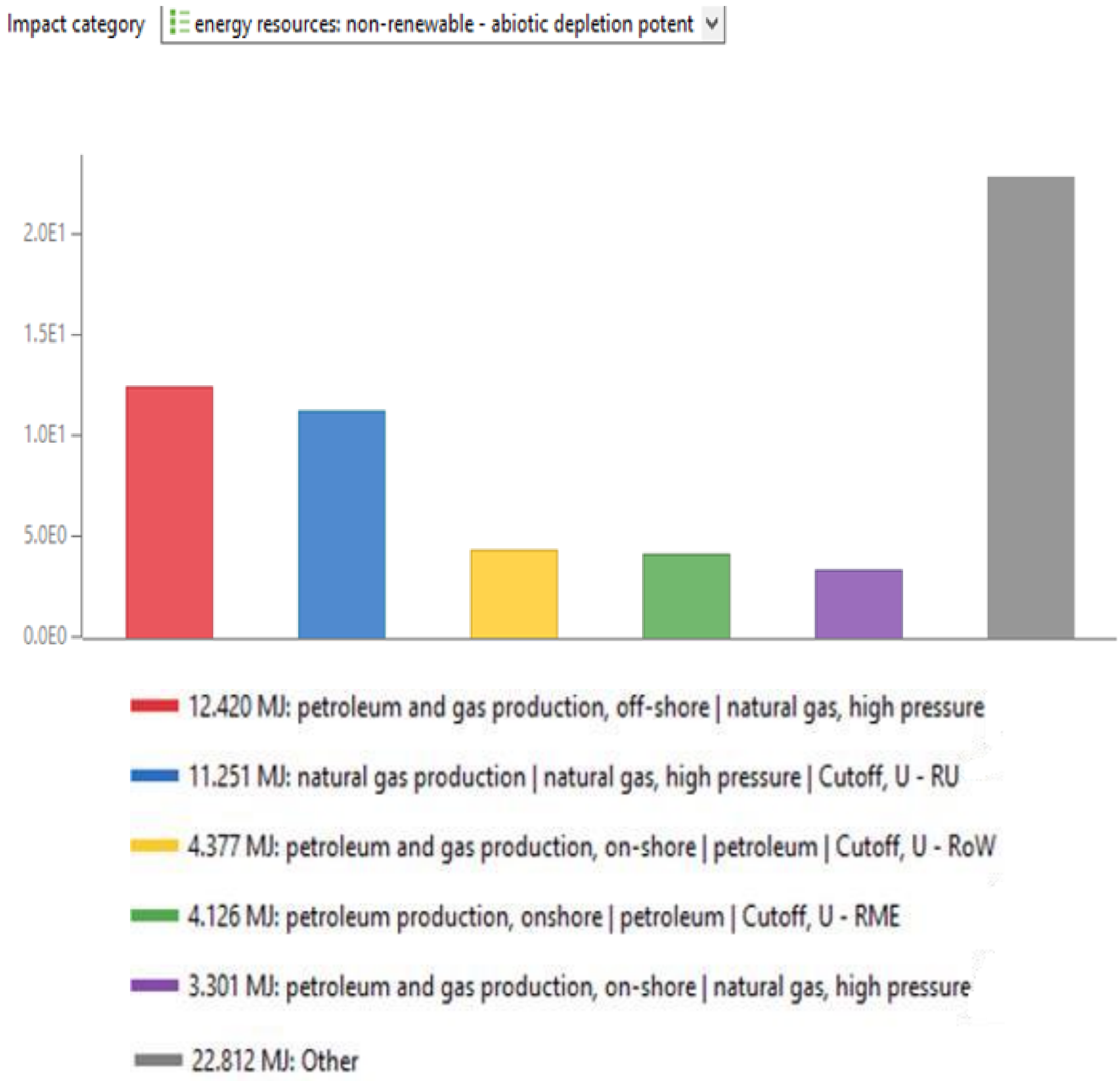


FIGURE 50 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "ENERGY RESOURCES: NON-RENEWABLE", CASE EOL SCENARIO A).

The results recorded for this impact category are lower. This could be linked to the short-term residence of polluting substances in freshwater ecosystems compared to marine ones. The top 5 direct contributions within the "freshwater ecotoxicity" impact category are attributed to municipal solid waste treatments. Specifically, the "treatment of basic oxygen furnace slag" (green bar) is linked to the production of cement to be used in unspecified landfills. The "treatment of sulfidic tailings" (violet bar) involves the open burning of municipal solid waste.

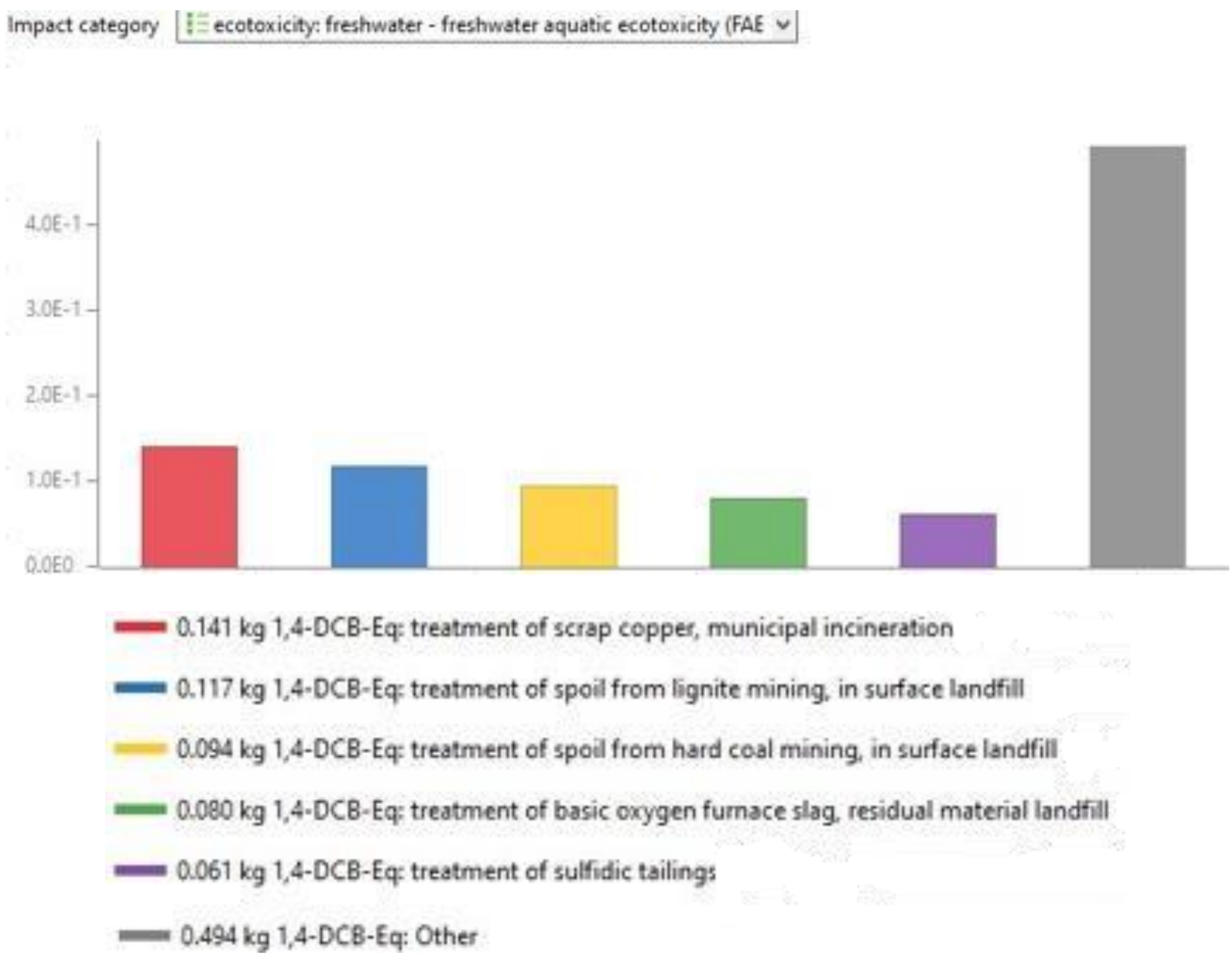


FIGURE 51 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "ECOTOXICITY: FRESHWATER", CASE EOL SCENARIO A).

The five processes that have the major direct contributions to this impact category are shown in Figure 58. The highest value is linked to the "surface landfill treatment of spoil from lignite mining," thus to long-term emissions from rainwater infiltration leaching, with nitrates as direct impact contributions.

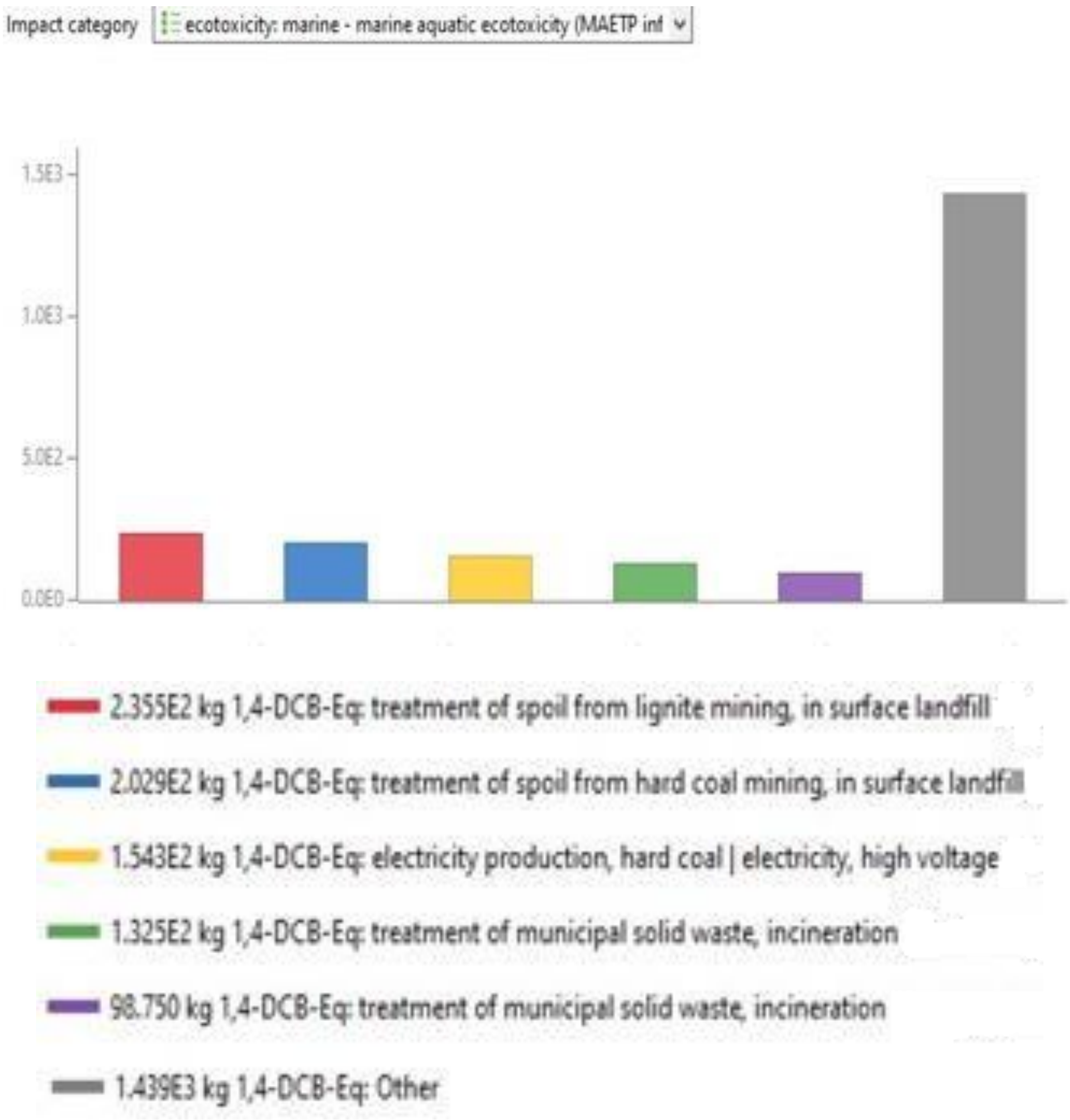


FIGURE 52 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "ECOTOXICITY: MARINE AQUATIC", CASE EOL SCENARIO A).

