

BIOPOLYMERS, THE GREEN ALTERNATIVE TO CONVENTIONAL PLASTICS: A  
SCIENTIFIC-TECNOLOGICAL SURVEILLANCE STUDY

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### **Dedicatoria**

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## Table of Contents

Introduction .....	9
1. Objectives .....	11
1.1 General Objective .....	11
1.2 Specific Objectives .....	11
2. Methodology .....	12
2.1 Source selection, search equation and window frame .....	13
2.2 Search, selection and organizing information .....	13
2.3 Bibliometric Analysis .....	13
2.4 Identification of advances.....	13
2.5 Assessing challenges and research gaps .....	13
3. Result Analysis .....	14
3.1 Selection of information – PRISMA protocol.....	14
3.2 Bibliometric analysis .....	16
3.3 Scientific advances .....	17
3.3.1 Chitosan .....	18
3.3.2 PLA .....	22
3.3.3 Cellulose .....	24
3.3.4 Starch .....	26
3.4 Scientific improvements .....	27
3.4.1 Nanoparticles .....	28
3.4.2 Nanoclay and nanotubes .....	29
3.4.3 Natural Fibers.....	30
3.4.4 Crosslinking .....	30
3.4.5 Plasticizers .....	31
3.5 Future challenges .....	32
3.5.1 Life Cycle Assessments .....	32
3.5.2 Unintended Green House Gases (GHG) emissions .....	33
3.5.3 Biodegradation of bioplastics .....	33
3.5.4 Real food applications.....	33
4. Conclusions.....	34
5. Recommendations.....	35

6. Bibliography .....	36
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### **List of Tables**

Table 1 Number of results while narrowing the equation search.....	14
Table 2 Biopolymer fabrication from Chitosan .....	20
Table 3 Biopolymer fabrication from PLA .....	22
Table 4 Biopolymer fabrication from Cellulose .....	25
Table 5 Biopolymer fabrication from Starch .....	27

### List of Figures

Figure 1 General methodological scheme developed during the research work.....	12
Figure 2 Process of exclusion .....	15
Figure 3 Trend through the years 2001-June 2021 of publication of articles related to the use biopolymers in the food packaging industry .....	16
Figure 4 Countries with the highest number of publications on the study field .....	17
Figure 5 Network of keywords.....	18
Figure 6 Structure of Chitosan .....	19
Figure 7 Structure of PLA .....	22
Figure 8 Structure of Cellulose .....	24
Figure 9 Structure of Starch .....	26

## Resumen

**Título:** Biopolymers, the green alternative to conventional plastics: Scientific-Technological Surveillance Study\*

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**Palabras clave:** biopolímero, bioplástico, biopelícula, revisión, alternativa verde

**Descripción:** Los plásticos convencionales se fabrican y utilizan para aplicaciones de embalaje en diferentes sectores. A medida que aumentan las industrias alimentarias, también aumenta la demanda de material de embalaje. En consecuencia, la atención a los plásticos amigables con el medio ambiente, como los biopolímeros, también ha aumentado drásticamente. Sin embargo, los biopolímeros generalmente presentan malas propiedades mecánicas y poca durabilidad a largo plazo. En este sentido, es de gran interés conocer los avances científicos y tecnológicos relacionados con el desarrollo de biopolímeros por su alto potencial para sustituir a los polímeros tradicionales y reducir la contaminación y el consumo energético. Esta monografía considera un análisis bibliométrico de publicaciones científicas relacionadas con los avances y desafíos de los biopolímeros realizados entre 2001 y junio de 2021. La información recopilada se clasificó por fuente del biopolímero, así como las mejoras en las propiedades mecánicas. Se destacaron los avances más significativos del quitosano, PLA, celulosa y almidón, así como las desventajas que cada uno conlleva. En las mejoras se mencionaron nanocompuestos, fibras naturales, plastificantes y nanotubos y como estos reforzaron las propiedades mecánicas de los biopolímeros. Además, se discuten los desafíos futuros en el campo, proponiendo estudios que aún no se han realizado, así como el refuerzo de los que necesitan más investigación, principalmente relacionados con los Análisis de Ciclo de Vida (LCA).

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### Abstract

**Title:** Biopolymers, the green alternative to conventional plastics: Scientific-Technological Surveillance Study\*

**Author:** Silvia Valentina León Acero\*\*

**Keywords:** biopolymer, bioplastic, biofilm, review, green alternative

**Description:** Conventional plastics are manufactured and used for packaging applications in different sectors. As the food industries are increasing, the demand for packaging material is also increasing. Consequently, the attention to environmentally friendly plastics such as biopolymers has increased dramatically as well. However, biopolymers generally present poor mechanical properties and poor long-term durability. With this matter on mind, it is of great interest to know the scientific and technological advances related to the development of biopolymers due to their high potential to replace traditional polymers and reduce pollution and energy consumption. This monograph considers a bibliometric analysis of scientific publications related to biopolymer advances and challenges carried out between 2001 and June 2021. The information collected was classified by source of the biopolymer, as well as the improvements in the mechanical properties. The most significant advances of chitosan, PLA, cellulose, and starch were highlighted as well as the disadvantages that each entailed. In the improvements, nanocomposites, natural fibers, plasticizers, and nanotubes were mentioned and how did they reinforce the mechanical properties of biopolymers. Furthermore, the future challenges in the field are discussed, proposing studies that have not been done yet as well as the reinforcement of the ones that need further investigation, mainly related to Life Cycle Assessments (LCA).

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## Introduction

Plastic has evolved along with society. It has revolutionized our daily lives bringing practicality, convenience, and safety to tasks that have now become simple (Santos *et al.*, 2021). Particularly, single use plastics, also often referred to as disposable plastics, has been widely used for packaging to protect food from chemical and biological contaminations and physical damage by preserving its quality and safety (Patrício Silva *et al.*, 2021). The list of disposable plastic includes items such as grocery bags, food containers, bottles, straws, containers, glasses, and silverware.

However, some of the features that make synthetic plastics commercially successful- its prices, durability, and resistance – also contributes in a harmful way to the environment, especially when poorly managed. Their non-biodegradable nature causes negative impact on both land and marine ecosystems due to the fact that it takes hundreds of years for these polymers to breakdown into innocuous soil components, it being one of the most global concern (Siva *et al.*, 2021). In this regard, the food packaging plastics are mostly single used representing the 36% of the total world plastic production in the industrial sector during 2015, and most of these disposable plastics are discarded the same year they were produced (United Nations Environment Programme, 2018).

