biology and life sciences forum

MDPI

Proceeding Paper

Food Packaging Film Preparation: From Conventional to Biodegradable and Green Fabrication †

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† Presented at the 1st International Conference of the Red CYTED ENVABIO100 "Obtaining 100% Natural Biodegradable Films for the Food Industry", San Lorenzo, Paraguay, 14–16 November 2022.

Abstract: It is undeniable that suitable packaging will extend the shelf life of the food. The packaging industry has had to renew and innovate in a world where consumers are increasingly environmentally conscious in order to deal with the impact of the production of petroleum-derived plastics and the management of the waste generated by them. In this way, the use of biopolymers has been proposed, mainly those produced from renewable sources and with biodegradability and/or compostability properties. However, these types of materials are more expensive and do not have the same performance as petroleum-derived materials. Besides, the technologies for film preparation are not adapted for these materials. Therefore, new technologies must be studied and implemented to make the packaging industry a sustainable industry. Recently, non-solvent phase inversion (NIPS) and electrospinning techniques, which are widely used for membrane fabrication, have been proposed for the fabrication of films for food packaging applications from biopolymers and green solvents.

Keywords: biopolymers; films; green chemistry; bio-based polymers

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Citation: Ferreiro, O.B.; Monteiro, M. Food Packaging Film Preparation: From Conventional to Biodegradable and Green Fabrication. *Biol. Life Sci. Forum* **2023**, *28*, 11. https://doi.org/10.3390/blsf2023028011

Academic Editor: Mary Lopretty

Published: 14 November 2023

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1. Introduction

Packaging was identified as an important stage of the food supply chain since it is responsible for food safety by extending the life of the food and maintaining its organoleptic characteristics until the final consumer. The goal of the food packaging industry is to provide a quality product as well as safe transportation and distribution to the last stage of the supply chain (the consumption stage) at the lowest possible packaging cost [1]. Consumers have been one of the growth drivers of the food packaging market due to their interest in less processed foods as well as their awareness and expectations regarding the environmental sustainability aspects of food packaging [2–4].

The packaging material depends on many aspects, such as the type of food, industrial processing, shelf-life, the means of transport as well as the distribution and storage requirements [5]. These materials will be in contact with food, so they must be safe and special care must be taken to avoid the eventual migration of undesired constituents into the food. The development of new materials for food packaging is a challenge that concerns researchers together with the food industry to meet consumer expectations as well as avoid socioeconomic problems. Among these socioeconomic problems, it stands out that suitable food packaging could contribute to reducing food loss and waste [6]. It is estimated that by stopping food waste it is possible to save enough food to feed 2 billion hungry people [7]. On the other hand, there is a growing interest in reducing the generation of waste derived from the use of packaging, mainly those made of synthetic plastic.

Conventional plastics are typically made from petrochemical processes and are considered synthetic plastics. These plastics are usually non-biodegradable, which greatly contributes to pollution. For this reason, biodegradable materials have been used as a

substitute, mainly for single-use plastics (SUP) [5,8–10]. SUP are widely used in food industry packaging, resulting in a great volume of waste generated [8,11]. In this way, the food packaging industry, which is responsible for over 40% of plastic waste, has promoted the search for new strategies [4]. Furthermore, environmental and regulatory aspects have also contributed to the increase of new eco-friendly materials researched for food packaging towards a green economy [12]. By 2030, the European Commission expects all plastic packaging to be reusable or recyclable [13]. This will contribute to a sustainable packaging industry.

Bio-based plastics are one of the most explored strategies aimed at developing sustainable food packaging materials [3,4,9,14]. However, these materials are not necessarily biodegradable or compostable and need to be recycled. For many years, recycling has been a strategy to reduce the volume of plastic waste and has an environmental benefit since it allows to reduce the use of virgin materials in plastic production [15]. However, it is often impracticable for food packaging due to the possibility of contamination by residual products and particles [5]. Furthermore, it has been reported that recycled food packaging could not be safe due to the increase in potentially hazardous chemical levels in the packaging that could migrate into the food [16]. Biodegradable and compostable packaging are of interest when it is not possible to reduce, reuse or recycle; however, more research and development are needed to satisfy the bio-packaging market [6,17]. It is also worth mentioning that most of the costs involved in manufacturing food packaging are attributable to raw materials, mainly when bio-based plastics and biodegradable materials are considered [5,12,18,19]. This way, it has encouraged the development of technology for obtaining eco-friendly, cheaper and degradable materials from organic waste streams (such as underutilized byproducts from food processing industries and agro-industrial biomass, among others) toward a circular economy not competing with food usage of agricultural resources [6,14,20].

