



Hydration and water vapour transport properties in yeast biomass based films: A study of plasticizer content and thickness effects



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ABSTRACT

Research on biodegradable films is a topic of strategical interest in the field of food science and technology. These films are membranes conformed by biopolymers that interact strongly with water and they generally require the addition of plasticizer in order to improve its integrity and mechanical properties. The study of the hydration and water transfer properties through these films, as well as understanding in what way the thickness and the addition of the plasticizer affect these properties, is essential for a better knowledge and optimization of the required protective function of these membranes. In this work the hydration kinetics and water transport through biodegradable films obtained from yeast biomass were studied, in order to know the performance against changes in thickness and glycerol content. Hydration kinetics experiments allowed obtaining solubility of water in the matrix film, and the diffusion coefficient. Results demonstrated that when glycerol content was increased, the solubility of water increased while diffusion remained constant. Moreover, when the film thickness was grown, the solubility of water decreased but the diffusion increased. Experimental water vapour permeability obtained through traditional cup method was compared to the theoretical permeability calculated by multiplying diffusion coefficient and water solubility. The good agreement observed between both values allowed the analysis of how solubility and diffusion contributed to permeability. As a result this study revealed that the increment of permeability with the plasticizer content was due to the increase in solubility, while the effect of the thickness increasing permeability was dominated by diffusion.

1. Introduction

Research and development of new methods to extend shelf-life and make a better use of available resources are very active areas in the field of food science and technology. In this regard, the study of biodegradable coatings and films based on biopolymers has taken great interest in recent years [1–5]. These films are a thin matrices preformed from a solution or dispersion of biopolymers. To form the film matrix it is necessary to remove the solvent by an appropriate method in order to decrease the distance between polymers and favouring their interaction [5]. This interaction promotes an interleaving of polymers chains that increase the viscosity of the system, allowing the formation of a polymer network that will be ended with a film conformation [5].

For food applications, these films should present certain flexibility and they must offer a good protection acting as a permselective membrane [6,7]. Plasticizers increase film flexibility due to their ability to reduce internal forces between polymer chains while increasing molecular space [8]. The best plasticizers will generally resemble most closely the structure of the polymer that they plasticize, thus polyols

such as glycerol are commonly used in biopolymers based films [9].

The analysis of interactions between these films and water molecules has a great importance in the area of packaging, in order to know if films can be applied as an efficient barrier to avoid hydration or dehydration of the food. Depending on the type of food where the coating or film will be applied, in some cases they may require low water vapour permeability [7]. For other products, it may act as sacrificing agent losing in first place its own humidity and preserves the humidity of the food product, extending the shelf-life [10].

Polysaccharides and proteins interact strongly with water; therefore films made from these biopolymers are hydrophilic films. Films based on yeast biomass represent a good model for the study of hydrophilic films and their interaction with water because they are formed basically by polysaccharides and proteins [11].

In general, hydrophilic films made from biopolymers show water sorption isotherms with a slight increase in hydration water content for low values of water activity (a_w), and a significant increase for $a_w > 0.6$ [5,11]. This shape of sorption isotherms suggests that the hydration is presented in a multilayer form, with a small amount of water strongly

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bounded to the polymeric matrix forming the monolayer and then, most of the hydration water forming multilayers [5,11], which are indirectly bounded to the polymer matrix [12]. As follows, hydration water in these materials is susceptible to be moved by diffusion mechanism.

The study of the water transport properties through films is essential for a better understanding and optimization of the required protective function of these membranes [13,14]. Hydration water affects the main structural and functional properties of hydrophilic films [15], because water acts as plasticizer by embedding itself between the polymers chains, spacing them, lowering the glass transition temperature (T_g), and affecting flexibility [16].

The water transport through polymeric films does not occur through pores, but can be understood through a simple model that proposes that process occurs in four steps: (i) absorption of water vapour on to the polymer matrix surface; (ii) solution of water vapour into the polymer matrix; (iii) diffusion of water vapour through the matrix; and (iv) desorption of water vapour from the other surface of the film [17,18]. Therefore water vapour permeability in these materials depends on the hydration or solubility of water in the film, as well as the water mobility within the matrix [18,19]. Thus, the chemical structure, polarity, degree of crystallinity, density, crosslinking degree, molecular weight and polymerization, as well as the presence of other plasticizers, are factors affecting permeability, because they influence hydration properties, degree of molecular mobility and diffusion through the material [13,20,21].

The mechanism in which water molecules are transported through hydrophilic polymeric films is a complex phenomenon not well understood. It was reported some anomalies that occur in biopolymeric films strongly interacting with the species that permeates, especially the strong dependence of permeability coefficient with film thickness [22–25] and plasticizer content [26–29]. Therefore, understanding in what way the thickness and the addition of the plasticizer affect the permeation process, is essential for a better knowledge and optimization of the required protective function of these membranes. The aim of this work is contribute to the explanation of these aspects, not completely lightened up to now [30].