With this matter in mind, different studies had been focusing on the development of green alternatives of conventional plastic, paying special attention to biopolymers. Biopolymers are made of renewable biodegradable materials. Their biodegradability is mainly the result of the presence of oxygen and nitrogen atoms available in their chemical structure. They can be divided into three categories depending on their raw materials and their ease biodegradability: 1) biopolymers that are made from renewable raw materials (bio-based) and are biodegradable

(biodegradable bio-based biopolymers); 2) biopolymers that are made from renewable raw materials (bio-based) and are not biodegradable (non-biodegradable bio-based biopolymers); and 3) biopolymers that are made from fossil fuels and are biodegradable (fossil-based biodegradable)(Khademian *et al.*, 2020).

However, biopolymers have yet to be studied in every single aspect with the aim to meet all the expectations a food packaging requires (Alamri *et al*, 2021). Two of the most important aspects are their lower mechanical properties and their higher price in the production in comparison to synthetic polymers. The latter has limited the market and occasionally petroleum plastic blending has been necessary. Also, a careful selection is required to avoid negative effects on the quality, safety, and shelf stability of products.

Based on the above premises, the objective of the present work was to recap the research made in the biopolymer field and more specifically on the food packaging context during the last 20 years. The information was classified using the PRISMA methodology. The scientific and technological advances in the production of biopolymers is presented based on analysis and comparison of works published in the data bases with major influences. Finally, the challenges in the field and the research gaps for future studies are also discussed to contribute for a better and promising future.

## **1. Objectives**

### **1.1 General Objective**

To develop a scientific-technological surveillance study related to current challenges in the production of biopolymers in the food packaging market.

### **1.2 Specific Objectives**

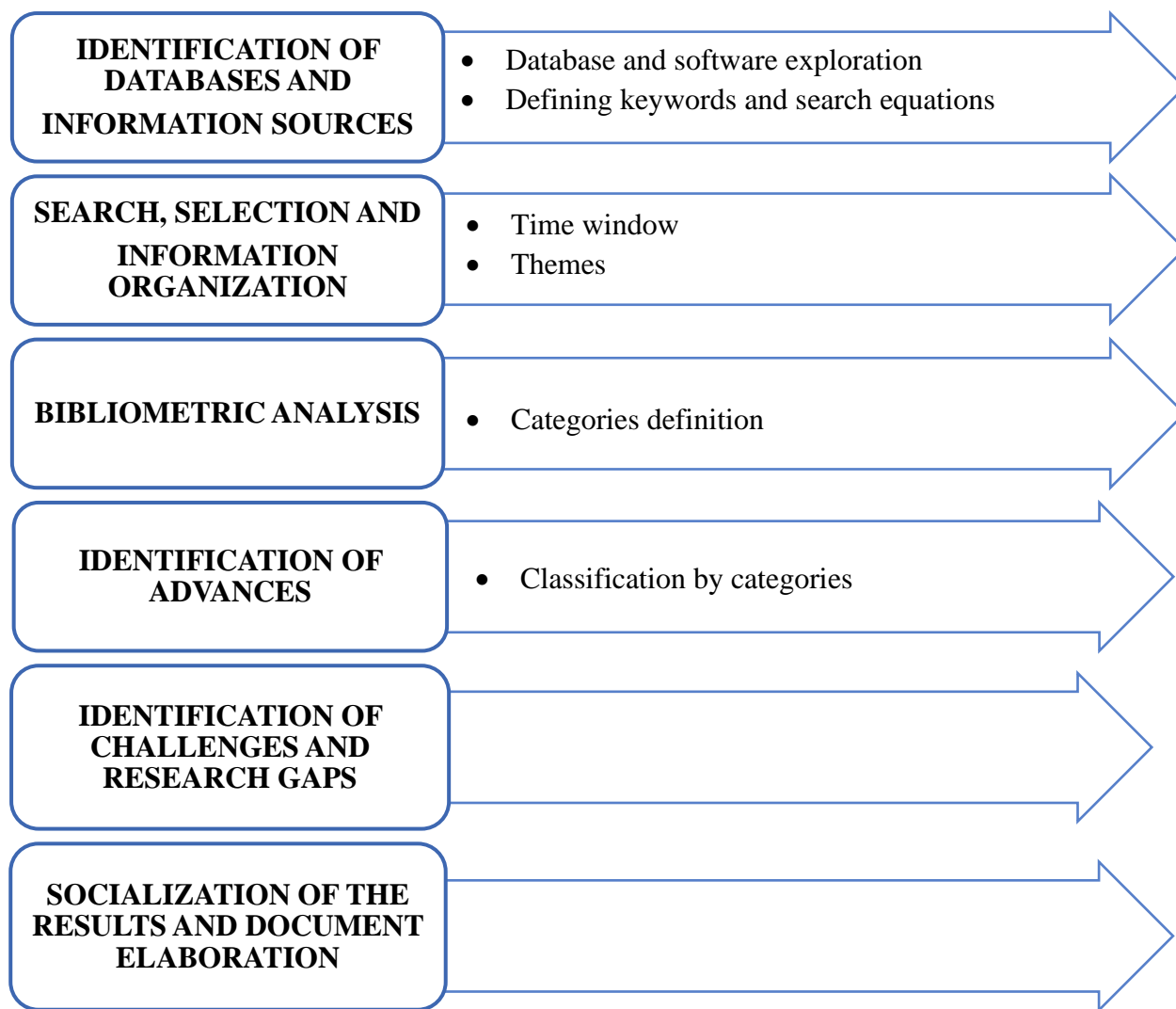
- To develop a bibliometric analysis of the production of biopolymers obtained from renewable sources, establishing the countries, authors, and research groups with the greatest contribution on the subject associated with food packaging.
- To analyze relevant scientific and technological advances in the production of biopolymers, based on the analysis and comparison of works published in indexed journals.
- To assess the challenges and gaps in the field of study that may serve as basis for future proposals of the research group.

## 2. Methodology

The following figure describes the general scheme of the methodology applied to this work.

**Figure 1**

*General methodological scheme developed during the research work.*



## **2.1 Source selection, search equation and window frame**

The information sources employed in this study were Web of Science (WOS), Scopus and Taylor & Francis platforms. Several search equations were created by considering different keywords and search operators in order to retrieve a major number of documents as possible. The window frame selected was from 2001 up to June 2021.

## **2.2 Search, selection and organizing information**

Once the search equation and the selected time window were established, the search of the investigation was undertaken from the three data bases and exported into Rayyan software, which is designed to help researchers working on systematic reviews and other knowledge synthesis projects. The records obtained were classified according to different thematic categories.

## **2.3 Bibliometric Analysis**

With the results of the search, a bibliometric analysis was carried out to classify the research according to: year of publication, country and common keywords in each publication.

