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# Evaluation of Polymeric Films Based on Starch and Antimicrobial Compounds as an Alternative to Replace Food Wrap

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**ABSTRACT**

One of the macromolecules most used as a replacement alternative for some conventional packaging materials is starch. This is due not only to its high bioavailability and economy, but also to its positive environmental impact. However, this material presents important physical-mechanical limitations, such as: high permeability to water vapor, rigidity, fragility, hygroscopicity and tendency to retrograde. Additionally, the incorporation of antimicrobial compounds in the polymer matrix enhances the use of these films, and the addition of plasticizers and reinforcing materials improves their mechanical properties so that they can increasingly compete with synthetic polymers.

This review article presents information on films based on starch obtained from tubers, and the effect of incorporating natural antimicrobial compounds present in some essential oils.

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## Introduction

One of the most commonly used synthetic polymers as food wrapping material is polyethylene. The final disposal and removal of this plastic is expensive, and represents serious environmental problems, which is why alternatives are sought that minimize the environmental impact generated by conventional packaging waste [1].

One of the alternatives that has aroused great interest in the industry is the use of biocompatible polysaccharides such as starch, since they are renewable, abundant, economical and also have the ability to form continuous matrices due to their hydrophilic properties. Even so, its technological application is limited by its physical, mechanical and barrier properties, since starch-based films have high hygroscopicity, rigidity, water vapor permeability and retrogradation compared to synthetic materials and [2-7].

A strategy to counteract the aforementioned limitations is the incorporation of reinforcing materials such as cellulose, which can be obtained from different agroindustrial waste, making it a material of interest not only for its high bioavailability, low cost and biodegradability, but also because, depending on its concentration, size (aspect ratio) and dispersion among other variables can increase the mechanical resistance and decrease the water vapor permeability of the films, which favors their application in food systems. Likewise, the incorporation of glycerol as a plasticizing material in the matrix can desirably alter physical-mechanical properties such as flexibility or elongation and some optical properties of the biofilms [8-10].

Likewise, the incorporation of glycerol as a plasticizing material in the matrix can desirably alter physical-mechanical properties such as flexibility or elongation and some optical properties of the biofilms. A source of antimicrobial compounds are some essential oils, these active agents not only provide antibacterial protection, they also improve the mechanical, functional and barrier properties, in addition, they can replace some synthetic additives and prolong the useful life of the product since it has been identified that during During storage and transportation, there may be contamination due to the characteristics of these package. Several studies showed the successful incorporation of antimicrobials in starch films [11-15].

This review work aims to understand the processes of obtaining starch-based films with the incorporation of antimicrobial compounds of natural origin, properties of the films, techniques to evaluate the migration phenomenon of antimicrobial compounds and finally, possible uses of these films. as food packaging materials.

## Methods for Incorporation of Antimicrobial Compounds in Biobased Films

The correct incorporation of antimicrobial compounds in biobased films is one of the great challenges for researchers, since it has not been possible to incorporate essential oils in a homogeneous manner distributed in the film. For this reason, various methods of incorporating bioactive compounds that can achieve this uniformity have been studied.

Ensuring the stability of an emulsion with the use of an additive called polysorbate 80, better known as Tween 80, has been one of the most used methods to promote compatibility between the components of the film when oils or oleoresins are incorporated, allowing there to be a complete and uniform distribution of the antimicrobial agent in the film. Polysorbate 80 is a nonionic surfactant that is in a liquid state, yellowish and viscous appearance that is derived from sorbitan. polyethoxylated and oleic acid. Polysorbate 80 is used as an emulsifier in foods, as it increases the resistance to fusion within the matrix.

suggest using this additive proportionally to the concentration of oil to be incorporated into the film. After forming the emulsified film, it is subjected to the drying process for two days at temperature and relative humidity conditions of 50°C and 50% respectively, to evaluate the concentration of oil retained in the film after drying and thus determine the antimicrobial potential against Staphylococcus aureus and Escherichia coli from lavender oil (Lavendula angustifolia) [16].

On the other hand,  proposed Tween 80 to form an emulsified mixture with the film, however, they propose that the concentrations of this additive were lower than the concentrations of oregano essential oil (Origanum vulgare), since this would allow effective antimicrobial activity to be achieved in the biobased films against strains of S. aureus and E. coli [17].