Emissions resulting from the heat production necessary for manufacturing processes represent the highest direct contribution to this impact category. All the top 5 direct contributions to the results of this impact category have similar values and are also attributed to municipal solid waste treatments. Specifically, the yellow bar represents municipal waste incineration without any pollution control, suitable for informal recycling, while treatments of uranium tailings (green bar) are associated with waste treatment facilities themselves (below figure 59).

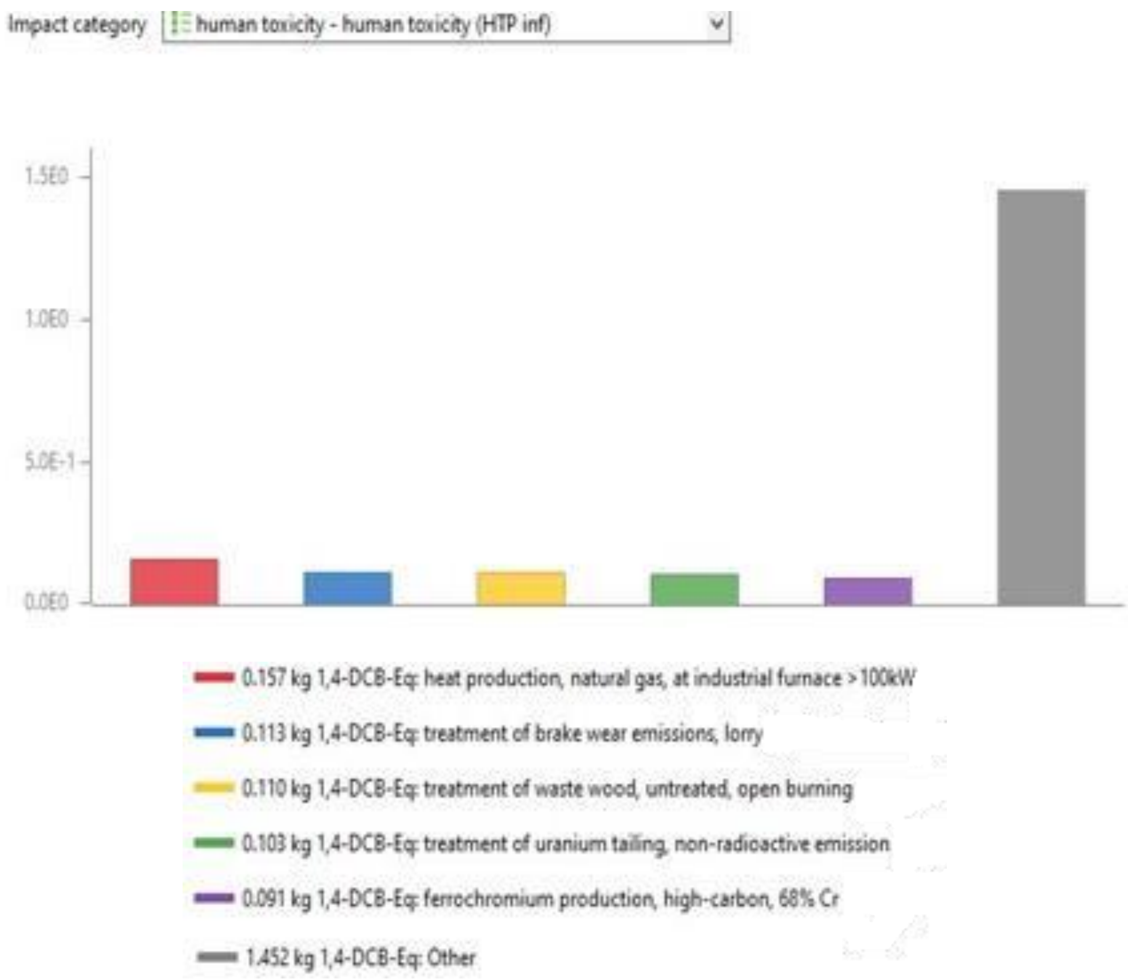


FIGURE 53 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "HUMAN TOXICITY", CASE EOL SCENARIO A).

End-of-Life scenario B) waste-valorization in processing for paper recycling.

For the scenario we are currently examining, the top five results are associated, in descending order, with the impact categories of marine aquatic ecotoxicity, energy resources, climate change, freshwater ecotoxicity, and human toxicity. The mentioned order differs from that of the ecological profile and End-of-Life scenario A) for the last two impact categories. The highest contribution for this impact category is the production of natural gas required for manufacturing processes, followed by raw material transportation. Interestingly, the lowest value is attributed to "production of quicklime, in pieces," a material that could be used as a process material in chemical industries and for wastewater treatment. This contribution is closely related to waste valorization in paper recycling processes.

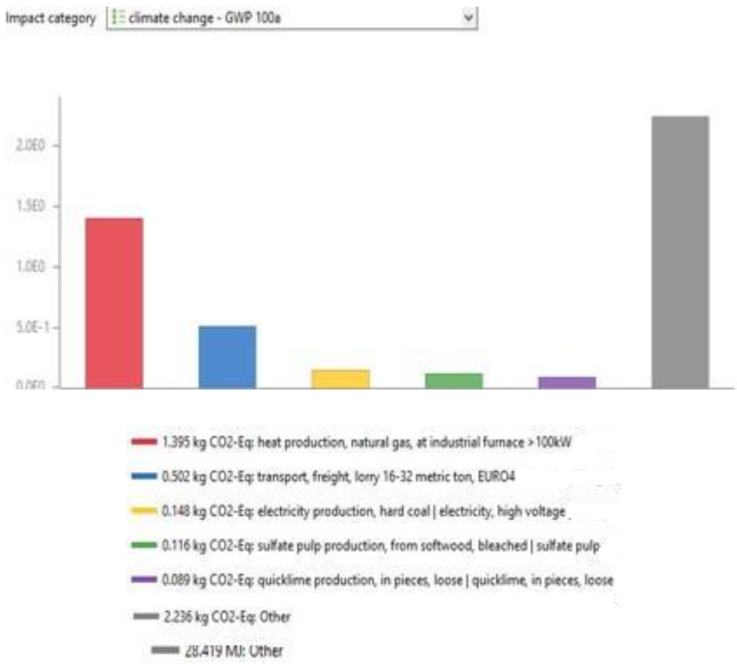


FIGURE 54 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "CLIMATE CHANGE", CASE EOL SCENARIO B).

All five top direct contributions to this impact category are linked to the production of petroleum and gas, primarily required by manufacturing phases but also by municipal solid waste treatments.

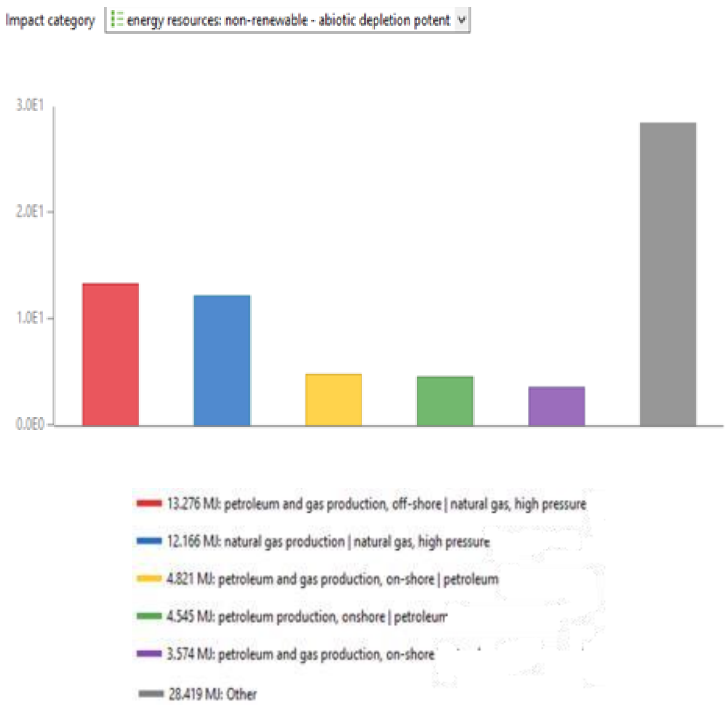


FIGURE 55 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "ENERGY RESOURCES: NON-RENEWABLE", CASE EOL.

The contribution of "lignite mining spoil treatment" is about an order of magnitude higher than those related to municipal solid waste. However, "hard coal mining spoil treatment" (yellow bar) is not among the top 5 contributions for the "marine-aquatic ecotoxicity" impact category. Overall, the results recorded for "freshwater ecotoxicity" in this end-of-life scenario are higher than those of industrial composting. This is directly linked to further treatments of co-products as well as the utilization of raw materials to produce recycled paper. Below are the results.