## **2.4 Identification of advances**

After classifying the information through the different topic categories, the most relevant scientific advances associated were identified. The classification was made using VOSviewer which is a free software tool for constructing and visualizing bibliometric networks

## **2.5 Assessing challenges and research gaps**

Finally, the main challenges and research gaps faced by the production of biopolymers in the food packaging industry were defined which will serve as a basis for future research proposals.

### 3. Result Analysis

#### 3.1 Selection of information – PRISMA protocol

Table 1 comprises the different search equations that were applied for a selected window frame between 2001 until the first semester of 2021. The last search was made on June 4, 2021.

The first equation in Table 1 was not very useful since the keyword “packaging” was pulling up documents related to any kind of packaging but not specifically “food packaging”. The second equation, which included the term “food packaging” in any field, showed almost every article related to another subject rather than the one it was really aimed for. On the other hand, equations 3 and 4 considered “bioplastic” instead of “biopolymer”, while equation 5 and 6 replaced “biopolymer” by “biofilm”.

**Table 1**

*Number of results while narrowing the equation search*

Keywords / Source	Number of Results				Number of Equation
	Scopus	WOS	Taylor & Francis	Total	
<b>TITLE: biopolymer KEYWORD: packaging</b>	810	913	14	1,737	1
<b>TITLE: biopolymer ALL: food packaging</b>	450	573	1,711	2,734	2
<b>TITLE: bioplastic KEYWORD: food packaging</b>	50	82	212	344	3
<b>TITLE: bioplastic KEYWORD: packaging</b>	123	150	273	546	4
<b>TITLE: biofilm KEYWORD: packaging</b>	43	9	5	57	5
<b>TITLE: biofilm KEYWORD: food packaging</b>	20	22	0	42	6
<b>TITLE: biopolymer OR bioplastic OR biofilm KEYWORD: food packaging</b>	<b>104</b>	<b>101</b>	<b>3</b>	<b>208</b>	7

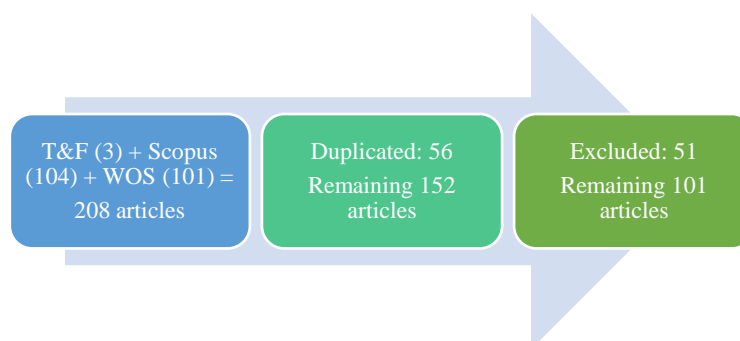
On the other hand, the employment of any other keyword such as “chitosan”, “cellulose”, “scratch” was too specific and narrowed the equation up to a very explicit subject.

Table 1 presents the seven different search equations that were applied and highlighted the selected one for the present study. Equation 7 is the final equation, in which the OR operator was included since equation 1 and 2 vs equation 3 and 4 used different terminology to refer to biopolymer and excluded articles that used terms such as biofilm or bioplastic. It should be mentioned that, although the chosen equation was not the one showing the highest number of results, it presented the best results in terms the of the subject-related research works.

Equation 7 was plugged in “Scopus” with a yield of hundred and four (104) results, “Web of Science” with a hundred and one (101) results and “Taylor and Francis” with three (3) results adding to a total of 208 articles. After identifying the duplicate articles using Rayyan, fifty-six (56) articles were deleted. With the fifty-two (152) remaining, accordingly, 51 pieces of information were excluded because they: i) were not related to the subject; ii) present a different study field; iii) or did not provide useful information to have any discussion about it. After this refining process, a hundred and one (101) pieces of information were considered for the final revision and analysis shown in Figure 2.

