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Physical and Mechanical Properties of Hydroxypropyl Methylcellulose–Coated Paper as Affected by Coating Weight and Coating Composition

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Hydroxypropyl methylcellulose (HPMC)–coated papers without plasticizer and plasticized with polyols were prepared, and the effects of coating weight, different plasticizers (glycerol (GLY), sorbitol (SOR), and polyethylene glycol (PEG)), and plasticizer contents (20% to 50%) on the physical and mechanical properties of the resulting biopolymer-coated papers were studied. Coating weight was the most important factor affecting mechanical properties. Conversely, increasing coating weight led to a decrease in gloss and to an increase in tensile strength (TS), elongation at break (%E), and tearing resistance of coated papers. The application of unplasticized HPMC coatings (3 g/m^2) on paper reduced water vapor permeability (WVP) and water absorption capacity by 25% as compared with uncoated paper. All plasticizers significantly ($p < 0.05$) increased WVP and $Cobb_{60}$ values of the films. With the exception of PEG, no effect was found with plasticizers on TS and %E of coated papers compared with those without plasticizer. HPMC-coated papers with PEG as a plasticizer showed significantly lower TS and higher %E and tearing resistance than the other plasticized films ($p < 0.05$). HPMC coating improved tensile properties and tearing resistance of paper and could be regarded as a reinforcement layer.

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INTRODUCTION

Packages protect food from the loss of nutrients, functional properties, color, aroma, and taste. They also preserve the general appearance expected by consumers and act as a barrier to microbiological contamination. Losses are due to a series of physical interactions, as well as chemical, biological, and biochemical reactions, and are accelerated when oxygen and water are present in the packages. Synthetic food packaging is classically used to prevent these losses and to maintain and improve food quality and to increase food product shelf life. However, most synthetic packaging films are composite polymeric structures that are difficult and expensive to recycle.

Over the last few years, increased environmental concerns over the use of certain synthetic packaging and coatings in combination with consumer demands for both higher quality and longer shelf life have led to increased interest in alternative packaging materials research. Several different renewable materials have been studied for packaging applications, but only a few are on the market today (Gällstedt and Hedenqvist 2006). The most common renewable packaging materials are cellulose-based, including paper

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and paperboard.

Although biodegradable and environmentally friendly, paper contains cellulose, whose hydrophilic nature causes poor water-vapor–barrier characteristics. Paper packaging also easily absorbs water from the environment or from the food and loses its physical and mechanical strengths. Paper is often coated with a polyolefin material to improve its barrier properties to water vapor, oxygen, and aromas. Unfortunately, the obtained material loses its biodegradation and recyclability characteristics due to the addition of synthetic polymer layers.

In an effort to produce more environmentally friendly and renewable materials, biopolymers have been investigated as surface-coating materials on paper or paperboard (Han and Krochta 2001; Lin and Krochta 2003). Biopolymer coatings on paper packaging materials can serve as moisture, grease, and oxygen barriers in many food packaging applications. Moreover, they have the potential to replace current synthetic paper and paperboard coatings (Khwaldia *et al.* 2010).

In the literature, renewable biopolymers prepared from polysaccharides, proteins, and lipids, or combinations of those components have already been incorporated into the production of paper products to provide interesting functionalities while maintaining the environmentally friendly characteristics of the material (Trezza *et al.* 1998; Gällstedt *et al.* 2005; Ham-Pichavant *et al.* 2005; Despond *et al.* 2005; Khwaldia *et al.* 2005; Kjellgren *et al.* 2006; Khwaldia 2010).

Han and Krochta (1999) showed that whey-protein–coating of paper improves the packaging-material performance of paper by increasing its oil resistance and reducing its water-vapor permeability. Despond *et al.* (2005), as well as Kjellgren *et al.* (2006), used paper coated with chitosan or chitosan/carnauba wax to obtain a packaging material with good barrier properties toward oxygen, nitrogen, carbon dioxide, and air. Bilayer coatings composed of beeswax and whey protein isolate/cellulose xanthate or poly(vinyl butyral)/ zein decreased the water-vapor–transmission rate by 92% to 95%, hence approaching commercial attributes required to ensure water-vapor barrier in paperboard-based food containers (Han *et al.* 2010). When paper was coated with sodium caseinate (NaCAS), its water-vapor barrier and mechanical properties were improved (Khwaldia 2010). According to Rhim *et al.* (2006), water-barrier properties of paperboards can be increased by alginate coating with a $CaCl_2$ post-treatment.

Polysaccharide-based polymers such as hydroxypropyl methylcellulose (HPMC) are used in food industries as an emulsifier, a protective colloid, a suspending agent, and a film former (Tharanathan 2003). HPMC presents excellent film-forming properties that enable the production of a tough and protective coatings (Villalobos *et al.* 2006), with efficient oxygen, carbon dioxide, and lipid barriers. Akhtar *et al.* (2010) showed that HPMC films containing suitable edible colors act as an adequate light barrier to avoid photo-oxidation of salmon oil during extended storage. This polysaccharide is available in a wide range of degree of substitution, molecular weight, viscosity, and particle size, influencing the properties of the subsequent film coatings (Capan 1989). In addition, its nontoxic nature, ease of handling, and relatively simple manufacturing technology (Shah *et al.* 1996) make HPMC a promising coating material for paper to improve some of its functional properties. Sothornvit (2009) reported that HPMC-based coatings improved paper flexibility and durability and reduced WVP, and further reduction was obtained when beeswax was incorporated in the HPMC-lipid composite–coated paper.