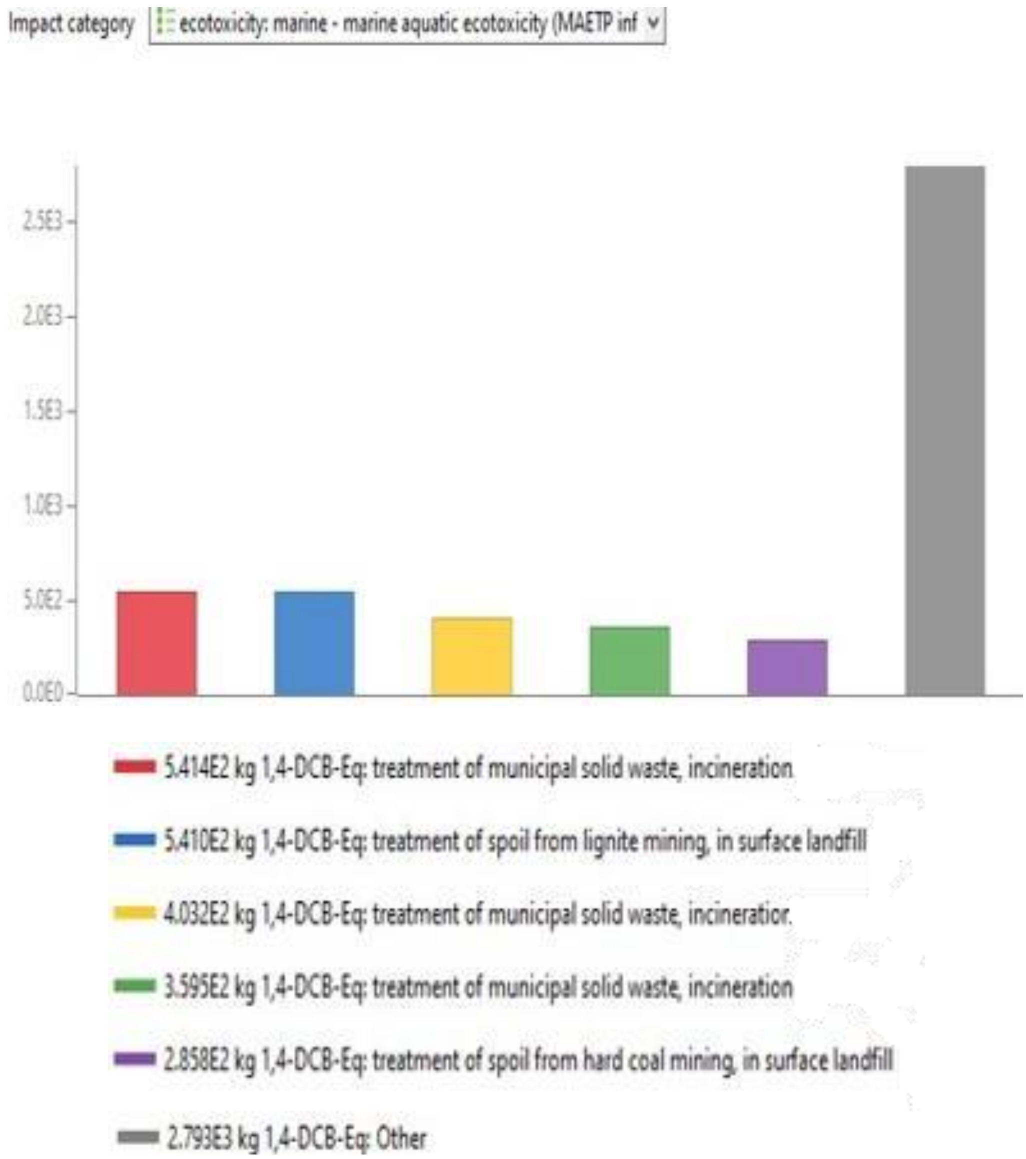


FIGURE 56 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "ECOTOXICITY: MARINE AQUATIC", CASE EOL SCENARIO.

For the impact category of human toxicity, it represents the fifth highest result. The main contributions are related to activities connected to waste treatments (blue and yellow bars), but also to transportation and energy production required by manufacturing phases. Interestingly, the production of ferrochromium is considered, a material used for constructing treatment plants (green bar).

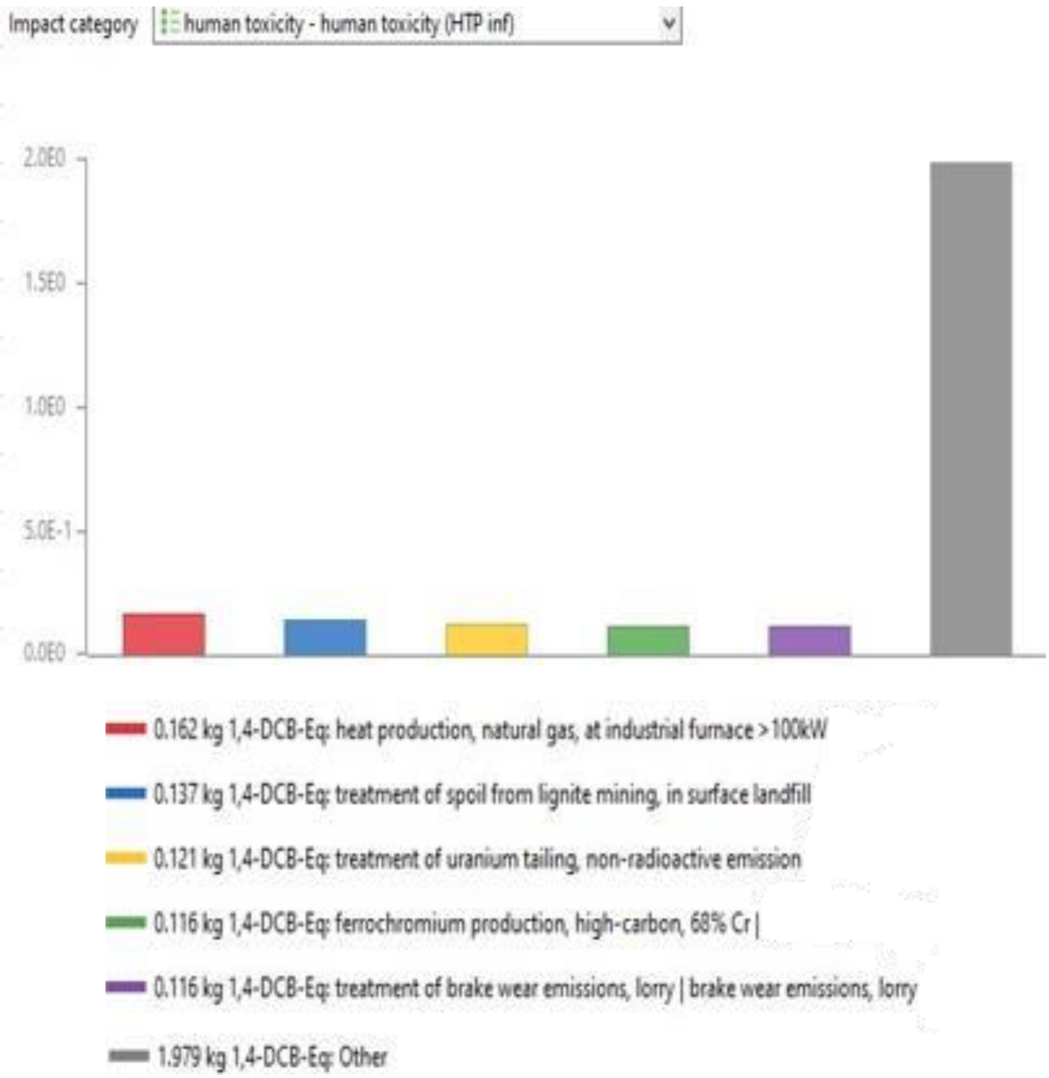


FIGURE 57 OVERVIEW OF TOP 5 CONTRIBUTION TO RESULTS OF THE IMPACT CATEGORY "HUMAN TOXICITY", CASE EOL SCENARIO B).

Conclusion

The study conducted has allowed us to identify some critical points along the product's production chain. The major contributions among the recorded environmental impacts were associated with the "Lamination" phase as well as the production of the semi-finished product to produce the finished pie. This is directly related to the inclusion of paper production and raw materials transportation within this process. Indeed, the production of pulp and paper is traditionally considered a significant source of pollution due to its high energy consumption and the use of large quantities of chemicals, fuels, and water. Additionally, the supplier of paper located approximately 2200 km away from the company would generate environmental impacts resulting from its transportation that cannot be overlooked. Therefore, the highest environmental burdens are attributed to processes upstream of those performed by Ecopack.

Two different end-of-life (EoL) scenarios were hypothesized for the analyzed product to assess the impacts of composting and recycling pies among paper-based items.

In general, higher environmental loads are associated with the production of 1 kg of recycled paper from 1 kg of wasted pies. For all the Impact categories assessed, data related to the selected recycling process were at least an order of magnitude higher than those of composting products. This could be attributed to the environmental loads of chemicals required for the waste valorization as recycled paper. However, the results recorded for these two EoL scenarios are based on rough estimations from secondary data. This is because currently the product "8018030B" is not classified by national unions as "compostable", and no primary data on specific industrial composting and paper recycling facilities were provided. Therefore, considerations about any potential reduction of environmental impacts that could be achieved with precise waste treatment are limited and bound to the assumptions made for the LCA study. More in-depth studies can be addressed, however, already through these data, it is clear what further directions to follow to improve the environmental sustainability of the product.

5. Innovative sticks for Chupa-ChupsPremises[38,79]

The Single-Use Plastic (SUP) Directive came into effect across all European Union countries starting from 2019, undergoing continuous updates and being transposed by all EU member states. Its aim is to prohibit producers of single-use plastic packaging from manufacturing and placing certain packaging articles on the market. Some examples of food and non-food packaging products not subject to this ban include cotton swabs for ear cleaning, cutlery, plates, straws, drink stirrers, balloon sticks (excluding industrial or professional use), containers made of expanded polystyrene (EPS) for immediate consumption or takeaway of foods without further preparation, beverage containers, and cups, as well as all single-use oxo-degradable plastic items.

Many food products, especially in the confectionery sector, rely on support sticks typically made of plastic. For instance, the popular lollipop consists of candy of various shapes attached to the end of a plastic stick.

In the healthcare sector, products like cotton swabs often feature a stick, usually made of plastic, with its ends coated in hydrophilic cotton. Due to significant environmental concerns related to the use and accumulation of plastic, especially near water bodies, the use of plastic sticks has sharply declined since the late 20th century, with many countries banning them.

An alternative material for these sticks has been identified in paper. While paper is environmentally preferable, it presents functional challenges, mainly due to its degradation when exposed to water. It's important to note that products using paper sticks instead of plastic ones are typically used in situations where contact with water or liquids is unavoidable.

As can be understood, if the paper stick deteriorates upon contact with water, its supportive function is compromised, rendering the product unusable. Paper degradation can also lead to a loss of rigidity in the stick, resulting in reduced functionality. Regarding the rigidity of the support stick, it's worth emphasizing that high rigidity is considered beneficial for functionality.

Objective

There was a need for support sticks made of paper, characterized by technical properties that ensured both high water resistance and considerable rigidity.

Through a preliminary market analysis, it was possible to understand the appropriate research and development phases to develop a new high-performance product for process applications. Therefore, the development process focused on creating a product with improved features compared to the latest configurations available on the market.

The improvement actions involved an eco-design approach to the product and focused on two main aspects:

- achieving a lighter product in terms of raw material usage without sacrificing performance properties
- having a recyclable product made of paper and economically sustainable compared to the stick market.

Below is a representation of the machine that produces the sticks (Figure 60).

FIGURE 58 THE PRODUCTION SYSTEM OF PAPER STICKS.[80]

The main parts of the plant are as follows:

- Stick winding and formation (left)

The first part of stick winding and formation involves components consisting of the feeding group where the paper coil is loaded; the tensioner that provides the correct tension to the paper for processing; a peristaltic pump for dosing the water needed to moisten the paper and allow it to be wound on itself for a precise number of times, and finally, the coil width adjustment group which serves to adjust the length of the sticks.