## Figure 2

### *Process of exclusion*

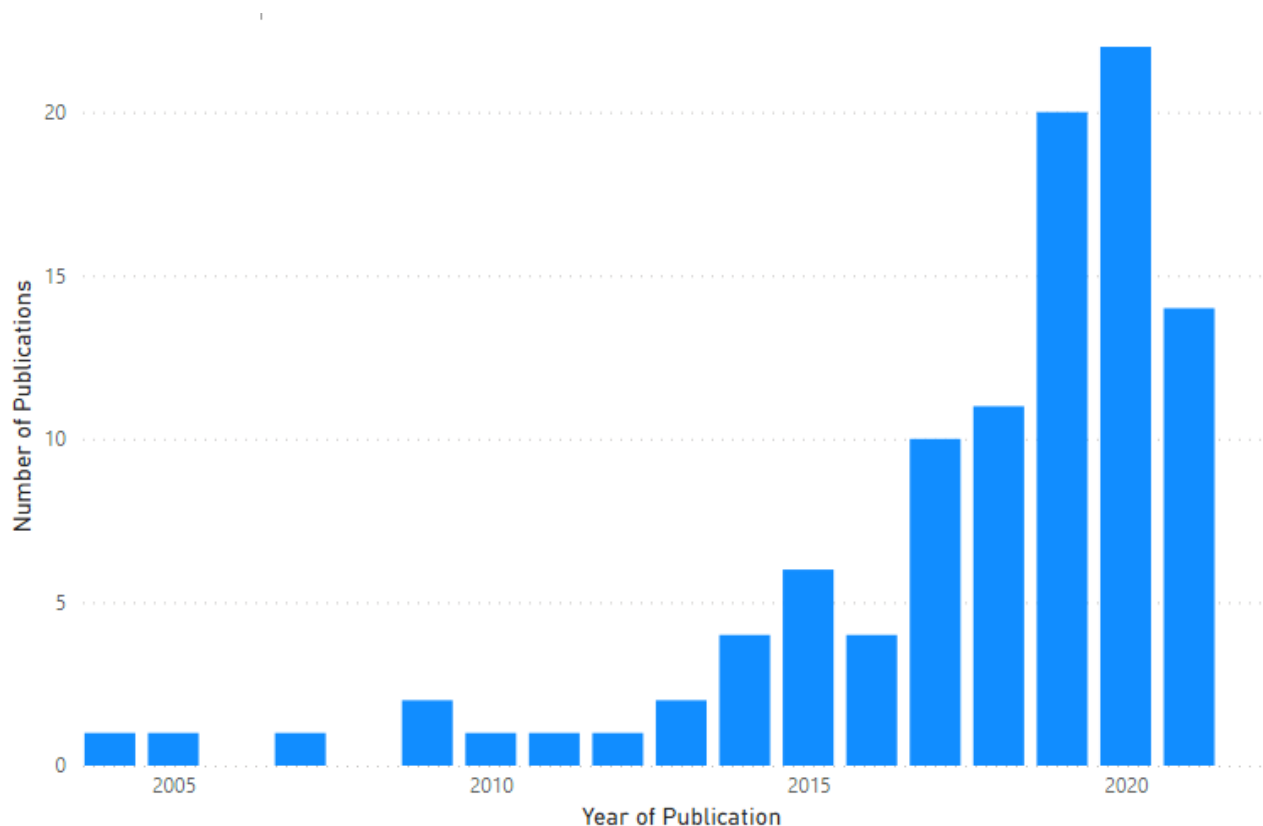


### 3.2 Bibliometric analysis

Figure 3 displays the distribution of publications in the time window analyzed. According to this figure there is an evident growing interest of this type of research. In this regard, the intellectual productivity is concentrated in the last 5 years representing 76.2% of the total of articles published since 2001. From Figure 3 it follows that the last 3 years 2019, 2020 and 2021 are the years with the highest number of publications, even though for 2021 only the articles published until June were taken into account.

**Figure 3**

*Trend through the years 2001-June 2021 of publication of articles related to the use biopolymers in the food packaging industry.*



On the other hand, the production of articles by country is shown in Figure 4. The country with the highest number of publications is India with 16 articles, which represents 11.3% of the

total records, followed by United States of America (USA) with 12 articles, 8.5%, and the Malaysia and Indonesia both with 8 articles, 5.6%.

**Figure 4**

*Countries with the highest number of publications on the study field.*



It is important to notate that, regarding the authors with major publications on the subject, no relevant correlation was found. The latter is related to the fact that “biopolymer in food packaging” is still a growing topic in which different authors are researching in.

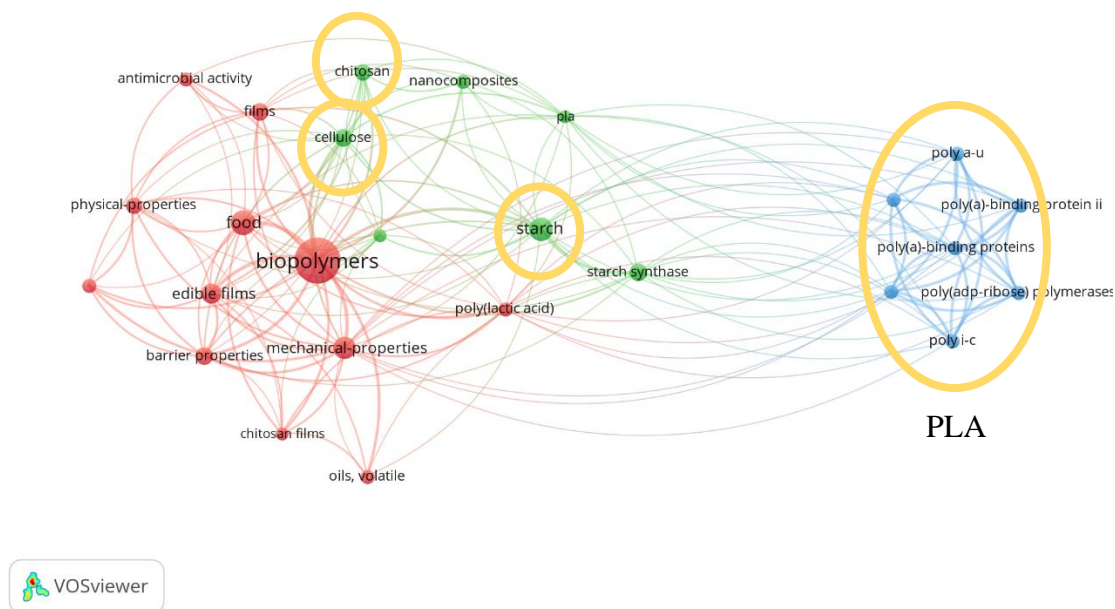
### 3.3 Scientific advances

Figure 5 displays the most common keywords found in all the documents and how they are related to each other. The size of the word represented how often the keyword was found,

the bigger the size, more times the keyword appeared. In this case, the categories designated were the following: chitosan, polylactic acid (PLA), cellulose, and starch.

**Figure 5**

*Network of keywords*



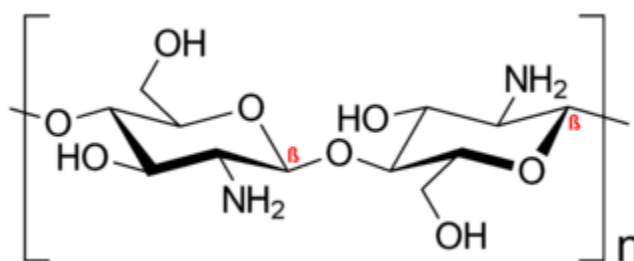
### 3.3.1 Chitosan

Chitosan (CS) is a natural polymer, widely used in applications in the fields of pharmacology, biomaterials technology, biomedicine, agriculture, and cosmetics and food industries due to its good biodegradability, biocompatibility, and antimicrobial and mechanical properties (Santos *et al.*, 2021). In this regard, chitosan (CS) has been one of the most common materials found in the formulation of biodegradable packaging together with polysaccharides, proteins, and lipids attributed to its fine film forming ability with reasonable mechanical strength (Díaz-Montes & Castro-Muñoz, 2021).

CS is a linear macromolecular polysaccharide, as shown in Figure 6, which is derived from the abundant chitin in the shell of crab and shrimp (Joseph *et al.*, 2021). It can also be obtained from the cell wall of some fungi (Miteluț *et al.*, s. f.). CS is composed mainly of repeating units of D-glucosamine and N-acetyl-D-glucosamine linked through  $\beta$ -(1–4) bond. Its structure is very similar to cellulose since the only difference is the amine group ( $\text{NH}_2$ ) on the position C-2 instead of the hydroxyl group ( $\text{OH}$ ) found in cellulose (Agusnar *et al.*, 2018).

**Figure 6**

*Structure of Chitosan*



*Note.* Adapted from Qniemiec, 2020.

Regarding the CS properties, the antimicrobial properties are attributed to its cationic nature that interacts with the negatively charged microbial cell surface, hence resulting in its membrane damage (Ilk *et al.*, 2018). On the other hand, its solubility depends on the degree of deacetylation, the distribution of acetyl groups along the main chain, the molecular weight and the nature of the acid used for protonation. Moreover, its molecular weight can also affect the properties of the final film such as elasticity or brittleness (Miteluț *et al.*, s. f.). CS has been studied by adding other materials such as biopolymers, polysaccharides, fatty acid and essential oils or mixtures to produce more competitive biopolymers (Agusnar *et al.*, 2018).

