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Design of biodegradable composite films for food packaging

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1. Introduction

The materials most used for food packaging are the petrochemical-based polymers, due to their availability in large quantities at low cost and favourable functionality characteristics, such as, good tensile and tear strength, good barrier properties to O_2 and heat sealability [1]. However, these materials are totally non-biodegradable, leading to serious ecological problems. As a consequence, the consumer demand has shifted to eco-friendly biodegradable materials, especially from renewable agriculture by-products, food processing industry wastes and low cost natural resources.

The biopolymers commonly used to produce films are carbohydrates, often vegetal starchy and pectic materials and proteins, vegetal and animal [2,3]. Usually, these biopolymers require that their mechanical and rheological properties be improved by molecular restructuring or by the inclusion of food grade additives. In addition to the appropriate mechanical properties, the films must have also the adequate permeability to water vapour and gases. The specific barrier requirements of the packaging depend upon the products characteristics and the intended end-use application. In the case of a packaged product whose deterioration is related to its moisture content, the barrier properties of the package relating to water vapour will be of major importance in extending shelf life. Similarly, the oxygen concentration in a permeable package will affect the rate of oxidation of nutrients such as vitamins, proteins and fatty acids.

The required specific permeability properties of the films can be obtained by inclusion of inert impermeable barriers and/or reactive compounds in the polymer matrix. The inert barriers can reduce permeability by increasing the diffusion path, while the reactive compounds interact selectively with the diffusing species increasing the time before a significant permeability occurs.

The objective of this work is to develop model composite films based on commercial pectin and κ-carrageenan, and characterize them in terms of their hygroscopic and mechanical properties, and permeability to gases (oxygen and carbon dioxide) and water vapour. In order to design films with specific permeability properties, reactive compounds (ascorbic acid and/or calcium hydroxide to

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interact with oxygen and carbon dioxide, respectively) are added to the polymer matrix, while mica flakes are used as impermeable barriers.

2. Experimental

Commercial κ-carrageenan (SKW Biosystems, France) and pectin from citrus fruit (Sigma–Aldrich, Spain) were used to prepare model carrageenan–pectin aqueous mixtures, with different proportions of the two components. The films were prepared by wet casting of the aqueous solutions and further drying. Their mechanical properties were studied using a TAXT2 (Stable Micro Systems, England) in accordance with ASTM D-882-91 (1996), and the glass transition temperature (T_g) was measured using a Shimadzu DSC-50 differential scanning calorimeter. Water sorption isotherms were determined by placing the samples in desiccators with different relative humidities, imposed by the use of saturated saline solutions, and weighed periodically until they reached constant weight. The water vapour permeance was measured gravimetrically, based on the ASTM E-96-80. In order to determine the gas permeability, the films were placed between two sealed chambers containing nitrogen at the same temperature and pressure. A known quantity of the test gas was injected in one of the chambers ("feed" chamber) and the solute flux across the membrane came indirectly from measuring the total pressure increase with time in the "permeate" chamber.

3. Results and discussion

In the water sorption experiments, the amount of water at equilibrium in the film was higher for films with pure carrageenan, which indicates that carrageenan is more hydrophilic than pectin. It was also observed that the amount of carrageenan in the films leads to a decrease in the glass transition temperature (T_g). This result can be explained by the plasticizing effect of carrageenan itself, since the 100% carrageenan film revealed a lower T_g than the pure pectin film. Alternatively, the increased hygroscopic properties of the pure carrageenan film and the inherent water plasticizing effect could be at the origin of the observed depressed T_g.

The addition of carrageenan resulted also on an improvement of the films mechanical properties, by decreasing the Young modulus and increasing the strain at break. These results are in agreement with the observed plasticizing effect of carrageenan.

It was also observed that, the films become more permeable to water vapour as the carrageenan content increases, until a plateau is reached. This increase in the water vapour permeance can be explained by the higher hydrophilic character of carrageenan when compared to pectin, already observed in the water sorption experiments. However, beyond a critical content of about 67%, a further increase in films carrageenan content has not a significant influence on the water vapour permeance. The inclusion of mica flakes in the polymer matrix (10% w/w in a dry basis) led to a water vapour permeance decrease of about 27%. The flakes act as impermeable barriers that increase the diffusional path of the water molecules.

Ongoing work is focused on the characterization of the films in terms of permeability to gases (oxygen and carbon dioxide). The results suggest that these films are more permeable to water than to oxygen and carbon dioxide, and it is anticipated a decrease of the gas permeability with the inclusion of mica flakes. Moreover, the inclusion of ascorbic acid, which reacts with oxygen, leads to an improvement of the films selectivity (CO_2/O_2).

4. Conclusions

The model carrageenan–pectin films studied showed increased mechanical properties, lower glass transition temperatures, increased water permeability and hydrophilic properties with increased carrageenan content. The water permeability was significantly reduced by including mica flakes in the polymer matrix, which act as impermeable barriers. These films revealed to be more permeable to water vapour than to oxygen and carbon dioxide, and the films selectivity (CO_2/O_2) was enhanced by adding ascorbic acid to the polymer matrix. The permeability properties obtained can be advantageous for the preservation of fresh fruit and vegetables.

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