The food packaging industry could contribute to fulfilling some of the United Nations' Sustainable Development Goals by using renewable resources or recycled materials and implementing innovations in the design and production of biodegradable, compostable and recyclable packaging materials [21,22]. However, even meeting these requirements and necessities for being used in the food industry, the materials must be economically competitive with the conventional plastics to be viable to survive in the market and to stimulate the development of this kind of material [4,5].

2. Films for Food Packaging

Polymeric films act as barriers by controlling water vapor and atmospheric gases permeation through them and are, therefore, a popular choice for food packaging purposes [23]. The food packaging films market was worth USD 49.8 billion in 2021 and is projected to reach USD 72.3 billion by 2027 [12]. Multilayer films combine several types of plastic consisting of 3–9 layers and have become important in the food packaging for developing high-performance food packaging in a cost-effective manner [5,24,25].

The most suitable material for film preparation will depend on the product characteristics, but in any case, any undesirable migration from the film to the food must be avoided, which means that the material must be safe [26]. Generally, it is important to know the characteristics of gas permeation, such as oxygen and carbon dioxide, or the exchange rate of these gases through the film. Furthermore, it is important to also consider the permeation of moisture, flavor, and other volatile compounds as well as the selectivity of these gases or vapors. Films with a low moisture permeability are often desired [3,5].

The gas and vapor permeation will depend on the environmental conditions in which the product will be kept, such as humidity, temperature, oxygen, light (which can induce degradation reactions during storage), among others. The mechanical resistance of the film will allow safe handling and transportation of the product through the supply chain. The material must also be easy to process and have migration limits according to the regulations [5,27].

The modification in the gas atmosphere near food products during storage has a great influence on their shelf life [28]. In this way a technology known as modified atmosphere packaging (MAP) has been proposed, in which the atmosphere inside the food package is modified by altering the gas composition or by removing it (vacuum packaging). For fresh products, the goal is to create a balance within the package where the respiratory activity of a product is as low as possible by maintaining a low concentration of oxygen and/or a high concentration of carbon dioxide [29]. However, there are very few materials that are sufficiently permeable to match the respiration rate of fruits and vegetables and each material has to be optimized for specific demands [26,30]. Modifying the atmosphere of packaged foods can improve their visual appearance, texture, and nutritional appeal, and it is considered minimal processing compared to applying food chemicals, preservatives, or stabilizers since it inhibits the microbial growth [28,29].

When antioxidants, nutraceutical or antimicrobial agents, enzymes, oxygen/ethylene carriers, flavor delivery or absorption systems are incorporated into the film matrix or are applied as a coating, the system is considered as active or bioactive food packaging in which the product, package and package environment interact to provide a positive effect on the food [17,31–34]. All active packaging technologies include some physical, chemical, or biological action to generate interactions between the packaging, the product and the space left between the product and the packaging, aiming to increase the shelf life of the food by a controlled absorbing or releasing of active ingredients or by scavenging undesirable substances [9]. The active compounds can be added to or be naturally present in the raw materials used for the film preparation, and it is important to know the toxicity of these compounds, their action mechanisms and their stability [32].

3. Eco-Friendly Materials for Film Preparation

In 2021, 390.7 million tons of plastics were produced worldwide, and 90.2% of these came from fossil sources, while post-consumer recycled plastics and bio-based/bio-attributed plastics accounted for 8.3% and 1.5% of world production, respectively. This year, packaging, building and construction applications were the two largest world plastics markets. Particularly in Europe, more than 50 million tons were produced, and packaging represented 39% [35]. For packaging applications, the most commonly used polymers are low-density polyethylene (LDPE), high-density polyethylene (HDPE), polypropylene (PP), polytetrafluoroethylene (PTFE), nylon (polyamide), polyethylene terephthalate (PET), polyesters, polyvinyl chloride (PVC), polystyrene (PS) and ethylene vinyl acetate (EVA) [17,29,35].