In this work, the hydration kinetics and water transport through biodegradable films obtained from yeast biomass were studied against changes in film thickness and glycerol content. Through kinetics experiments, the solubility of water in the matrix film and the diffusion coefficient of water were obtained. Taking into consideration that the theoretical permeability coefficient is expressed as the contribution of solubility of water in the film and the diffusion of the permeant in the matrix [17–20], this theoretical quantity was compared with experimental permeability obtained by the traditional cup method [31]. The agreement between theoretical and experimental permeability allowed studying the effect of thickness and plasticizer on the water vapour permeability, in a more comprehensive manner analysing the contribution of solubility and diffusion.

2. Materials and methods

2.1. Materials

Saccharomyces cerevisiae yeast cells were obtained from commercial pressed cells from Calsa, AB Mauri (Tucumán, Argentina). Glycerol, silica gel, and all salts used were purchased from Biopack (Buenos Aires, Argentina).

2.2. Preparation of films

Films were prepared by using *Saccharomyces cerevisiae* yeast cells that were previously cleaned with distilled water and centrifuging at 750g. The supernatant was discarded and replaced by distilled water to reach 10% w/vol (dry matter of yeast/volume). As described in a previous work [11], the standardized dispersion was submitted to high

pressure homogenization (H) at 125 MPa in a continuous flow during 9 min, followed by a thermal treatment (T) in a water bath at 90 °C for a time lapse of 20 min. A second high pressure homogenization step was applied at the same conditions of the first homogenization process to obtain the sample named HTH [11]. The final pH of the prepared dispersions was 6. Glycerol was added to dispersion at levels of 0, 10, 20, and 30% w/w with respect to dry matter (d.m.). Volumes of 5, 10, and 20 mL of HTH dispersions were placed in 8.6×10^{-2} m diameter Petri dishes, in order to obtain films of thicknesses close to 7, 15, and 30×10^{-5} m respectively. Evaporation of water was done at 40 °C in a ventilated oven, until the remaining water content of the films was between 10 and 15%. Next, films were stored at 24 °C and 43% r.h. generated by a saturated solution of K_2CO_3 .

2.3. Density and thickness measurements

For density determination of dried films, samples with $5.8 \times 10^{-3} \text{ m}^2$ of circular area were dried at 0% r.h. into desiccators containing silica gel until a constant weight was achieved; this process lasted around 7 days. Films were weighed using analytical balance ($\pm 10^{-4}$ g). Thickness was measured by a digital calliper ($\pm 10^{-6}$ m) at ten different places of the film, obtaining for each specimen an average value with an error lower than 5%. The values measured were close to 7, 15, and 30×10^{-5} m, as expected according to the volumes of 5, 10, and 20 mL of dispersion placed in the Petri dishes. Film density was calculated through Eq. (1)

$$\rho_{d.f.} = \frac{m}{AL} \quad (1)$$

where $\rho_{d.f.}$ is the density of dried film (g m^{-3}), m the dry mass (g), A the area (m^2), and L the thickness (m).

2.4. Microstructural characterization by scanning electron microscopy (SEM)

The equipment used was a scanning electron microscope SEM LEO EVO 40-XVP (Carl Zeiss AG, Germany). For best viewing under a microscope, the samples were coated with a gold layer. Magnifications used were $500\times$ with a potential difference of 5kV. The absolute pressure during the study was 80 Pa (High vacuum conditions).

2.5. Kinetics of water sorption experiments

Dried films with different content of glycerol and thickness, were placed in Petri dishes and located into a sorption container at 90% r.h. generated by a saturated solution of $BaCl_2$. A fan was used to maintain uniform conditions inside the desiccators following the recommendations of previous authors [22]. Samples were removed at specific intervals of time and weighted for data collection. Weighing of samples was made using analytical balance ($\pm 10^{-4}$ g). The water content h , given in units of g of water per g of dried mass was evaluated as function of time t , taking the difference between the mass of the hydrated film and that of the dried film. Three replicates of each film were tested. Experiments were carried out at 24 °C.

Water content as function of time, $h(t)$, was fitted with Fick's mass transport differential law solution in Fourier series, for one-dimensional diffusion of water in a plane infinite sheet as was described by Crank [32], and done by Eq. (2)

$$h(t) = h_{\infty} \left\{ 1 - \sum_{n=0}^{\infty} \frac{8}{(2n+1)^2 \pi^2} \exp \left[-D_w^{\text{eff}} (2n+1)^2 \frac{\pi^2 t}{4L^2} \right] \right\} \quad (2)$$

where h_{∞} is the water content at equilibrium, L is the film thickness, and D_w^{eff} is the apparent diffusion coefficient in units of $\text{m}^2 \text{ s}^{-1}$. Hydration experiments were performed with one surface of the film exposed to the wet atmosphere, where water molecules penetrated in

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