The results of the three data bases regarding the use of chitosan are summarized on Table 2. According to Table 2 the pursued results are an increased tensile strength, and elevation of water vapor and oxygen barrier properties, however it was not the case in a few documents.

**Table 2***Biopolymer fabrication from Chitosan*

Film	Addition	Result	Reference
Chitosan	Gelatin	Increased tensile strength and decrease water vapor and oxygen barrier properties.	Benbettaïeb <i>et al</i> , 2014
Chitosan	Garlic, black pepper, caraway and cinnamon.	Decreased tensile strength and elongation at break	Hromiš <i>et al</i> , 2016
Chitosan	Cellulose nanofibrils (CNFs) and corn starch	Improve food quality and safety reinforcing oxygen and water vapor barrier properties.	Yu <i>et al</i> , 2017
Chitosan	Patchouli Oil	Increased tensile strength, moisture content ability and thermal stability.	Agusnar <i>et al</i> , 2018
Chitosan	Octadecylamine-Montmorillonite NC Nigella arvensis Extract	Significant inhibition against the human and food-related bacteria was achieved	Ilk <i>et al</i> , 2018
Chitosan, pectin, and starch	Rosemary and mint essential oil, nisin and lactic acid	Increased film microstructure heterogeneity and reduced water barrier properties and tensile strength of the films while improving their flexibility.	Akhter <i>et al</i> , 2019
Chitosan and potato starch	Critic Acid	Improved water resistance properties and enhanced mechanical and antimicrobial properties	Wu <i>et al</i> , 2019
Chitosan and Poly(3-hydroxybutyrate-co-3-hydroxyvalerate (PHBV)	ZnO- Ag NC	Improved thermal stability and tensile strength	Zare <i>et al</i> , 2019
Chitosan, potato protein and linseed oil	ZnO NPs	Considerable mechanical properties, acceptable moisture barrier capability and good compatibility.	Wang <i>et al</i> , 2020
Chitosan	Nano-liposomal garlic essential oil (NLGEO)	Thickness, water- solubility, elongation at break, some microstructural properties and antioxidant activity of films containing NLGEO improved.	Kamkar <i>et al</i> , 2021
Chitosan	Methylcellulose/PEG/chitosan NP	Enhanced mechanical and barrier characteristics, high antibacterial, antifungal, antioxidant and UV-protective activity and serve as time-temperature indicators.	Kritchenkov <i>et al</i> , 2021
Carboxymethylcellulose	Chitosan and turmeric	Improvement in the elasticity of the film, good barrier properties (WVP)	Santos <i>et al</i> , 2021

Chitosan have been used in two different ways, as the biopolymer film or as a nanoparticle (NP) addition, both with the aimed to improve mechanical properties. As a film, CS have been studied with different additions in order to enhanced mechanical properties. Unfortunately, there were some studies that did not achieve the expected result. In this regard, Hromiš *et al.*, (s. f.) demonstrated that with the addition of spice oleoresins the tensile strength decreased contrarily to what expected. Likewise, Akhter *et al.*, (2019) studied the addition of rosemary, mint, nisin and lactic acid resulting in a reduction of water barrier properties and tensile strength.

By contrast, İlk *et al.*, (2018) introduced organo-clays fillers as surface-modifiers with organic surfactants by in situ intercalation process. ODA-MMT is one of commercial organo-clay derivate and widely used in producing nanocomposites. It can be used as a carrier of natural active substances (antioxidant and antimicrobial) which could contribute to the enhancement of shelf life and offer a more control release of any food ingredient. With this effort in mind, the article exposes the first report of chitosan/ODA-MMT/black cumin nanocomposite that exhibits significant antibacterial effect against human and food-related bacteria. In addition, the crosslinking technique is a promising procedure because it helps improved film water sensitivity but its employment has been limited due to the high cost and cytotoxicity. Accordingly, Wu *et al.*, (2019) presented an interesting crosslinking route employing citric acid that provided an appropriate formula to crosslink the polymer structure with low cost and a non-toxic nature.

On the other hand, chitosan has been used as a NP addition due to the need of research in active or smart packaging. Nanoparticles have smart or active properties, which can also act as mechanical reinforcement of the food films and coatings. This was the purpose of Kritchenkov *et al.*, (2021) and Santos *et al.*, (2021). From Kritchenkov *et al* the addition of CS NPs into a chitosan film resulted in enhanced mechanical and barrier characteristics and high antibacterial

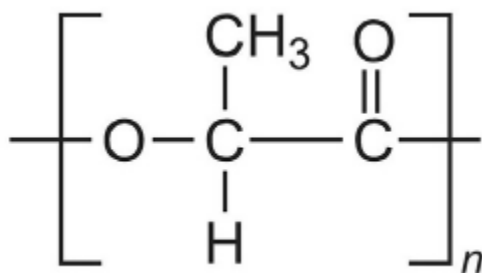
properties. Moreover, Santos *et al* presented the increase in elasticity of the carboxymethylcellulose film, in addition to making it more rigid, both characteristics that are favorable for the use as edible packaging.

### 3.3.2 PLA

Poly (lactic acid) (PLA) is a compostable bioplastic manufactured by the polymerization of lactic acid monomers derived from the fermentation of starch (sugar, corn, sugarcane, or tapioca) as a feedstock (Swaroop and Shukla, 2018). Its structure is display in Figure 7.

**Figure 7**

*Structure of PLA*



*Note.* Adapted from Karamanlioglu *et al*, 2017.

PLA has gained importance due to its mechanical properties that are similar to the petrochemical based plastics polystyrene (PS) and polyethylene terephthalate (PET) (Auras *et al.*, 2003). Different approaches have been made regarding PLA to overcome the few drawbacks that it can present, such as addition of plasticizers, polymer blending, and nanotechnology (Table 3).