- Thermoforming and consolidation of the stick (center)

In this section, a series of elements are essential to allow for the formation of the paper sticks and consist of a deburring blade that helps the paper take the correct fold to be then wound on itself; a cutting element that cuts the paper sticks to the correct length, and finally, a linear vibrating discharge cup for the formed products.

- Semi-automatic packaging (right)

The paper sticks then undergo a thermal treatment process to remove the retained moisture and allow for satisfactory final rigidity. Heating occurs through passage in a heated vibrating tower at 300°.

The device has been specifically designed and built to meet the technical specifications required by the customer for the finished product. The time spent in this phase is three minutes.

The machine is efficient and production proceeds smoothly until the coil is depleted. However, changing the coil requires a significant amount of time, so optimizing the system to reduce changeover times is necessary. The optimal coil dimensions allow to produce sticks with specific dimensions, but production may be affected if different formats need to be produced. The plant is composed of standard commercial devices, including pneumatic, hydraulic, and electrical systems. The machine is equipped with integrated safety features, although the functionality of the devices has not been verified.

Experimental application phases and testing

It has been discovered that applying a specific layer of surface coating to a stick made of a particular type of paper can effectively meet the requirement.

The subject of this innovative solution is a paper support stick to which at least one functional element is attached. This support stick is characterized by comprising a paper stick made of paper with a grammage between 50 and 90 g/m2 and a fibrous material content of 80% by weight or more, and a surface coating layer comprising nanometric silica (SiO_2).[81]

This surface coating layer is derived from the application of an aqueous treatment solution containing 5 to 50% by weight of nanometric silica, with this aqueous treatment solution being subjected to a thermal treatment at a temperature between 100 and 250°C after being applied to the paper stick.

Preferably, the paper used for the paper stick has a grammage between 55 and 65 gsm and a fibrous material content of 90% by weight or more. Additionally, the paper of the paper stick should have a breaking load between 4.8 and 7.2 KN/m and a surface roughness between 115 and 155 ml/min.

The surface coating layer should be present in an amount between 3 and 10 g per m2 of the paper stick's surface and may include tetraalkoxysilanes and/or alkyltrialkoxysilanes.

Another object of this innovative solution is a product comprising a paper support stick according to any of the preceding characteristics and a food composition or a cleaning material attached to said support stick. Preferably, such a product is a lollipop or a cotton swab.

The above-mentioned sticks were made of paper, each having a length of 75 mm, an outer diameter of 3.5 mm, and a weight of 0.755 grams.

These sticks were made from paper with characteristics as listed in Table.

Properties	Values
Weight ISO 536 (Gsm)	60
Bursting strength ISO 1974 (KN/m)	5,6
Tear resistance ISO 2758 (mN)	460
Bursting strength ISO 1974 (Kpa)	225
Air resistance ISO 5636 (Sec)	30
Cobb 60″ ISO 535 (Gsm)	27
Surface roughness ISO 8791-2 (ml/min)	125

Opacity ISO 2471 (%)	72
ISO brightness ISO 2470 (%)	84
Fibrous material (%)	95
Humidity ISO 532 (%)	5
Filler substances (%)	5.7

TABLE 34 CHEMICAL AND PHYSICAL PROPERTIES OF THE PAPER SELECTED FOR THESE APPLICATIONS.[82]

The values reported in the claims are calculated according to the standards listed in the tables of this description.

The above-described paper sticks underwent a coating treatment with an aqueous solution of nanometric silica (SiO_2) as described below.[83]

The coating treatment involved the application of an aqueous suspension of nanometric SiO_2 and a drying phase at a temperature between 100 and 250°C.

Specifically, the aqueous suspension used contained 30% by weight of nanometric SiO_2.

Properties	Values
Specific weight (kg/m^2)	1.3
Grammage (Gsm)	3
Contact angle (°)	120
Melting point (°C)	1100
Fire reaction (UNI EN 13201-1:2009)	A1
Recyclability	In variation
Compostability	Already composted

TABLE 35 FUNCTIONAL BARRIER PRODUCT PROPERTIES FOR PAPER STICKS.

The aqueous suspension was applied at a rate of 15 gsm. Subsequently, after the deposition of the aqueous suspension, the paper sticks underwent a drying phase through thermal treatment in an oven at a temperature of 160°C for three minutes. The resulting coating layer obtained after the drying phase amounted to 5 gsm of the stick's surface area. The paper sticks obtained from the thermal treatment underwent thermogravimetric analysis, water resistance testing, and mechanical stress rigidity testing.

To support the effectiveness of the innovative solution, the same analyses were conducted on two comparison products: (i) a plastic support stick used in conventional lollipops and (ii) an untreated paper stick (referred to as "untreated stick").

Comparing with the plastic stick serves to assess against a solution effective in terms of functionality but no longer feasible for environmental reasons, while comparing with the untreated paper stick serves to assess against a solution that, although environmentally acceptable, has poor functional efficacy.

- Thermogravimetric Analysis

Thermogravimetric analysis determined the relative humidity of each sample, and the conditioning of the samples required for subsequent analyses.

The analysis was conducted according to ISO 287 method.

The table below shows the recorded values of relative humidity.

Sample	Values (HR% Relative umidity)
Stick of the invention	3.1
Plastic sticks	1
Uncoated sticks	2.7

TABLE 36 COMPARATIVE RELATIVE HUMIDITY ACROSS SAMPLES IN DIFFERENT MATERIALS.

The results reported in the table show that the invention stick, and the uncoated paper stick have approximately the same moisture content, while the plastic stick has a lower moisture content.

- Water resistance analysis

The Cobb test was performed to determine the amount of water absorbed in a standardized time of 30 minutes. The analysis complied with TAPPI 441 standard.

The detected values are reported in Table below.

Sample	Weight (gr)	Weight (gr of water absorption)
Stick of the invention	0.755	0.17
Plastic sticks	0.500	n.a.
Uncoated sticks	0.800	0.21

TABLE 37 WATER RESISTANCE VALUES OF THE ANALYZED SAMPLES.

From the values reported above, it is evident that the stick of the present invention absorbs 24% less water than the uncoated paper stick. The innovative stick is therefore more water-resistant, demonstrating a highly hydrophobic character compared to the uncoated paper stick. It has been observed that the paper stick tends to disintegrate once it meets water, obviously compromising its functionality. However, the plastic stick is not comparable due to the nature of the material itself. In fact, the analysis under examination is specific to applications on paper products. The hydrophobic properties of plastic are not detectable with this instrument due to the intrinsic nature of the material.

- Analysis of stiffness under mechanical tensile

This analysis was performed using a Taber-type tester, which allows evaluating the force required to permanently deform the sample under examination at a defined folding angle (7 or 15°). The reference standard used is ISO 2493-1:2010.

In particular, the force measured and expressed in mN is evaluated by applying the force at the midpoint of the sample, and the higher the force value, the greater the stiffness of the sample. Obviously, for a correct comparison, it is necessary for the samples to be under the same temperature and humidity conditions.

The recorded values were reported based on the weight of the sample and expressed as mN/g. Specifically, the analysis was performed by comparing the values obtained at t0 under standard conditions (25°C and 50% humidity) and the values obtained at t30 under the water resistance conditions mentioned above (Cobb test 30 min).

The detected stiffness values according to the above are reported in table 35.

Sample	Mechanical strength t0 (mN /gr)	Mechanical strength t30” (mN /gr)	Δ Mechanical strength
Stick of the invention	6.6	6.5	0.1
Plastic sticks	5.2	5.1	0.1
Uncoated sticks	6.1	5	1.1

TABLE 38 VALUES OF MECHANICAL STIFFNESS OF THE ANALYZED SAMPLES.

The values reported in the table above demonstrate that:

- at t_0, the innovative stick is 27% stiffer than the plastic stick and 8% stiffer than the uncoated stick.
- at t_{30}, and therefore under prolonged water contact conditions, the innovative stick maintains its stiffness performance, unlike what was observed for the uncoated stick.

It is therefore noted that, for the duration of the expected applications, the innovative stick has stiffness comparable to the plastic stick, even though it is constantly in contact with water. While the advantages recorded in terms of hydrophobicity could somewhat be expected, the stiffness values obtained were unexpected and significantly support the innovative nature of the present invention.

The surprisingly obtained results on the innovative stick are attributed to the combination of the two selected elements: paper and barrier product, which interact with each other to achieve successful outcomes. Through a literature search, it is possible to interpret what has been achieved. The high purity paper allows for facilitated penetration of the barrier product through capillary transport, enabling the product to occupy the nanopores of the cellulose (characteristic spaces of the selected cellulose type).[81]

Above is a chemical illustration explaining the interaction between the two elements that functionalize the finished product. Therefore, the interaction between solvent evaporation in which the barrier product is dispersed, its micellar concentration, and its capillary transport rate through the cellulose material enable the functionalization of cellulose pores, resulting in changes in both hydrophobic properties and structural rigidity.[84]

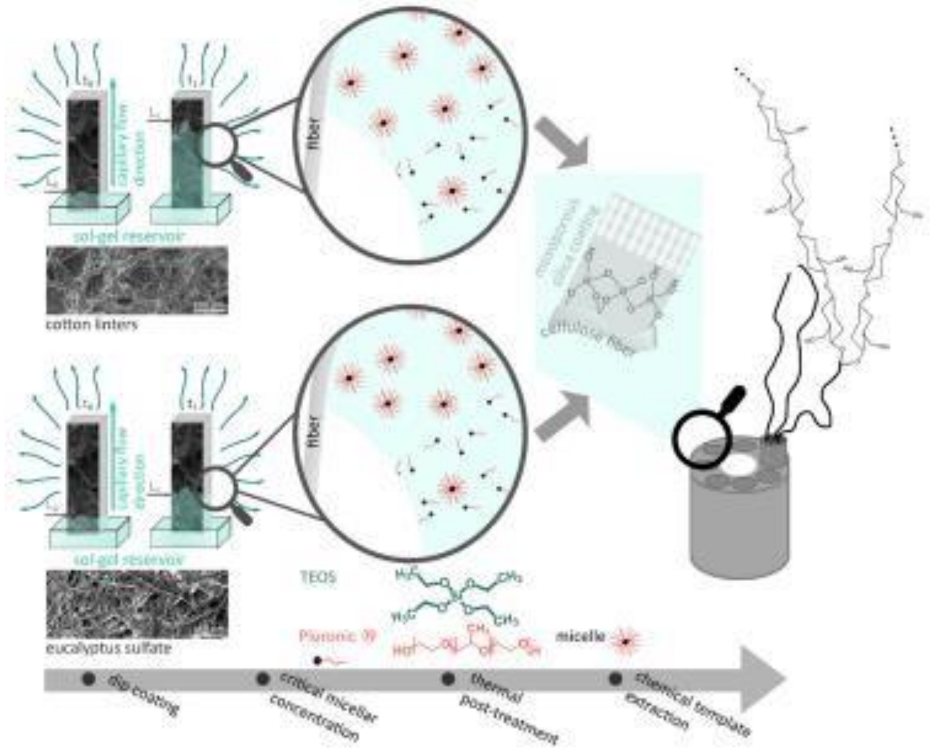


FIGURE 59ILLUSTRATION OF THE PRINCIPLES OF INTERACTION BETWEEN THE BARRIER AGENTS.[81]

The illustrated process provides a chemical interpretation of what occurs: the functionalization of the involved components through barrier product hydrolysis activity followed by a polycondensation reaction allows for the formation of chemical bonds between the groups of the elements involved.