Similarly, stated that it is important that the concentration of antimicrobial compounds such as the essential oil of thyme (Thymus), flowers belonging to the Laminaceae family (Zataria mutiflora) and eucalyptus (Eucalyptus globulus) exceed the concentration of the emulsifying agent (Tween 80), since the loss of antimicrobial potential during the film drying stage must be considered [18-20].

Other studies evaluated the retention of clove essential oil (Syzgium aromaticum), and olive oil (Olea europaea) in the films based on Job's tears starch, potato and starch-chitosan mixture in which the incorporation of the emulsifying agent exceeded that of the bioactive compound so that the latter was uniformly retained in the film, after drying [14,21].

Another method of incorporating bioactive compounds consists of the association of liposomal compounds, which promotes the retention of compounds of interest in the films obtained after drying. Liposomals are spherical structures that form spontaneously when the forming lipids are dispersed in an aqueous medium. These structures depend on the composition of the cell membrane and are characterized by having a polar part and a hydrophobic part, which give them amphiphilic properties. These vesicles act as an encapsulating agent. However, the use of this method has been more evaluated in the pharmaceutical and cosmetological industry due to the high compatibility with the cell membrane acting here as transport vehicles [18].

On the other hand, different studies have developed films with the incorporation of antimicrobial agents present in the essential oils of oregano (Origanum vulgare) and rosemary (Salvia rosmarinus) without the addition of emulsifying agents, which can alter the physical or barrier properties of the resulting films and using the high shear rate dispersion technique. However, the authors reported that at the conditions evaluated (13000 rpm; 30 - 50 °CY 50% RH) this method decreased the homogeneity in the incorporation of the compounds of interest, affecting their retention in the film, and consequently, the antimicrobial activity [22-25].

## Properties of Starch-Based Films with Incorporation of Natural Antimicrobial Compounds

### Mechanical Properties

Regarding the impact of the incorporation of essential oils containing antimicrobial compounds on the properties of films based on biopolymers, various results have been reported. As reported by Potato starch films containing lavender essential oils manage to reduce the tensile strength of the film, going from 70 MPa to 50.8 MPa since the incorporation of a lipid agent can hinder the interaction between the chains. of polymers and create flexible areas in the film [16].

Similarly, showed decreases in tensile strength and tensile strength, as well as an increase in flexibility after the incorporation of oregano essential oil, which leads to the formation of a structure heterogeneous with discontinuities compared to the film without essential oil.

This same behavior was reported by, who mentioned that the incorporation of zataria multiflora oil reduced the tensile strength values compared to those obtained for the control film but increased the percentage of elongation to the break. The bioactive compounds present in the essential oil of zataria multiflora such as thymol and carvacrol are mainly responsible for the plasticizing effect, interfering in the interaction between the polymer chains. As a result of this interaction, the plasticizing effect and flexibility can improve on the one hand, but the strength and rigidity of the edible films decrease . It has been reported that the changes in the mechanical properties of the films are attributed to the partial replacement of starch-starch intermolecular interactions in the film matrix by starch-essential oil intermolecular interactions, resulting in a decrease in the rigidity of the polymeric starch network and greater flexibility of the films [17,19,21,22,26].

Similar results were obtained by who mentioned that the incorporation of olive oil in the formulation of potato starch films caused a significant decrease in tensile strength (TS) from 21.95 MPa to 10.57 MPa. This behavior may be due to heterogeneity in the film due to the addition of a lipid, decreasing the cohesion and integrity of the starch matrix [14].

Data obtained by showed that the incorporation of cinnamon essential oil (Cinnamomum verum) at a concentration of less than 1% in cassava starch films did not significantly increase the tensile strength and elongation properties. This result may be due to a possible heterogeneity of the film, that is, most of the antimicrobial agent was not distributed uniformly. However, when the concentration of this active compound increases, the values of mechanical properties such as resistance to elongation also increase; This being an effect of the interaction of monounsaturated lipids with the matrix of the biopolymeric film forming a flexible complex. The cassava starch films with 3% cinnamon essential oil had a tensile strength and elongation of 19.74 MPa and 16.03% respectively [27].