**Table 3**

*Biopolymer fabrication from PLA*

Source	Result	Reference
PLA + PHBV + PBAT	Low thermal stability and poor ductility, intermediate mechanical and barrier performance	Quiles-Carrillo <i>et al</i> , 2018
PLA + MgO (NP)	Improved tensile and gas barrier properties, antibacterial efficacy, and UV screening ability	Swaroop and Shukla, 2018

<b>Starch + PLA</b>	Improved mechanical and water barrier properties	Abdullah <i>et al</i> , 2019
<b>PLA + crude palm oil with starch</b>	Good tensile strength and poor elongation at break	Krishnamurthy and Amritkumar, 2019

Quiles-Carrillo *et al* (2018) and Abdullah *et al.*, (2019) developed a ternary blend PLA+PHBV+BAT highlighting that the one that shared similarities with conventional plastics (PET) was the blend with the highest content of PHBV. Krishnamurthy & Amritkumar, (2019) presented a significant advance in the addition of glycerol as a plasticizer blended with another biopolymer, starch. This study showed that the elongation at break (EAB) passed from 3.49% (without glycerol) to 73.38% (with glycerol), which is a notable change in this property. At last, Swaroop and Shukla were the first study that included MgO NP with PLA resulting in an improvement in tensile and gas barrier properties, as well as in antibacterial efficacy, and UV screening ability. Due to this, PLA with MgO Np, can be considered as promising material for food packaging applications.

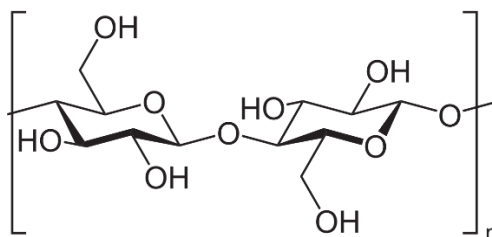
Despite the good results in improving mechanical properties, there is a growing concern about PLA contamination in the environment. It is crucial for future studies to also focus their efforts in solving PLA degradation issues that are mainly influenced by pH and UV light. In this regard, it degrades faster in alkaline conditions because during hydrolysis, cleavage of ester groups is catalyzed by hydroxide ions, therefore, the high concentration of hydroxide ions in alkaline media enhances PLA degradation. In addition to the contamination, some toxicological studies on PLA reveal that there might be instances of lactic acid release into the food. Even though a release of a limited amount of lactic acid does not create any harm to human body, it may create some complications to individual's health, especially people who are intolerant to lactic acid (Rajesh *et al*, 2020).

### 3.3.3 Cellulose

Cellulose is one of the most abundant polymers on earth, which can be easily obtained from the cell wall of plants. The structure of cellulose (Figure 8) is very similar to chitosan one's. Cellulose is a low-cost material carrying suitable mechanical properties. However, cellulose and its derivatives have various limitations, such as high-water absorption capacity and insufficient interfacial adhesion, hence it has been studied and discussed in research for many years (Y. Liu *et al.*, 2021).

**Figure 8**

*Structure of Cellulose*



*Note.* Adapted from Neurotiker, 2007.

Cellulose is used in different types, as fiber, nanocrystal, nanofibrils, as an acetate or even a more specific type of cellulose, *i.e.* carboxymethylcellulose (CMC). CMC is a cellulose derivative with carboxymethyl substituents ( $-\text{CH}_2\text{COOH}$ ) bound to some of the hydroxyl groups of the cellulose (Choi *et al.*, 2017).

Looking at Table 4, Han *et al.*, (2015) proposed an interesting combination of CMC and soy protein trying to compensate the weak parts of each source. Studies have shown that films made from cellulose derivatives have the tendency to be stiff but brittle, while films composed of proteins are flexible but weak. Therefore, the mixture of both cellulose derivative (CMC) and protein (soy protein isolate) resulted in an improved film with combined properties to be used in food

packaging. Choi *et al.*, (2017) stated that the CMC addition is helpful to improve the film strength and the gas barrier property due to the formation of the film network structure with high polysaccharide and protein contents and the firmly linked chemical bonds, respectively. In addition, Rodsamran & Sothornvit, (2017) synthesized CMC from rice stubble (CMCr), which can successfully replace to the commercial one (CMCc) up to 50% to form a blended film.

**Table 4***Biopolymer fabrication from Cellulose*

Type	Addition	Result	Reference
<b>Carboxymethylcellulose (CMC)</b>	Soy protein + CMC + catechin	Lower water vapor (WVP) and oxygen permeability. Increase in TS and a decrease in %E of the films.	Han <i>et al</i> , 2015
<b>CMC</b>	apple skin powder + CMC + apple skin extract and tartaric acid	WVP and tensile strength decreased and the elongation at break increased	Choi <i>et al</i> , 2017
<b>CMC</b>	Rice Stubble + CMC + plasticizers	Acceptable water barrier and extended the shelf life of food products	Rodsamran and Sothornvit, 2017
<b>Nanofibrils</b>	CNFs + corn starch, + chitosan	Increased the rigidity and opacity of the films	Yu <i>et al</i> , 2017
<b>Cellulose acetate (CA)</b>	poly-carbonate (PPC) + CA + oregano waste	High UV protection and antioxidant activity	Nga <i>et al</i> , 2020
<b>Nanocrystal (NC)</b>	CNC-R + PHBV	Improved strength/toughness, thermal stability, melting temperature, and barrier/migration ability	Li <i>et al</i> , 2020
<b>Fiber</b>	Cellulose fiber + fungal fiber	Significant effect on the mechanical strength + increased air permeability.	Irbe <i>et al</i> , 2021
<b>CMC</b>	CMC+ Turmeric + NP of chitosan	Improved elasticity of the film, in addition to making it more rigid.	Santos <i>et al</i> , 2021

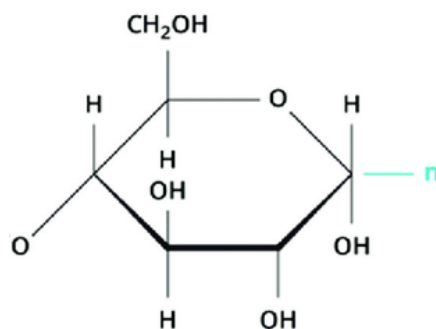
Even though most of the cellulose studies presented enhancement in the mechanical properties there is a need for an application test in order for determining its industrial feasibility.

### 3.3.4 Starch

Starch is a renewable biopolymer that consists of amylose and amylopectin. Starch is also the most commonly used agricultural raw material for edible film manufacturing because its inexpensive sources, its relatively easy to handle, its biodegradability. It is widely available in nature from various sources, such as cereals, roots, tubers, others (Pagno *et al.*, 2015). Figure 9 exposed the structure of starch.

**Figure 9**

*Structure of Starch*



*Note.* Othman *et al.*, 2018.

Starch-based bioplastics have been studied recently with different sources such as corn starch, potato starch, cassava starch, rice starch, canna starch, etc. However, the use of starch has a main disadvantage that is its hydrophilic characteristic, which will decrease its properties as bioplastic. Therefore, the addition of other ingredients is needed to improve typical starch-based bioplastic (Table 5) (Abdullah *et al.*, 2019). One relevant study to highlight was carried out by Pagno *et al.*, (2015). They added gold NP as microbial agents with promising results since gold NP's enhanced the tensile strength and increased the UV radiation absorption.