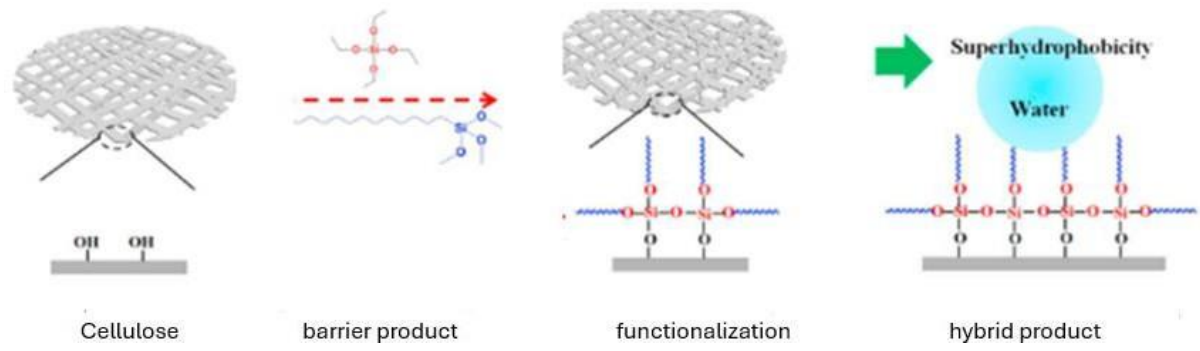


FIGURE 60 CELLULOSE FUNCTIONALIZATION PROCESS THROUGH THE BARRIER PRODUCT.[85]

To achieve effective chemical functionalization between the components, it is essential that the cellulose has a high degree of purity to allow for the best interaction with the barrier product. The characteristics of element a (selected paper) are:

- High degree of surface smoothness
- High compatibility with treatment processes
- High purity of cellulose (100% virgin fiber)

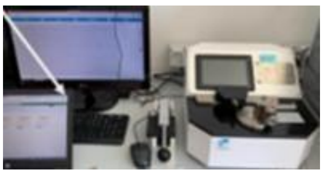
In fact, it was unforeseeable that the treatment provided by the present invention could ensure the stiffness subsequently observed. It has been identified that the effectiveness of the innovative solution depends on the type of paper used to produce the paper stick to which the aqueous treatment solution is applied. In fact, following experimental evidence obtained with different types of paper, it has been demonstrated that paper with a high percentage of fibrous material and a certain grammage ensures better interaction with the substances present in the aqueous treatment solution.

Below are some images depicting the main tests conducted following the development of the innovative sample for comparison with the standards.

Reproduction of the process phase and storage of the paper sticks

Reproduction of the paper sticks in the final usage phase

Bending Resistance instrument evaluation of stiffness on the samples



Analytical balance for gravimetric evaluation of water absorption of the samples

FIGURE 61 THE INSTRUMENTS USED FOR CHARACTERIZATION ON THE SAMPLES TO EVALUATE AN IMPROVEMENT SOLUTION.

Conclusion

The development project for this innovative new product has combined scientific expertise in chemical products and raw materials, particularly paper, with technical process engineering skills. The newly developed product would replace current plastic sticks, fully complying with the requirements of the SUP (Single-Use Plastic Directive), which mandates the replacement of specific packaging items with more sustainable alternatives.

The environmental impact of the innovative packaging will be significantly reduced, while still ensuring the functional characteristics required for the specific application. The tests conducted have confirmed the feasibility of industrial-scale production. The innovative packaging will serve to support and hold an element (such as a lollipop), and therefore, the paper's grammage must be between 50 and 90 gsm. This range is necessary to ensure sufficient initial stiffness, which will then be optimized with the barrier treatment.

The silica (SiO_2) coating layer, obtained from the application of an aqueous treatment solution, is subjected to a thermal treatment between 100 and 200°C, to ensure good surface protection of the finished product.

The development of this innovative new product could bring benefits to the food industry in the coming years, as it increasingly needs to meet environmental sustainability criteria.

6. Development of a new line of grass paper

productsDesign requirements

In recent years, the food packaging industry has highlighted increasingly targeted needs towards sustainability, pursuing an approach to the circular economy through the creation of eco-friendly products made from new, innovative materials that are also engineered.

Objective

Thus arose the proposal to develop new packaging containers for the bakery sector, simple in appearance yet featuring high technical and eco-sustainability characteristics. The preliminary approach focused on identifying a new and innovative raw material suitable for market needs, aiming to envision the success of introducing a new product line that meets sustainability requirements.

To conduct experimental activities related to research and development within the scope of my PhD program thesis applied to packaging sustainability, it was decided to present a project aimed at developing a new product line to be proposed as more eco-friendly paper food packaging. These packaging solutions will be manufactured using innovative and compostable raw materials, scalable at an industrial level, and economically sustainable.

The preliminary approach focused on identifying a new and innovative raw material suitable for market needs, with the aim of ensuring the success of introducing a new product line that meets sustainability requirements.

The goal is to establish a new product line that meets predefined sustainability criteria; recyclable, compostable, and economically sustainable enough to be compatible with market economic standards. Currently, there are no grass paper packaging products for the bakery sector, as confirmed by thorough market research.

Market research

It is evident that the only products available on the market are shoppers, adhesive labels, wrapping paper, bags for fruits and vegetables at supermarkets, cardboard boxes for moving or shipping (including smaller ones), tissues, toilet paper, napkins, shipping envelopes (also in cardboard), supermarket tomato trays or packaged fruit trays, product displays (ranging from large displays to business card holders), copy paper (plain sheets), gift wrapping paper, sticky notes, notebooks, diaries, and sample trays.

Below are some samples, Figure 64:

What are the current uses?

New application for a new market

FIGURE 62 PRODUCT CATEGORIES IN GRASS PAPER.

Description of the project phases

To address the design requirements, research was conducted to identify the most suitable raw material that would meet the specified criteria, selecting paper with distinctive characteristics and good technical features compatible with Ecopack' s production processes.

The chosen paper, named "Kazan," is produced by the Burgo paper mill, a part of the Mosaico Group. collaborating with the paper mill, experimental activities were initiated to enhance performance, adaptability to the production process, and suitability for the bakery sector.

Kazan paper is part of the green paper family, made from virgin cellulose fibers partially sourced from grass. Grass fibers, obtained through grinding and drying grass from urban green areas, replace a significant portion of cellulose fiber that would otherwise require sulfate-based chemical processes.

From this grade of paper, the initial experimental development phases were undertaken, resulting in a new paper named Kazan C Barr. These activities were conducted in collaboration with the paper mill under a confidentiality agreement between the parties.

Preliminary project phases

The development of this new and innovative paper involved initial laboratory-scale phases focused on testing surface treatments with additives and barrier products to identify the most suitable option in terms of process and performance, ensuring compatibility with the final application on the product. Subsequently, adjustments were made to the production process to create Kazan C Barr, a printable offset paper with pleasing visual, tactile, and olfactory characteristics.

Kazan C Barr paper features include:

- Composition of up to 40% virgin grass cellulose fiber added to wood cellulose fibers.
- Reduction of CO_2 emissions by up to 75% compared to equivalent products made solely from wood cellulose.
- Biodegradability
- Recyclability according to European regulation EN 13430[78]
- Compostability according to European regulation EN 13432[78]
- Excellent resistance to high temperatures, meeting BFR 36/2 requirements.[86]

Following the selection of the paper, collaborative activities were defined with Ecopack to commence initial studies and prototyping tests for the new product.

Experimental application phases and testing

In this section, the activities of prototype development and subsequent product sampling tests are reported to verify and make any necessary improvements to achieve the predefined objectives and thus present the potential new Ecopack packaging line.

Below is a table outlining the application phases, objectives, and additional notes:

Phase	Design solution. Phase 1	Deliver products. Phase 2
Objective	To certify the innovative and eco-sustainable paper on our production Technologies.	To approve categories of products on Ecopack production line
Price (euro/kg)	Δ≈- 15% vs standard product	To provide an economic advantage for most possible product categories

TABLE 39 STAGES OF THE PROJECT ACTIVITY.

A preliminary study was conducted to ensure the economic sustainability of introducing a new product line made from innovative grass paper into the market. By analyzing the costs of raw materials, production, and additional fixed costs, we evaluated the economic benefits that the new grass paper product line could bring compared to standard paper and similar innovative paper types. As observed, if the prototyping, sampling, and production testing activities yield the desired results, the market introduction of the new line will not only bring innovation as an environmentally sustainable product but also offer economic advantages due to the reduced cost of the raw material used.

Items	Type paper (Gsm)	paper (€/Kg)	Raw (material/k)	Processing (€/k)	Total (cost €)	Delta Saving
"Tulip" standard	50	2.825	4.58	2.78	7.36	+7% tulip grass
"Tulip" cocoa paper	50	2.875	4.52	2.78	7.30	+6% vs tulip grass
"Tulip" grass Kazan c bar (paper's name)	50	2.45	4.07	2.78	6.85	saving
"Lotus" standard	60	2.825	4.97	3.29	8.26	+6% vs Lotus grass
"Lotus" cocoa paper	60	2.875	5.04	3.29	8.33	+7% vs Lotus grass
"Lotus" grass "Kazan c barr"	60	2.45	4.46	3.29	7.75	saving

TABLE 40 SOME EXAMPLES OF ECONOMIC SAVINGS THAT THE NEW INNOVATIVE PRODUCTS WOULD CONTRIBUTE.

In defining the requirements for our project, we have focused on several key characteristics.

Firstly, we have considered the material's slipperiness, measured through the COF test, the static coefficient of friction is used to evaluate whether the paper has adequate slipperiness, necessary for being easily machinable and meeting the final requirements requested by the customer. The measurement is done by applying a force to the sample: the greater the force needed to initiate movement, the higher the sample's friction. The value of the applied force and the separation force determine the coefficient of friction. This parameter plays a crucial role in the industrial processability of the products we intend to manufacture, ensuring smooth and efficient processing.[87]

Additionally, we have emphasized the grease resistance, a feature requested by our customers to prevent the grease from cooked products from penetrating the paper of the mold, thus avoiding unwanted stains on the finished product.

Similarly, we have considered moisture resistance, essential to ensure that the packaging maintains its functionality even in standard environmental conditions and retains its original shape.

Another crucial characteristic is resistance to high oven temperatures, which must comply with the applications required in the bakery sector.

Finally, we have ensured the recyclability of the material and obtained the necessary certifications for its sustainability, including FSC certification and OK COMPOST INDUSTRIAL certification, ensuring that our product is free from hazardous and persistent substances for human health and the environment.

Following the initial phases of characterization on the paper, some limitations related to the composition and properties of the raw material have been highlighted.