Barrier and mechanical properties of coated cellulosic materials strongly depend on the cellulosic substrate, the coating application technique, and the nature of the

coating materials and their final compositions. Thus it is important to choose the proper biopolymer matrix and coating ingredients such as plasticizers. The addition of a plasticizer is required to overcome the brittleness of films, to obtain structures with enough flexibility, and to impact the resistance of coatings. Plasticizers increase the free volume or molecular mobility of polymers by reducing hydrogen bonding between polymer chains. Film permeability always increases with increasing plasticizer content. Proper selection of a plasticizer for a given polymer will allow optimization of the film mechanical properties with a minimum increase in film permeability (Sothornvit and Krochta 2005). The selection of a plasticizer for a polymer matrix is normally based on the compatibility and permanence of the plasticizer, the amount necessary for plasticization, and the desired functional properties of the films (Sothornvit and Krochta 2001). Good compatibility results from the plasticizer and polymer having a similar chemical structure. For polysaccharide-based edible films and coatings, hydrophilic plasticizers containing hydroxyl groups, which form hydrogen bonds with polysac-charides, are the best suited for this use. Plasticizers used for polysaccharide-based films and coatings are glycerol, sorbitol, xylitol, mannitol, polyethylene glycol, ethylene glycol, and propylene glycol (Sothornvit and Krochta 2005).

This study aimed at developing a variety of plasticized HPMC coatings on paper, and investigating the effects of coating weight, different plasticizer types, and plasticizer concentrations on the physical and mechanical properties of the resulting biopolymer-coated papers.

EXPERIMENTAL

Materials

Paper packaging used as a support for coating was supplied by SOTEFI (Soukra, Tunisia) with a grammage of 79.15 ± 0.89 g/m^2 and an average thickness of 98 ± 1.21 μm at 23 °C and 50% RH. HPMC (Methocel E-19, Food grade, molecular weight ~ 50,000 Da) was provided by Dow Chemical (Midland, USA). Food grade glycerol (GLY, >97% purity), sorbitol (SOR), and polyethylene glycol (PEG, molecular weight ~200) were used as plasticizers and were purchased from Sigma Aldrich (Steinheim, Germany).

Coating Solutions

Preparation of HPMC-based coating solutions was adapted from Imran *et al.* (2010). Seven grams of HPMC powder was dissolved in a mixture of distilled water (65 mL) and ethanol (35 mL) heated at 65 °C with constant agitation, until all particles were thoroughly dispersed. Then, appropriate amounts of plasticizers (GLY, SOR, or PEG) were added individually to the film-forming solution dispersions to give a final concentration of each plasticizer at 20%, 30%, 40%, and 50% (w/w) based on HPMC weight).

Coating Method

A control coater (model KCC 101, RK Print-Coat Instruments, Hertz, UK) was used to deposit the coating solutions on the paper at ambient temperature. This automatic machine uses standard wire-wound bars to produce a uniform and repeatable coating. Four coat weights were applied: 3, 5, 7, and 9 g/m^2, where the coat weight was varied by varying the diameter of the wire on the rod. The coating process was performed at a

speed of 6 m/min. After wet coating, the papers were dried at 40 °C for 30 min (dryer model 400, TECHPAP, Gières, France). Before testing of properties, all samples were conditioned for two days in an environmental chamber at 50% RH and 23 °C. Coating weights (g/m^2) were obtained by subtracting from the weight of a defined area of coated paper, the weight of the same size area of the uncoated paper.

Paper Thickness Measurements

Paper thickness was measured using a ProGage thickness tester (Thwing-Albert Instrument Co., Philadelphia, PA) in accordance with ISO 534-2005. Ten replicates were performed on each sample.

Water Vapor Permeability

Water vapor permeability (WVP) measurements were determined with the gravimetric method described in the AFNOR NF H00-030 standard (AFNOR 1974), with the HPMC-coated side towards the humid air. The test film was sealed in a permeation cell containing a desiccant (silica gel) to maintain an RH of 0% in the cell. The permeation cells were 6.4 cm (internal diameter) by 8.9 cm (external diameter) by 4.8 cm deep with an exposed area of 26.42 cm^2. The permeation cells were placed in a controlled temperature (38 ± 1 °C) and relative humidity (90 ± 3%) chamber fitted with a variable-speed fan to provide a strong driving force across the film for water-vapor diffusion. The water-vapor transport was determined from the weight gain of the cell. Changes in the weight of the cell were recorded as a function of time. Slopes of weight changes *vs*. time (after steady state was reached) were calculated by linear regression, and the correlation coefficient for all reported data was >0.99. The steady transfer rate was reached after ~1 h. The water-vapor transmission rate (WVTR) was defined as the slope (g/d) divided by the transfer area (m^2). After the permeation tests, film thickness was measured and WVP was calculated as follows (McHugh and Krochta 1994),

$$WVP = \frac{WVTR \cdot X}{\Delta p} \qquad\qquad \text{[g μm/m}^2\text{/d/ kPa]} \qquad\qquad (1)$$

where X is the coated paper thickness, Δp is the difference of partial water-vapor pressure across the film ($\Delta p = p(RH_2 - RH_1) = 5.942$ kPa, where p is the saturation vapor pressure of water at 38 °C, $RH_2 = 90\%$, $RH_1 = 0\%$). Four replicates were made for each type of paper.