On the other hand, one aspect to improve as mentioned by Mohan *et al.*, (2017) is that, although there are several studies based on the effect of nano clay on thermal and mechanical

properties of biopolymer, its influence on biocompatibility and degradation behavior has not been studied in detail yet.

**Table 5**

*Biopolymer fabrication from Starch*

Source	Addition	Result	Reference
<b>Quinoa</b>	Gold NP	Strong antibacterial activity against food pathogens, enhanced the tensile strength, increased UV radiation absorption and a decreased solubility.	Pagno <i>et al</i> , 2015
<b>Corn</b>	Nanoclay fillers	Reduced water absorption, moisture uptake, and oxygen permeation.	Mohan <i>et al</i> , 2017
<b>Corn</b>	CNF chitosan	+ Increased the rigidity and opacity of the films and reinforced oxygen and water vapor barrier properties.	Yu <i>et al</i> , 2017
<b>Tapioca</b>	ZnO NPs	Increased the efficacy of the film to inhibit pathogenic bacteria	Warsiki and Bawardi, 2018
<b>Corn</b>	PLA	Improved mechanical and water barrier properties	Abdullah <i>et al</i> , 2019
<b>Wheat</b>	Glycerol	High protective barrier and tensile strength	Saiful <i>et al</i> , 2019
<b>Potato</b>	Citric Acid + Chitosan	Improved the water resistance properties and enhance the mechanical and antimicrobial properties	Wu <i>et al</i> , 2019
<b>Dialdehyde</b>	Silica solutions	Better tensile strength and long biodegradability	Oluwasina <i>et al</i> , 2020
<b>Cassava</b>	Polyvinyl alcohol (PVA)	Higher tensile strength, slightly increased bioplastic elongation and more thermally stable.	Syamani <i>et al</i> , 2020

### 3.4 Scientific improvements

From the previous section, it was possible to identify that regardless of the source used for the films, there are several limitations from the mechanical point of view and from the permeation of water and oxygen that can be improved by nanoparticles, nanoclay or nanotubes, natural fibers, crosslinking, or plasticizers. This section presents the scientific advances related to the use of the aforementioned compounds and how they make an improvement.

### 3.4.1 Nanoparticles

NP have been discussed in material science since the early 1990's; consisting in materials with nanoscale reinforcement containing particle sizes less than 50 nanometers. They are used in the plastic industries to enhance mechanical properties, increase thermal stability, improve tensile strength, and decrease gas permeability, which is valued in the packaging industry. The only downside of their employment is the higher cost of NP-polymers when compared to conventional polymers.

ZnO had been wide applied due to its unique properties such as photo-catalytic properties, anti-UV properties, antibacterial properties, semiconductor properties as well as low risk for human health. In addition, zinc oxide can be consumed and has no potential to pollute the environment. Besides, ZnO has a low price, an abundant supply in nature, a stable chemical structure and non-toxicity (Warsiki & Bawardi, 2018). The ZnO NPs are found to have large surface to volume ratio, chemically alterable physical property, increased surface reactivity, unique thermal, mechanical and electrical properties (Kanmani & Rhim, 2014). Furthermore, although Ag has been traditionally applied as an antibacterial substance in food items and beverages, ZnO NPs provide a more reasonable and safe food packaging solution for mass commodities. Zare *et al.*, (2019) observed that the incorporation of ZnO NPs into PHBV induces antimicrobial activity, which potentially increases the shelf life of food. Also, it improves the tensile strength and Young's modulus of PHBV and increases its maximum decomposition temperature.

Another incorporation is gold NPs as antimicrobial agent. Active biofilms of quinoa starch containing gold NPs were successfully obtained by (Pagno *et al.*, 2015). It was proven that the

addition of gold NPs resulted in an enhancement in the tensile strength, an increase in UV radiation absorption and a decrease in the solubility, this last providing an improvement for the protection to packaged food, as well as enlarging the possibilities of applications.

In addition, chitosan nanoparticles have also been incorporated in the study of edible films. Santos *et al.*, (2021) presented the incorporation of chitosan nanoparticles into the polymer (CMC), in an effort to reinforce and enhance the natural properties. Results showed the permeability values were higher when chitosan NP's was added, possibly due to the interaction with the hydrophilic groups of the CMC chain, reducing the content of free groups and thus facilitating the passage of water through the polymer matrix. The formulations of CMC with chitosan NP's presented satisfactory visual characteristics for application in packaging, such as good homogeneity, and handling, as well as contributing to the improvement of physical, mechanical and barrier properties.

### **3.4.2 Nanoclay and nanotubes**

Studies have proven that the addition of nanoclay in biopolymers resulted in improved thermal, mechanical and physical properties (Navarchian *et al*, 2015). Moreover, it also displayed an improved CO<sub>2</sub>, O<sub>2</sub>, and moisture barrier properties. Thus, (Mohan *et al.*, 2017) determined the role of nanoclay in reducing the water uptake and its outcome on biodegradation properties. They observed that the fillers reduced about 30–40% water uptake when compared to an unfilled biofilm. Also, it was proven that, depending on its concentration, nanoclay addition delays the film biodegradation decay. This phenomenon suggests that the nanoclay-filled biofilm biodegradation activity can be controlled and suitable for food packaging.

Among the clay NPs, halloysite nanotubes (HNT) are an emerging nanofiller with hollow tubular morphology that improves physical barrier properties of polymeric materials such as poor barrier properties to water vapor (Biddeci *et al.*, 2016). Halloysite is a clay mineral and a biocompatible material as shown by several studies Bellani *et al.*, (2016); Lvov, & Fakhrullin, (2016); Piana *et al.*, (2015) . It is suitable as nanofiller for sustainable packaging. In addition, HNT loaded with essential oils were successfully used to obtain composite biofilms appropriate for packaging applications.

### **3.4.3 Natural Fibers**

Natural fibers have been promising candidates when replacing synthetic reinforcing fibers in biopolymers, finding bamboo on top of the list. Bamboo is a good fiber as reinforcement because of the high modulus of its microfibrils. On the study made by Abdul Khalil *et al.*, (2018), an increase in the mechanical strength of films by the addition of bamboo fibers was exhibited. The latter was associated with the strong interaction between polymer matrix and bamboo fiber through the formation of hydrogen bonds. The extent of the increase in mechanical strength depends directly upon the average amount of the dispersed fibers, which is reflected in an increase in mechanical properties when the fiber load increases. The results also show that there is a limit in addition of the bamboo fiber. Both mechanical and physical properties increase when fiber is used in an excessive amount. It can be explained, considering that, the filler might weaken the film due to a poor adhesion at the interface.