- Low grease resistance (limitation on the final product)
- Low slipperiness (limitation on the Ecopack process)
- Low moisture resistance (limitation on the product)

Furthermore, the first version of Kazan C Barr does not meet all the requirements for environmental sustainability (e.g., absence of compostability certification according to EN 13432)[78] and applicative compliance (e.g., FDA 176.170 compliance for fatty and moist foods; compliance for the oven according to BFR 36/2 requirements).[86]

The second phase thus involved testing to verify and validate the paper requirements. In collaboration with the paper mill, specific modifications were made to the raw material to achieve the stated objective.

A compatible coating was identified, and compliance with the necessary quality standards was reached, obtaining all the approvals for the new version of Kazan C Barr.

The paper's approval required the implementation of some aspects related to the production process technologies and the study of the process phase in which the barrier product was to be applied to bestow the desired properties to the paper.

Performance tests, both technical and applicative, were carried out again on the new version.

The results of the assessment of the properties of the new paper product have been satisfactory, considering the reliability and preliminary importance of the tests conducted to validate the suitability of the innovative paper for the development of new innovative products.

The new product has surpassed the grease resistance requirement compared to the standard. Using the DIN 53116 method, the data indicates a level 3 grease resistance, corresponding to a medium-high grease resistance on the finished product.

Furthermore, excellent moisture resistance has been achieved, with data obtained through the Cobb 60" test indicating a value below 10 gsm, approximately 20% better than the product standard.

Surface slipperiness of the paper has been significantly improved, with measured coefficient of friction (COF 0,12) values providing results comparable to the slipperiness of standard papers.

The suitability of the new product for baking applications for fatty and moist foods has been confirmed.

The new paper product represents a significant innovation, offering an exceptional combination of grease resistance, moisture resistance, slipperiness, and suitability for baking foods, ensuring superior performance compared to industry standards.

The missing requirements for environmental sustainability and applicative compliance have also been achieved as per the table below:

Parameters	Values
Grammage	50 – 130 gsm
FDA (food contact with bakery food)[88]	In compliance
Reg EU directive for packaging food[78] 1935/2004 & 2023/2004	In compliance
DM 21/03/1973 [78]	In compliance
FSC	In compliance
Recyclable[78]	In compliance Method Aticelca system level B
Compostable[78]	In compliance OK Compost Industrial TA 8012207184
PFAs Free[89]	In compliance

TABLE 41 COMPLIANCE AND SUSTAINABILITY PROPERTIES ON THE RAW MATERIAL.

Prototype testing and product samples.

Once all the above requirements were confirmed, it was possible to proceed with the production of the first product prototypes.

The initial samples were produced starting from some paper samples that were thermoformed on the machine in semi-automatic mode.

Below is the image of the first prototype of the muffin cup product.

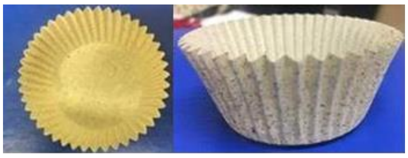


FIGURE 63 FIRST PROTOTYPE MADE USING THE INNOVATIVE GRASS PAPER.

The prototypes confirm their adaptability to the die-cutting and thermoforming production process; the machine parameters remained the same as those applied in standard productions, and the evaluated performance on the final product was achieved.

The prototypes have excellent shape memory, good resistance to the grease of the cooked dough, which is maintained for the entire time necessary for its function, and the aesthetic appearance is very distinctive and attractive.

Given the positive outcome of the initial prototype testing, technical samples were produced in production using process machines while keeping machine parameters constant.

Five samples were produced, creating six types of products to validate the feasibility of success on a production scale and confirm the success of the proposal for the new product line.

The sampling activities were managed following company schemes and workflows to track the technical and qualitative results obtained on the products.

All product samplings carried out yielded positive results, with an outcome confirmed by the quality team of the product compliance group.

It was possible to confirm the adaptability and success of the new product line in terms of production on an industrial scale.

Below is a table with the number of sampling cases and the outcomes of:

- machinability in the process
- applicative compliance on the final product

Items	Outcome of machinability in process	Outcome of applicative tests
Tulip grass paper	In compliance	In compliance
Lotus grass paper	In compliance	In compliance
Pie grass paper	In compliance	In compliance
Beking cups grass paper	In compliance	In compliance

TABLE 42 PRODUCT SAMPLES MADE USING PRODUCTION MACHINES.

To achieve compliance results for the sampled products, numerous adjustments regarding machine parameters and the specific paper thickness selection were necessary. The products listed in the table are indicated as compliant both in terms of machinability and paper behavior on the machine, as well as in terms of the required quality standards for the finished product, to validate and approve them as suitable to be proposed as new products to be introduced in the packaging market. The product's qualitative compliance involved adhering to regulations to consider the product applicable to the food packaging sector for bakery applications.

Below are some images regarding the performance evaluations of the sample products: Qualitative (aesthetic) appearance of grease resistance of the "lotus" samples.

FIGURE 64 APPLICATIVE TEST COMPARING SAMPLES IN GRASS PAPER TO STANDARD SAMPLES.

On the left, the sample in grass paper; in the center and on the right, the standard samples (standard paper and cocoa paper).

Following the internal testing flow, empirical tests were conducted on the innovative products, which represent those requested by customers to verify whether the prerequisites for testing samples on a small scale exist from the preliminary trials. Therefore, tests were carried out on sweet baking foods in the oven, following the methods and conditions used by large industries.

During these tests, the resistance of the product to food grease over the prescribed time, namely after 24 hours, was evaluated, and the desired result is that the grease does not penetrate the paper, remaining confined inside without passing from the inner side to the outer one. Additionally, for rectangular or round-shaped products, such as pastries or cakes, easy detachment of the food from the packaging after baking was required. It is evident that the best sample from this point of view is the grass paper one; the food cooked in the grass paper baking mold tends to release easily, surpassing the quality standard required on an industrial scale.

A similar result to the previous one was obtained on the "Pie" sample; in this case, the comparison was made with the standard sample in cardboard coextruded with PET.

FIGURE 65 APPLICATIVE TESTS WITH BAKED CAKE BATTER COMPARING THE GRASS PAPER SAMPLE WITH SAMPLES WITH PET BARRIER FILM.

Finally, as a last example, we took the "plumpy" sample and compared it with the same product in silicone-coated paper. In this case too, the outcome was positive; the sample meets the requirement demanded by the bakery market.

FIGURE 66 APPLICATIVE TEST IN THE OVEN OF A MOLD MADE OF GRASS PAPER (LEFT) VS. SILICONIZED PAPER (RIGHT).

On the left, the sample in grass paper vs. the sample in single-silicone-coated paper.

As can be seen from the image above, the food detachment on the innovative product is comparable to that achieved on standard paper products, therefore, it meets the required specifications.

During the applicative tests, the analysis plan was submitted to the TUV Austria certification body to assess further analysis tests to certify the product family with the TUV logo and mark.

A pre-analysis was conducted based on the evaluation of the samples to be certified, considering criteria such as paper grammage, paper thickness, and the same for the finished products.

The compostability requirements being present on the raw material, it was necessary to conduct the quantitative disintegration test to achieve "OK COMPOST INDUSTRIAL" TUV certification for the product family.

"OK Compost TUV" is a certification mark for compostable products, issued by TÜV Austria, an independent certification body. This certification indicates that the product has been tested and evaluated according to international standards for compostability, confirming that it can be safely and effectively composted in industrial composting facilities. The presence of the OK Compost TUV mark on a product provides users with assurance that the product complies with environmental regulations and can be sustainably disposed of through composting. Given that there are several marks with the TUV logo, it is important to distinguish them for specific applications. In this case, the certification involved for the new product line is OK Compost Industrial, which recognizes that the products are compostable in industrial composting facilities under specific conditions and process parameters, ensuring that the material composts in the required manner.

Below is the table and evaluation of the worst-case scenario selected for the disintegration test according to European method and standards.

Items	Minimum size (mm)	Maximum size (mm)	Paper (Gsm)
Muffin	45*35	63*40	65
Plumpy	80*40	-	125
Pie	40*20	90*40	125
Backing cups	20*17	102*53	50
Tulip	20*17	50*96	50
Lotus	50*62	82*85	50

TABLE 43 LIST OF SPECIFIC GEOMETRIC SPECIFICATIONS AND GRAMMAGE ON SAMPLED PRODUCTS.

The list above identifies the products that may be covered by the compostability certification accredited by the certifying authority.

Items	Thickness (µm)
Muffin	305
Plumpy	475
Pie	380
Backing cups	50
Tulip	180
Lotus	180

TABLE 44 SELECTION OF THE WORST-CASE SCENARIO FOR THICKNESS ANALYSIS.

The sample selected for the test is the "plumpy" because it has a higher grammage and thickness of both the paper and the finished product compared to the other samples. Despite having higher grammage and thickness, since the paper is tested and certified compostable, any potential influence that could compromise compostability on the product line would be solely due to quantitative disintegration over the required time. Therefore, the necessary test was conducted on the product that overall had the highest thickness, which in our case turned out to be the plumpy.

The test was conducted by the accredited laboratory, and the analysis results showed positive outcomes, allowing for the issuance of the compostability certificate for the developed product line.

To confirm that the raw material used to produce the products in the new line is the same as the one certified as compostable, a parallel FTIR analysis was required alongside the quantitative disintegration test. This allowed certifying the new product line as OK compost Industrial.

Qualitatively, the bands attributable to the coating (encircled in yellow) are similar in the spectra of the certified paper and the worst-case plumpy sample and are compatible with the hypothesis that the coating is a styrene-acrylate copolymer. Specifically, the red spectrum = matte side of the certified 2022 paper; purple spectrum = glossy side of the certified 2022 paper; green spectrum = glossy side of the worst-case plumpy sample.

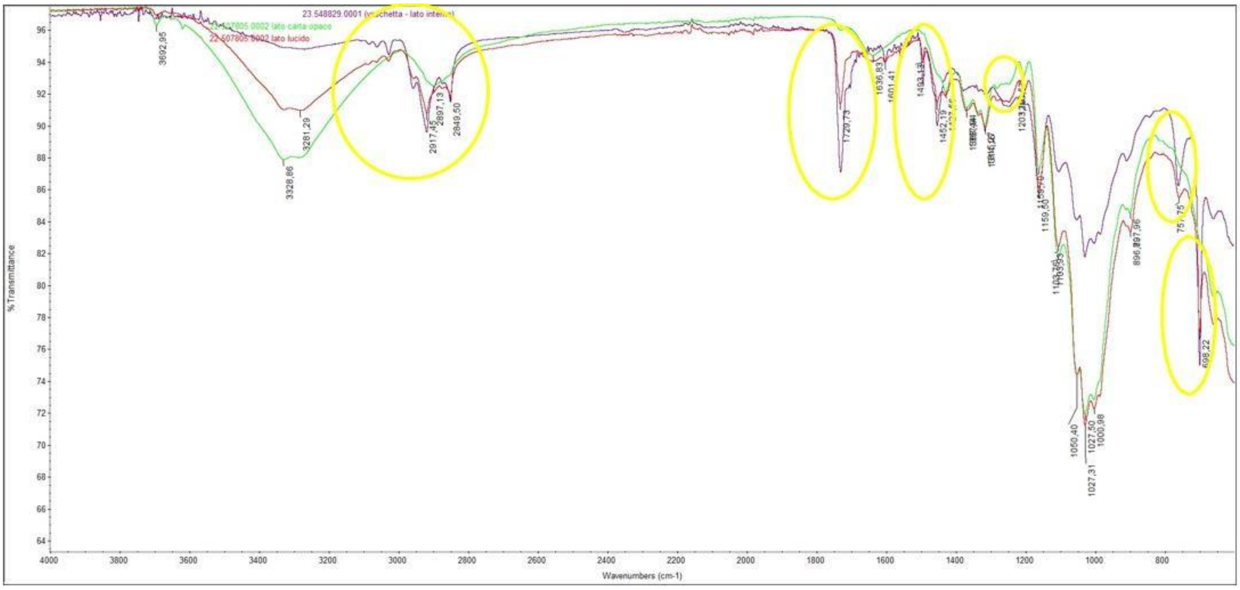


FIGURE 67 OVERLAY OF THE SUBTRACTED SPECTRA PAPER VS "PLUMPY "PRODUCT.