Most of these polymers have a low cost and appropriate physical, mechanical and transport properties to be used in food packaging. However, they are petroleum-derived polymers and have low or no biodegradability [29]. Degradability is the material ability to break down into carbon dioxide, methane, water, inorganic components, and biomass, and it could be done chemically or biologically. Biodegradability is performed using microorganisms, such as fungi and bacteria, to achieve the breakdown of matter into lower molecular-weight products that can then be used by other organisms until the complete decomposition of matter. It is worth mentioning that the rate of degradation is highly dependent on the chemical structure [36].

There has been a growing interest in reducing the environmental impact of plastics; therefore, new materials have been studied for food packaging, such as bio-based polymers and biodegradable polymers. Biopolymers include these two types of polymers and are polymers that can be extracted from biomass (such as cellulose or starch), synthesized from bio-derived monomers (such as Bio-PP or polylactic acid), and produced from microorganisms (such as polyhydroxy-alkanoates) [21,29,36,37].

These materials are being designed to replace synthetic plastic materials with the goal of achieving a minimal carbon footprint, high recycling value, or complete biodegradability or compostability. Although not all bio-based polymers are inherently biodegradable, some of them could exhibit antioxidant and antimicrobial activity and biocompatibility, among

other positive effects [21,36,38]. Moreover, if these materials come from renewable waste streams or biomass sources that are not competent with food and agricultural resources, sustainable development is achieved by promoting a circular bioeconomy [14,21,36].

As previously mentioned, polymers for food packaging must be thermally stable, flexible and have a good barrier to gases and chemicals, which will depend mainly on the packaging matrix. However, most of the biopolymers used for food packaging have been reported to have poor mechanical or barrier properties toward moisture and water vapor compared to synthetic polymers [37,39]. Furthermore, while the technologies to produce synthetic plastics are widely established, the technologies to produce bioplastics lack comparable scalability and productivity. These factors have delayed the widening applicability of biopolymers in food packaging [21,37].

The food packaging industry has been working on innovative solutions for improving the barrier performance, mechanical strength, and thermal stability of the packaging and, consequently, extending the food shelf life [31,40–42]. Among the strategies to improve the characteristics of biodegradable polymers have been suggested: the use of nanotechnology by the incorporation of nanofillers such as nanoparticles to modify or control the permeability or the release of active ingredients or to provide antioxidant, antibacterial, antifungal or antimicrobial properties [9,31,33,37,42–44]; the application of a surface treatment such as coating using a good film-forming [41,45] or inducing a crosslinking [46]; and the blending of biopolymers, which should be compatible [5,47].

Among the biopolymers most reported in the literature with a great potential to substitute synthetic polymers in the food packaging industry can be cited: starch, chitosan, polylactic acid (PLA), and polyhydroxyalkanoates (PHA) [5,48,49]. PLA is the biopolymer with the greatest potential to replace petroleum-based polymers (such as polystyrene (PS) and polypropylene (PP)) in packaging applications due to its excellent barrier properties [38,39]. Oriented PLA (OPLA) showed to be a good film for tomatoes and other breathable products because of the matching of the oxygen and carbon dioxide exchange with the respiration rate of these products [27]. Table 1 summarizes some of the advantages and limitations for the biopolymers more commonly used and reported in the literature.

Table 1. Advantages and limitations of biopolymers for food packaging applications.