### **3.4.4 Crosslinking**

Crosslinking is a promising technique to improve the performance and applicability of polysaccharide-based films, especially when concerning their water sensitivity. In crosslinking,

covalent bonds or short sequences of chemical bonds are generated to link two polymer chains together. Several crosslinking agents have been used in previous studies specially involving citric acid. Olsson *et al* (2013) found that thermoplastic starch films cross-linked with different amounts of citric acid show a lowering in moisture content, in diffusion coefficient and in water vapor permeability values. In another study, Priyadarshi *et al* (2018) incorporated citric acid into chitosan films. This modified biopolymer, displayed a better water resistance (moisture content, water absorption and water vapor permeability) than chitosan due to crosslinking. However, the actual application of those agents in the preparation of films or coatings for various packaging material is limited due to their cytotoxicity, high cost and efficiency (Wu *et al.*, 2019). Moreover, there is limited information on the crosslinking technique, because some studies displayed better water resistance while others state that it led to poorer mechanical properties. Consequently, there is more research that needs to be done under this subject of study.

### 3.4.5 Plasticizers

Plasticizers are generally added to the film formulations to overcome their weak mechanical properties, especially its brittleness. The most common plasticizer is glycerol because it can strengthen the bioplastic by improving the chemical bonding leading to higher tensility (tensile modulus). Also, the addition of glycerol makes the film flexible while increases the elongation at break with moderate tensile strength (Krishnamurthy & Amritkumar, 2019). However, glycerol is highly hydrophilic and hygroscopic, leading to significant augmentation in the water vapor permeability of films. Accordingly, Liu *et al.*, (2018) considered 1,8-octanediol (OD) as a plasticizer alternative, due to its largest water-soluble aliphatic diol, with no reported toxicity. However, its polarity is obviously weaker than that glycerol one. The results of Liu *et al*

suggested that, although the plasticizing efficiency of OD was not as good as that of glycerol in the films, OD-plasticized films reported higher water resistance.

Polyvinyl alcohol (PVA) is another plasticizer that have been used. PVA is a biodegradable synthetic material that has hydroxyl groups (-OH) in its structure. PVA has the advantages of good film forming, strong conglutination, and high thermal stability, hence it has been widely used in the materials industry, as well as in packaging industry due to it is tasteless, odorless, nontoxic, dissolvable in water, and resistant to oil and fat. The results from Syamani *et al.*, (2020) displayed that the addition of PVA into cassava starch produced bioplastic with higher tensile strength, almost 10 times more, compared to that without PVA addition.

### **3.5 Future challenges**

The green alternative of biopolymers has been of great interest in the past years due to the great potential to replace the conventional plastic. Numerous sources, and different ways of improving its mechanical properties, make biopolymers a great subject of research. There are numerous possibilities to create different types of biopolymers depending on its application, in this case, food packaging. However, it is of great interest to approach the series of challenges to successfully achieve its industrial application.

#### **3.5.1 Life Cycle Assessments**

The need of Life Cycle Assessments (LCA) studies is crucial. An LCA involves collecting information on the inputs and outputs, such as emissions, waste, and resources, of a process (life-cycle inventory) and translating those to environmental impacts, using standardized impact assessment methodologies (ISO 14040:2006). Only a systematic review by Kakadellis and Harris (2020) was found about application of LCA. Nevertheless, this study did not provide enough

evidence to state which polymer is the best at reducing food waste. Therefore, a more detailed studies are required in this regard.

### **3.5.2 Unintended Green House Gases (GHG) emissions**

It is important to mention that land use change associated with agricultural feedstocks for bioplastic production can cause significant GHG emissions, especially if rainforests, grasslands or peatlands are being displaced. Although in some countries, such as Europe, the landfill gas needs to be captured and used as energy, most landfills in the rest of the world have no gas collection systems and therefore, landfilled bioplastic wastes increase greenhouse gas emissions. In this case, LCA is also taken into account because with the LCA studies can be determined which type of biopolymer might reduce carbon emissions the most (Zhao *et al.*, 2020).

### **3.5.3 Biodegradation of bioplastics**

Biological degradation of bioplastics without the right conditions, such as micro-organism, temperature, and humidity can be very slow and can compromise the sustainability of the bioplastic (Zhao *et al.*, 2020). More scientific evidence is needed to understand the extent to which different biopolymers biodegrade in a range of managed and unmanaged environments.

### **3.5.4 Real food applications**

Fresh food is highly susceptible to spoilage during storage due to microbial growth and chemical deterioration, specially through oxidation (Alizadeh-Sani *et al.*, 2020). Various studies, *e.g.* Kanmani & Rhim, (2014) presented experimental results in which antimicrobial packaging films improved shelf life of food and therefore it could be used as promising alternative to synthetic or petroleum-based packaging films. Moreover, Benbettaïeb *et al.*, (2014) presented an

improvement on chitosan films that could be used for the incorporation of antimicrobial agents to create a controlled release systems for food preservation. Nevertheless, further studies are needed to analyze their potential performance in improvement of real food applications.

#### **4. Conclusions**

- A bibliometric analysis of research work from 2001 up to June 2021 was develop about biopolymers in food packaging applications. According to the information collected from WOS, Scopus and Taylor & Francis, the subject of study shows an increasing scientific interest due to biopolymers have a potential for the substitution of conventional plastics.
- The production of biopolymers is still a growing subject of study, being chitosan, PLA, cellulose, and starch the most common sources. In result, these biopolymers presented poor mechanical properties such as water permeability and low tensile strength. However, it has been demonstrated that the addition of nanoparticles, plasticizers, natural fibers or the use of crosslinking can improve the biopolymer mechanical properties.
- The development of bioplastics is not enough to mitigate the environmental issue. The need to study the emissions of greenhouse gases and the degradation of biopolymers is crucial to lead the investigation to an industrial scale. Furthermore, biopolymer production faces several challenges in order to completely replace conventional plastics. The challenges are in fields such as analysis of Life Cycle Assessment (LCA), unintended greenhouse gases (GHG) emissions, biodegradation and real food applications.

## 5. Recommendations

- Based on the results collected in this research and the bibliographic contribution of this monographic text, it is recommended to continue the research on bioplastics, since it is such a growing subject, every week there are new advances and contributions.
- It is put to the consideration of the reader and the educational community to investigate mainly on the nanocomposites used in biopolymers to see how easy or how hard will be to lead this investigation to the industrial level.
- To extend the PRISMA methodology used in this thesis to other types of studies, since it follows an advance methodology for scientific reviews.
- Carry out a study of the current status of biopolymers in Colombia and how can it be impulse and enhanced.

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