### Barrier Properties

mentions that incorporating 2%, 4% and 6% of lavender oil to composite films based on gelatin starch decreases solubility by 9.66%, 11.9% and 18.25 % respectively, since essential oils, as complex mixtures of various chemical compounds, show that the hydrophobic character is a variable characteristic, which affects their behavior and barrier properties of the film. In the same way, the solubility of the starch films of Job's tears decreased from 71.63% to 48.47% with the addition of 0.75% of essential oil, since the affinity of the starch films towards water decreased, thus increasing water vapor permeability. Similar results have been reported with an increase in WVP of the films in cassava starch film with oregano oil (Origanum vulgare) and a film of tapioca starch containing carvacrol [16,28].

Solubility is a factor that guides the application of the film as packaging for food products. In contrast to the results mentioned above, reports that films with oregano essential oil (Origanum vulgare), regardless of the concentration, showed greater solubility than the films without oil. This behavior may be a consequence of the breakage of the films, facilitating the insertion of water into the polymer matrix, and also of the increase in thickness and irregular surface structures of the films, increasing the contact area of the film and water. For any ideal food packaging material, the barrier properties should be low, the mechanical strength should be high. However, the high solubility can be beneficial for the application of the films on fruits and vegetables, for their subsequent disposal [20,24].

In the research carried out by  a significant decrease in water solubility was observed for the films after the addition of oregano essential oil, nisin and lactic acid. This behavior may result from the interactions of lipid polysaccharides, mainly through hydrogen bonds, which limit the interaction of hydroxyl groups with water molecules due to their lower availability, which would lead to the formation of more resistant films. water. The decrease in moisture content and solubility can be attributed to the hydrophobic nature of the lipids and the interaction between the oleic acid components and the hydroxyl groups of the film matrix [24,27].

On the other hand, the addition of olive oil to the starch film results in a reduction of the water vapor permeability value by 31%, compared to the value obtained for the simple starch film. This may be a result of a uniform distribution of olive oil in the plasticized starch network. In addition, oils, as hydrophobic and nonpolar substances, can increase the hydrophobic phase of the polymers and, therefore, reduce the diffusion of water through of the film  [14].

On the other hand, films with 4% concentration of Eucalyptus leaf extract globulus had the lowest water vapor permeability due to the uniform distribution of oil in the film matrix. The improvement in water vapor permeability could be due to the complex composition of the Eucalyptus leaf extract. globulus, since almost 41 phenolic compounds have been identified in Eucalyptus globulus leaf extract. These polyphenols, due to the hydrophobic groups they contain, are able to maintain cross-linking between polymer chains through hydrophobic interactions and may have led to a higher water barrier property of the films [29].

**Antimicrobial Properties**

The proliferation of microorganisms is the main problem in food deterioration, for this reason, the addition of antimicrobial agents present in lipid compounds such as essential oils can be an alternative to mitigate or reduce the presence of microorganisms. The antioxidant and antimicrobial activities of films incorporating essential oils are affected by many factors, such as the chemical nature of the oils, the base materials of the film, the interactions between the components of the films or the size of the particles. of oil droplets in the film matrix  [24,30].

A study carried out by showed that when lavender essential oil was incorporated into the films, the total antibacterial activity was significantly improved by 88.97%, compared to the control film (without oils). essential). However, some studies have reported that the main components of lavender essential oil do not present antibacterial activity. This may be due to the interaction between the compounds present in the oil and the film [16].

Similarly,  demonstrated that potato starch films without the addition of oregano essential oils did not present any inhibitory effect against S. aureus and E. _ coli , which indicated that native starch does not have antimicrobial properties. On the other hand, the incorporation of 0.5% oregano oil in films based on potato starch did not confer any inhibition against E. coli and a very weak inhibition against S. aureus, compared to concentrations between 1% and 7%. This behavior agrees with what was expressed by who stated that inhibition increases with the highest concentration of essential oil, in this case, Eucalyptus oil. globulus. The films incorporating this oil were more effective against Gram-negative bacteria (E. coli) than against Gram positive bacteria (S. aureus), with 4% (1 g of oil/1 g of starch) being the optimal concentration for eucalyptus oil, that is, the concentration at which the maximum inhibition zone was obtained for both microorganisms [17,20].