Biopolymer	Advantages	Limitations	Ref
Chitosan	• Availability • Nontoxic • Easy to form films • Selectivity to carbon dioxide • Oxygen permeability • Antibacterial, antifungal, and mechanical properties	• High sensitivity to water afecting its mechanical stability	[5,50]
Starch	• Low Price • Zero toxicity • High degradability • Easy availability • Already being commercialized	• Poor resistance to humidity • Poor thermal processability • Poor mechanical resistance • It is not stable to heat	[51]
PLA	• The existing technology for the manufacture of films can be used • It has properties similar to polymers of fossil origin (oxygen transfer; strength and stability)	• Fragile	[9,38]

Table 1. *Cont.*

Biopolymer	Advantages	Limitations	Ref
PHA	• Not toxic • Insoluble in water • Biodegradable/Biocompatible/Renewable • Good UV resistance • High tensile strength	• Cost of raw material when pure glucose is used • Low barrier properties	[9,44,52]

4. Films Preparation Techniques

Although biopolymers do not present the same performance for food packaging applications, another factor that has limited their widening application is that the current film preparation techniques are not always suitable for these materials. Among the techniques for synthetic polymers that have been tested for biopolymers is blown film extrusion. This technique has undergone extensive industrial development for synthetic polymers. However, it presents some limitations for biopolymers, as will be discussed below. Solution casting is the most reported technique for biopolymer film preparation, although this technique is difficult to implement on an industrial scale.

On the other hand, membrane preparation technology for separation processes is quite developed. In this way, it is possible to take advantage of this area to design biopolymer-based food packaging films. Non-solvent-induced phase inversion (NIPS) is a conventional method for fabricating polymeric membranes, while electrospinning is an emergent technique. Both techniques could be used to prepare films for food packaging.

The blown-extrusion technique allows for the production of polymeric films of a variety of thicknesses on a large scale due to its low cost, continuity and simplicity of operation. Multilayer films can also be produced by this technique. PLA films were successfully produced by this technique by adding nanoparticles of MgO, which enhanced the film plasticity [43]. Karkhanis et al. [53] prepared transparent PLA films containing cellulose nanocrystals with high potential to be used for food packaging due to the enhanced barrier performance obtained. Bilayer biodegradable films were also prepared by the co-extrusion of PLA and Bio-flex® in blown-extruder equipment, showing that it is possible to obtain multilayer films in a single step [54].

However, blown-extrusion processing requires a high melt viscosity resin, which limits the application of other biopolymers different from PLA, and it is necessary to blend the biopolymers with other polymers to improve their processability by modifying the rheological properties of the blend [5,29,51].

Solution casting is the most popular technique to prepare biopolymeric films on a laboratory scale because of its simplicity [5,51]. Besides, this technique allows for a better crosslinking between two or more blended polymers [45]. However, this technique has a high energy consumption for solvent evaporation (commonly, water), which has hindered its expansion to an industrial scale [51]. Furthermore, the incorporation of nanoparticles into the film is also limited due to the difficulties related to their uniform dispersion in the film [5]. This could be overcome using tip sonication; this way, Manikandan, Pakshirajan and Pugazhenthi [44] prepared PHA films with graphene nanoplatelets, which were uniformly distributed in the PHA matrix. On the other hand, Ochoa-Yepes et al. [55] reported better mechanical and lower moisture content and water vapor permeability for the starch films prepared by the extrusion/thermocompression process than the ones prepared by solution casting.

The NIPS technique for the preparation of biodegradable films has recently proposed by Liu et al. [56]. The authors prepared PLA films by NIPS for pork meat packaging to extend the shelf life of the food. Although the authors used N-methylpyrrolidone as a solvent, green solvents could be explored as an alternative to a sustainable film preparation. NIPS is a technique widely used for polymeric membrane preparation. The membrane industry is also looking for eco-friendly alternatives toward a sustainable membrane fabri-

cation process. In this way, biopolymers and green solvents have been proposed [57,58]. On the other hand, electrospinning technology has been widely investigated for the fabrication of nanofibrous membranes for water treatment [59] and more recently for the fabrication of food packaging materials [60]. PLA-based nanomaterials as well as active and intelligent packaging materials have been fabricated for food packaging applications by electrospinning with expected structures and enhanced barrier, mechanical, and thermal properties [39].

5. Final Considerations

The food packaging film industry is directed towards sustainable development; for this reason, it requires raw materials obtained from renewable or recycled sources, more efficient and green production, and proper waste management.

Author Contributions: All authors contributed equally. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Red Cyted ENVABIO100 121RT0108.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data sharing is not applicable to this article.

Conflicts of Interest: The authors declare no conflict of interest.

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