Routine FT-IR analysis can provide, in the case of a thin coating on paper, an output that is the sum of the coating and what is underneath it. The summation of spectra does not allow good confidence regarding the percentage of spectrum overlap, which is not very qualitative in any case, but can lead to compatibility.

Conclusion part 1

Having achieved positive outcomes in all project phases, the feasibility of successfully promoting the new line of grass paper products can be confirmed. The set objectives have been met, and the new line of grass paper products can enter the market and be easily accepted as it respects aspects of environmental sustainability both in terms of the raw material used and in terms of end-of-life packaging. The approach adopted in the project phases has been effective and aligned with the approach of sustainable product design. The design of the new line of grass paper products meets criteria of environmental, social, and economic sustainability, respecting the environment, and this can be further confirmed through additional studies such as Life Cycle Assessment (LCA) in accordance with environmental sustainability standards for packaging such as ISO 14044:2006. Marketing campaigns will be planned to introduce the product to market customers and new potential clients. Therefore, there will be an initial exploratory market phase to understand if the product can generate interest. Meanwhile, extensive production will be carried out to ensure the workability of the paper for all grammages. The new product line meets all sustainability requirements and is certified as compostable. Its end-of-life, as well as the raw material used, are virtuous and should significantly reduce environmental impact on indicators such as water consumption, energy, and CO2 emissions. Below is the LCA study on the paper compared to a standard paper, where the environmental benefits of the innovative paper can already be observed.

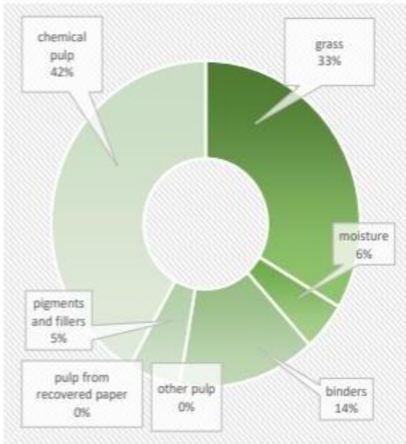
Grass paper "KAZAN C BARR"

ENVIRONMENTAL PARAMETERS

The figures are based on methods and procedures of measurement approved by the local (or national) environmental regulators at the production site. The figures include pulp and paper production.

Water	COD	7.3	kg/tonne
	AOX	0.033	kg/tonne
	NTot	0.111	kg/tonne
	PTot	0.013	kg/tonne
Air	SO2	0.032	kg/tonne
	NOx	0.625	kg/tonne
	CO2 (fossil)	673.5	kg/tonne
Solid waste landfilled		4.0	BDkg/tonne
Purchased electricity consumption /tonne of final product		264.9	kWh

PRODUCT COMPOSITION



This product contains biomass carbon, equivalent to 1380 kg of CO2 per tonne of product.

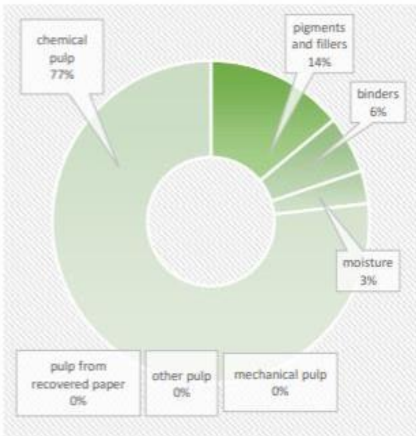
Standard paper

ENVIRONMENTAL PARAMETERS

The figures are based on methods and procedures of measurement approved by the local (or national) environmental regulators at the production site. The figures include pulp and paper production.

Water	COD	11.1	kg/tonne
	AOX	0.065	kg/tonne
	NTot	0.135	kg/tonne
	PTot	0.021	kg/tonne
Air	SO2	0.064	kg/tonne
	NOx	1.051	kg/tonne
	CO2 (fossil)	725.5	kg/tonne
Solid waste landfilled		6.9	BDkg/tonne
Purchased electricity consumption /tonne of final product		308.4	kWh

PRODUCT COMPOSITION



This product contains biomass carbon, equivalent to 1408 kg of CO2 per tonne of product.

FIGURE 68 COMPARISON OF SUSTAINABILITY DATA BETWEEN INNOVATIVE PAPER AND STANDARD PAPER.

Conclusion part 2

As a result of worst-case evaluations and tests, it was possible to obtain compostability certification for the entire line of packaging products intended for the market of the bakery and confectionery industries. The official certification document transmitted by the globally recognized certifying body demonstrating the compostability of the entire product line is transmitted below.

Product line certification code **Product Attachment**

CERTIFICATE FOR AWARDING AND USE OF THE
'OK COMPOST INDUSTRIAL' CONFORMITY MARK
TA8012408291

Issued by TÜV AUSTRIA GMBH

Product(s):
Domain: Industrially Compostable Products
Group: Finished Products
Family: Other
Type: Baking moulds
Trade mark: Terra moulds for bakery food
Description / Particularities: List of certified products in Annex 1; Maximum nominal grammage: 127 g/m²; Colour: Light brown; Unprinted

Licensee: Ecopack SpA, Via della Masolina 24, 10040 Piobesi Torinese (TO), ITALY

Criteria: • Certification Scheme with reference OK 01 edition F Including UNI EN 13432:2002 : « Imballaggi – Requisiti per imballaggi recuperabili mediante compostaggio e biodegradazione – Schema di prova e criteri di valutazione per l'accettazione finale degli imballaggi »

Validity: From 29 May 2024 till 29 May 2029

Conclusions of the examination: The products comply with the above mentioned certification criteria, as confirmed by the report no 65002156-2024-AG-0063cert.

Applicable certification system: Type examination followed by supervision through verification tests on samples from the distributor's stocks or of the market. The conformity of the product is guaranteed by the procedures for awarding and use of the 'OK compost INDUSTRIAL' conformity mark. This only applies for specimen bearing the 'OK compost INDUSTRIAL' mark.

Rotselaar, 29 May 2024

For the Certification Committee
Ph. DEWOLFS
President of the Committee

Annex: List of certified products (1 page)

This certification was carried out according to the TÜV AUSTRIA GMBH procedures for certification and is regularly monitored.
TÜV AUSTRIA GMBH | Deutschstraße 10 | A-1230 Vienna

Certificate TA8012408291
Validity of the certificate from 29 May 2024 till 29 May 2029
Annex 1

Product	Nominal grammage [g/m²]	Minimum dimensions [cm]	Maximum dimensions [cm]
Muffin	50 or 65	45 x 35	63 x 40
Plumpy	100 or 125	80 x 40	/
Pie	100 or 125	40 x 20	90 x 40
Pirottini	50 or 65	20 x 17	102 x 53
Tulip	50 or 65	20 x 17	50 x 96
Lotus	50 or 65	50 x 62	82 x 85

This certification was carried out according to the TÜV AUSTRIA GMBH procedures for certification and is regularly monitored.
TÜV AUSTRIA GMBH | Deutschstraße 10 | A-1230 Vienna

FIGURE 80 COMPOSTABILITY CERTIFICATION OF NEW GRASS PAPER PRODUCT LINE

You can go and visit the products by following the links below:

- https://www.ecopack.com
- https://lnkd.in/dq2T_wSr
- https://www.linkedin.com/pulse/terra-moulds-from-earth-back-incredible-journey-blade-grass-cqr7f/?trackingId=oFzVZqlBSR2b2oH7t7cBKQ%3D%3D

Academic research activities

During my doctoral studies, I had the opportunity to conduct chemical characterization tests both in academic settings and at the laboratory of the Department of Chemistry, within the research group MOF - Functional Organic Materials.

Under the guidance of Professor Claudia Barolo and the supervision of Dr. Matteo Bonomo, I performed tests to evaluate the performance of a functionalizing product aimed at creating high-performance electrodes.

The results obtained from the tests contributed to the publication of a scientific article titled "Engineered Surface for High Performance Electrodes on Paper", published in the journal Applied Surface Science.

My contribution to the article includes participation in the characterization and barrier testing phase on papers treated with functional coatings, as well as data processing and drafting of the initial article draft.

Below is a brief description of the article: “"Engineered surface for high performance electrodes on paper”[70]

The article proposes a scalable, eco-friendly, and cost-effective method for manufacturing highly conductive flexible PEDOT: PSS electrodes on paper. Using the blade-coating technique, a cellulose-derived polymer solution was applied to the paper, creating a suitable surface for the deposition of the conductive material.[70]

Among the various water-based coatings tested, hydroxypropyl cellulose at 20% weight proved to be the best solution, ensuring uniform coverage on the paper. By using a low molecular weight polymer, it was possible to reduce the amount of water in the coating dispersions, while simultaneously increasing the polymer quantity and maintaining necessary rheological properties such as viscosity.

This approach allowed for a flatter, less prone to curling, and stiffer paper, while also reducing drying times. Electrodes produced on this paper exhibited higher conductivity and a more stable response compared to those on untreated paper. Additionally, they proved to be more resistant to deformation and showed greater stability over time.

These advanced characteristics are advantageous for use in wearable devices, providing reproducible and durable performance for both sensors and energy storage systems. In summary, electrodes on treated paper offer a more reliable and stable response, reducing the impact of external pressures.

The apparatus depicted in Fig. was utilized to measure the variation of resistance under linear strain. A PEDOT: PSS electrode measuring 0.4x3 cm^2 was clamped between two electrodes. The sample was pulled from one electrode, which was mounted on a motorized linear stage (Stand 039801), operating at a fixed speed of 0.3 mm/min. While the strain was applied, a constant current of 5 mA was passed through the sample, and the voltage differences between the electrodes were monitored using a Keithley 2700 multimeter and recorded. Subsequently, the resistance of the sample (R) was calculated as a function of the relative strain (ε_r).