Likewise, the results obtained by showed that the films with the addition of olive oil presented inhibitory activity against both Gram positive bacteria (Escherichia coli) and against Gram-negative bacteria (Staphylococcus aureus). The addition of 1%, 2% and 5% (1 g oil/ 1 g starch) of olive oil managed to inhibit 12.5%, 4.12% and 12.5% respectively against Escherichia coli, although the decrease did not show a significant difference in activity. However, there was a significant difference in antimicrobial activity against Staphylococcus aureus, where the addition of 1%, 2% and 5% of olive oil allowed the inhibition of this microorganism by 25.00%, 16.62% and 20.87% respectively.

In the case of thyme essential oil, its antifungal effect varied depending on the concentration in the films, showing that at a concentration of approximately 7.4% its antifungal activity was more effective against B. fuckeliana than against A. alternata [18,31].

**Potential Applications**

Due to the modifications in the mechanical and barrier properties, starch-based films with the incorporation of antimicrobial compounds have great potential for use in the food industry.

One of the great potential uses that can be given to this type of film is packaging and coating, since starch has a great capacity to form edible films with reasonable mechanical resistance, and can be used to coat food products with the in order to avoid moisture loss or protect them from unwanted substance. Furthermore, the incorporation of lipid antimicrobial agents provides protection to the food from external factors such as water vapor that spreads microorganisms in the product, deteriorating it or shortening its useful life [14,18,24,32].

Likewise,studied the potential use and performance of films based on oxidized starch as packaging material and food systems and determined that, although these can behave differently, satisfactorily as a packaging material, these require modifications in the mechanical and barrier properties, since films based on oxidized starch present high degradation compared to films based on native starch, however, the low oxidation of these is They exhibit greater stability to heat and mechanical agitation compared to other biopolymers in their natural state [33,34].

Therefore, biofilms with the incorporation of natural antimicrobial compounds are perfect candidates as coating and packaging materials to prevent not only the oxidation of lipids in some foods, but also the protection of these against factors that alter the organoleptic characteristics in storage [11].

In addition to this potential use, point out that the application of films based on corn starch and Job's tears can be used in the meat industry, since studies have shown that The incorporation of essential oils such as clove oil and zataria multiflora flower oil not only improves the mechanical and barrier properties of the films, but also enhances the antimicrobial activity since they act as a barrier to chemical and microbiological agents that deteriorate the meat quality [19,21].

Essential oils manage to maintain the stability of the final product by controlling the growth of bacteria, so this incorporation increases the commercial potential of such foods, thus contributing to the food industry with safer and more sustainable foods for the consumer [34].

**Migration Phenomenon**

The ability to release antimicrobial components through direct contact is an important characteristic because, typically, the greatest microbial contamination occurs on the surface. Therefore, antimicrobial films would act directly at the most critical point. These interactions result in a gradual release of antimicrobial compounds and ensure their action for a longer period compared to direct application. Furthermore, the incorporation of essential oils in packaging is interesting because it is a method of using this natural extract in foods without the need to add them as an ingredient, thus reducing undesirable sensory interferences [20]. According to a study carried out by the release of active compounds from the starch film to meat was 42 to 51% for cinnamaldehyde and 38 to 48% for eugenol, at storage temperatures of 4 at 15 ° C. Likewise, it states that the diffusivity of these active compounds had a significant positive effect on the microbial, visual and chemical shelf life of lamb meat. By comparing the microbial, physical and chemical results, it was found that the shelf life of lamb meat increased by 1 week at a storage temperature of 10 °C and 3 weeks at a storage temperature of 4 °C. Therefore, these spice-fused edible films can be used as packaging to improve the shelf life of meat, which in turn can reduce economic losses for the meat industry [35].

Similarly, states that the essential oils of sage and oregano are compatible with the compounds of bio-based films to form edible films. The results of this study confirm that the interactions between essential oils and films have a critical effect on the diffusivity of the active compounds and, therefore, on the final antimicrobial activity. As a result, to obtain active edible films, it is necessary to find the balance point between the nature and concentration of the active compounds in the essential oil and the film formulation. In food applications, the use of edible antimicrobial films could potentially allow the control of the migration of antimicrobial agents from the film to the food surface, thereby having a continuous effect on it [36,37].

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