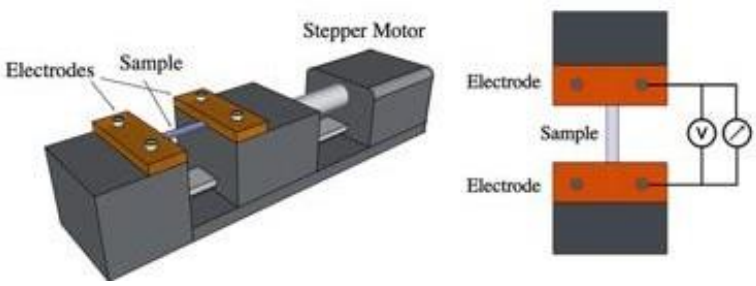


FIGURE 69 SET-UP FOR THE ELECTROMECHANICAL CHARACTERIZATION OF THE ELECTRODES.

Indeed, their enhanced performance, stability, and reproducibility open new possibilities for wearable electronic devices. I have contributed to a second academic project aimed at drafting a second scientific article, which is currently in the draft phase.

The title is "Enhancing Packaging Sustainability: Cellulose-Based Coatings with Improved Barrier Properties and Oxidative Stability".

This article aims to delve into a comprehensive study on the use of cellulose derivatives-based coatings to enhance the functionality of paper as packaging material.

Originating from renewable sources, cellulose derivatives emerge as a promising avenue to improve the mechanical strength and barrier properties of paper. Taking innovation a step further, we introduce an inventive approach by modifying these cellulose-based coatings with detonation nanodiamonds (DND).

Our investigation entails a thorough characterization of the modified cellulose-based coatings, providing insights into their structural and surface properties, and how these properties are altered with the addition of DND to the system. This integration imparts unique properties, including increased mechanical strength, enhanced barrier capabilities, and greater thermal stability. Significantly, we achieved a 70% increase in paper water vapor barrier resistance, a sixfold improvement in mechanical resistance, and enhanced chemical stability in strongly acidic oxidizing environments.

The results underscore the vast potential of cellulose-based coatings, particularly when customized with DNDs, offering sustainable solutions with superior performance characteristics. This research not only addresses the urgent need for eco-friendly packaging materials but also contributes to advancing the capabilities of cellulose derivatives in the packaging industry.

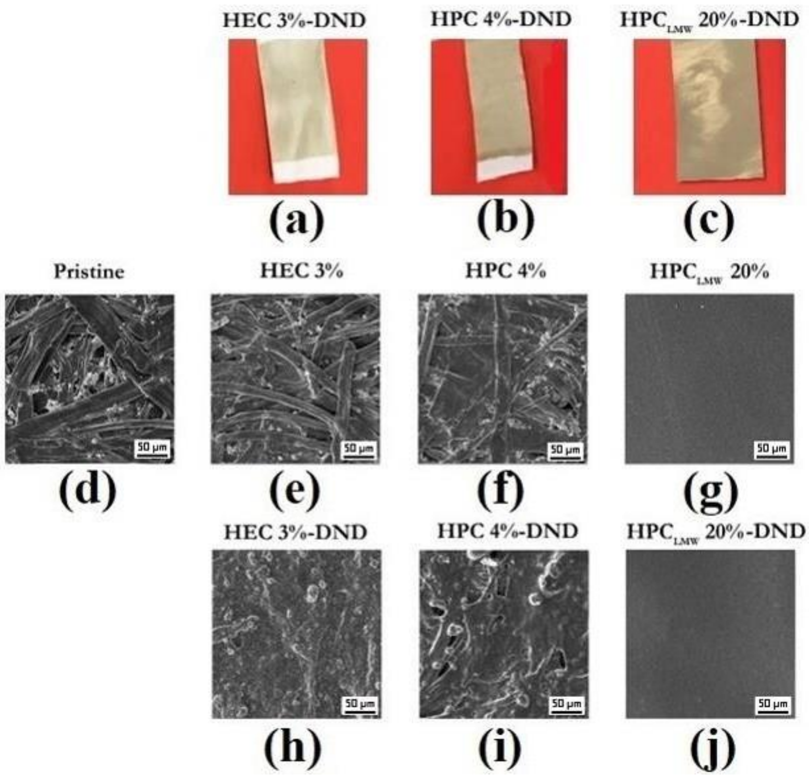


FIGURE 70 PICTURES OF PAPER COATED WITH (A) HEC 3%-DND (B) HPC 4%-DND (C) HPCLMW 20%-DND; SEM IMAGES OF UNTREATED PAPER (D), AND PAPER COATED WITH (E) HEC 3% (F) HPC 4% (G) HPCLMW 20% (H) HEC 3%-DND (I) HPC 4%-DND (J) HPCLMW 20%-DND.

The images in Figure 75 show paper coated with cellulose derivatives-DND coatings. When the dispersion of HEC 3%-DND is applied (Figure 2a), it results in almost homogeneous coverage, although some defects are visible. HPC 4%-DND (Figure 2b) achieves good uniformity of the layer, while HPCLMW 20%-DND (Figure 2c) produces a glossy-like appearance, indicating remarkablyhomogeneous coating deposition. Uncoated paper observed via SEM (magnification = 1000x) reveals a worm-like structure with alternating larger and thinner fibers and some brighter aggregates. EDX analyses confirm the presence of Ca, C, and O, consistent with the use of CaCO3 in the paper manufacturing process. The application of cellulose-based coating reduces surface porosity (Figure 2d).While HEC 3% and HPC 4% coatings still exhibit a fibrous structure (Figure 2e,f), HPCLMW 20% achieves complete coverage of fibers (Figure 2g), making the pristine worm-like morphology barely discernible even at higher magnifications. This transformation is attributed to the higher quantity of cellulose-derivative coating in HPCLMW 20%. DND-enriched coated papers show a uniformly decoratedsurface with small spheres (DND aggregates). In the case of HEC 3%-DND and HPC 4%-DND (Figure 2h,i), SEM images demonstrate that DND helps close paper pores and fill voids left uncovered by the polymeric dispersion. However, no significant enhancements are observed for HPCLMW 20%-DND (Figure 2j), as the polymer alone achieves impressive coverage of the paper substrate in this case.

Acknowledgments

I would like to express my gratitude to all the individuals I have encountered and had the privilege to collaborate with during these three years of research and development focused on environmental sustainability. Our efforts have primarily centered around the realm of food packaging, with significant implications for various other fields, including those addressed within academic pursuits related to the energy sector. I consider myself fortunate to have not only expanded my knowledge base but also enriched my social experiences throughout this journey. It is crucial, in my view, to consistently seize the opportunities presented to us, a principle that I have endeavored to live by during both my professional and academic endeavors, whether within the corporate setting or within the university environment.

Thanks to all those who supported me.

Collaboration and thanksgiving

I would like to thank the entities and academies with which we have collaborated in the implementation of individual development projects in business and research.

The first project was carried out with the collaboration of a company called Alter Eco https://alterecodisposable.com/ a producer of pulp packaging

Project number 2 was carried out thanks to the contribution and support with InnovhubSSI https://www.innovhub-ssi.it/ experimental station for innovation

Project number 3 and 4, on the other hand, required the involvement of the Department at which I do my Ph.D.; the company named Glueton https://www.glueton.it/ for the development of the innovative glue while the contribution regarding the LCA study and analysis was carried out thanks to the support of the University of Eastern Piedmont https://www.uniupo.it/it .

Project number 5 was carried out entirely within the company and was the result of a combination of intuition and technical knowledge of materials and market needs.

Project number 6, on the other hand, was conducted thanks to the collaboration of the Burgo paper mill in Mosaic https://www.mosaicopapers.com/ thus, it was possible to promote a new line of 100% eco-friendly and innovative products.

Overall Conclusion

All the projects undertaken during these years of applied research doctorate in the company have aimed to promote focused attention on the environmental sustainability of new raw materials, production processes, and finished products intended for the food packaging sector. This sector is closely linked to the strategic initiatives of the European Green Deal, which aims to guide the EuropeanUnion towards a green transition with the goal of achieving climate neutrality by 2050.

The innovation created through the development of these projects can offer a significant contribution towards achieving the European goals of ecological transition. The research conducted has led to the discovery and validation of more sustainable materials and processes, reducing environmental impact and improving resource efficiency. These advancements not only contribute to more ecological production but also demonstrate how the food packaging industry can evolve in harmony with sustainability principles.

In particular, the adoption of new biodegradable and recyclable materials, along with the optimization of production processes, represents a fundamental step towards a circular economy. This minimizes waste and valorizes production residues, transforming them into useful resources. Furthermore, the integration of innovative technologies and the implementation of best practices in the sector contribute to reducing CO_2 emissions, improving energy efficiency, and promoting a more responsible management of natural resources. The work carried out during this doctorate thus represents a concrete example of how applied research can provide practical and immediately implementable solutions to address global environmental challenges. The skills acquired and the results obtained can be used as a reference model for further developments in the sector and can positively influence other industries aiming to integrate sustainability into their operations.

In summary, the projects carried out not only meet current market needs but also anticipate future environmental directives, laying the foundation for a greener and more sustainable food packaging sector. This approach, in addition to being in line with European policies, strengthens the company's global competitiveness, while promoting a culture of sustainable innovation that can inspire similar initiatives in the industrial and academic context.

Future prospect

During my doctoral journey, I undertook and completed a research project that allowed me to deeply explore the development of innovative materials and processes aimed at creating new cellulose-based food packaging solutions within a circular economy framework. The primary objective was to create more sustainable solutions compared to current standards, reducing the environmental impact throughout the product's entire life cycle. Throughout my research, I analyzed the opportunities associated with using cellulose derivatives to enhance barrier properties and achieve sustainability goals such as compostability and recyclability of packaging. This led to the development of products like paper sticks and a new line of products made from grass paper, which represent more eco-friendly alternatives to traditional packaging materials like plastic and aluminum. A crucial element of my work was the life cycle assessment (LCA) of packaging. I conducted detailed studies on two specific products to demonstrate their eco-sustainability: Project 2 and Project 4. These analyses provided fundamental data on the environmental impact of the products throughout their life cycle, helping to identify the best strategies to reduce CO2 emissions and other negative impacts. Additionally, in the academic realm, I had the opportunity to collaborate on activities involving the functionalization of cellulosic substrates with innovative coatings, such as diamond nanoparticles (DND). The initial results of these experiments are promising and suggest potential applications in sectors different from those traditionally addressed in the company. Looking to the future, I see numerous development and application opportunities for the food packaging sector. It will be essential, if not fundamental, to expand the production of developed prototypes on an industrial scale, ensuring efficient and sustainable processes for mass production. Research, particularly in line with market trends, will continue to focus on new bio-based materials to promote solutions that offer better barrier properties and greater environmental sustainability, reducing dependency on synthetic polymers. Strengthening interdisciplinary collaborations will be crucial to accelerating innovation in sustainable packaging. Additionally, experimenting with and implementing new surface treatment and coating technologies to further improve the functional properties of cellulose-based packaging represents a promising direction. Promoting environmental awareness and adopting sustainable practices within the packaging industry and among consumers will be crucial. Therefore, it is important to have strong support for circular economy policies and contributing to achieving the Sustainable Development Goals (SDGs) represents essential steps toward a more sustainable future.

In conclusion, the work carried out during my doctorate has laid a solid foundation for developing innovative and sustainable food packaging solutions. These projects and new products pave the way for new growth and improvement opportunities within the circular economy framework. The efforts undertaken will continue to evolve, guiding the industry towards a more sustainable and environmentally responsible future.

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