Water Absorption Capacity

The Cobb test method was conducted for water-absorption measurement of paper samples. The water absorption was determined by measuring the amount of water absorbed by paper within 60 s. Three replicates of each type of paper were tested in a Cobb Sizing Tester (Gurley Precision Instruments, Troy, NY, USA) according to ISO 535-1991. The $Cobb_{60}$ values were expressed in g/m^2.

Tensile Testing

A material testing machine (Lloyd LRX, Lloyd Instruments, Royston, UK) with a 0.5 kN static load cell was used to determine tensile strength (TS) and elongation at break (%E) in the machine direction of the papers, according to a standard method of ISO

1924-2-1994. The experiments were performed under controlled conditions, at 23 °C and 50% RH. Ten rectangular paper samples (15 mm wide × 100 mm long) were cut from each type of coated paper and were tested using a double clamp with a separation of 30 mm at a test speed of 20 mm/min. The clamp separation was 100 mm, and the strain rate was 20 mm/min. The curve load *vs*. extension was recorded until the elongation at break was reached. The TS was expressed in MPa and was calculated by dividing the maximum load (N) by the cross-sectional area (m^2). Maximum elongation at break or percent elongation at break (%E) was determined by dividing the extension at the moment of breakage by the initial gauge length of the samples and multiplying by 100.

Tearing Tests

Tearing tests were performed on paper samples (50 mm wide × 63 mm long) according to ISO 1974-9290. Four replicates of each sample were tested in a Lorentzen & Wettre Tear Tester (Stockholm, Sweden) by the Elmendorf method. Tearing resistance, expressed in mN, is the force required to tear a test specimen after a cut has already been started. All samples were tested at 23 °C and 50% RH.

Gloss Measurements

Specular gloss was determined at an incidence angle of 60° using a flat-surface gloss meter (Elcometer 407 Statistical gloss meter, Manchester, UK), according to the ASTM standard D523 (ASTM 1999). Measurements were carried out on film specimens equilibrated at 23 ℃ and 50% RH and were taken in quintuplicate for each sample. Three films of each formulation were considered. Results were expressed as gloss units, relative to a highly polished plane surface of standard black glass with a value close to 100.

Statistical Analysis

Data were subjected to a multifactor analysis of variance with 95% significance level using Statgraphics Plus 5.1 (Manugistics, Rockville, MD, USA), while the least significant difference test (LSD, $p < 0.05$) was used to compare the different treatments and detect significant differences among storage times, at the 95% confidence level.

RESULTS AND DISCUSSION

Thickness

The thickness of coated papers varied significantly ($p < 0.05$) with coating weight and plasticizer type (Fig. 1). By increasing coating weight from 3 to 9 g/m^2, the dried thickness of HPMC-paper films was increased.

Considering that the uncoated papers had a thickness value of 98 ± 1.21 μm, HPMC could form coating layers of 3.9 to 7.4 μm thickness on the surface of the paper. However, coating thickness is not proportional to the coating weight. Indeed, the coating materials formed a continuous layer on the surface of the paper and also filled the internal porous space of the paper.

The thicknesses of the HPMC coating in this study are lower than those reported for NaCAS coating (Khwaldia 2010) and whey protein isolate coating (Han and Krochta 1999). By applying the same biopolymer, Aloui *et al.* (2011) observed similar coating thicknesses compared with this study. Reis *et al.* (2011) demonstrated using SEM images that chitosan impregnated the cellulose fibers on kraft paper, but the thickness of the

chitosan coating could not be measured due to low chitosan weight and the high penetration into the cellulose matrix. Guillaume *et al.* (2010) reported that the penetration of biopolymer into paper was related to the nature of the biopolymer, the coating weight, and the intrinsic properties of the cellulosic substrate (affinity toward biopolymer-coating solutions, topography, and porosity).

Plasticizer concentration exerted no effect on the coating thickness of GLY- and SOR-plasticized HPMC coatings on paper, while the higher the PEG concentration, the thicker the HPMC coating ($p < 0.05$). In addition, plasticized coatings with 40% and 50% of PEG were thicker compared with unplasticized coatings and coatings containing GLY and SOR. PEG-plasticized coatings were thicker than SOR-plasticized coatings, and the 50% PEG-plasticized HPMC coating was the thickest (Fig. 1). Conversely, Hong *et al.* (2005) reported that solid-phase plasticizers, including SOR and sucrose, tended to show higher coating thickness than the liquid-phase ones, such as GLY and PEG. They also pointed out that the coating thickness increased with the molecular size of the plasticizers used.
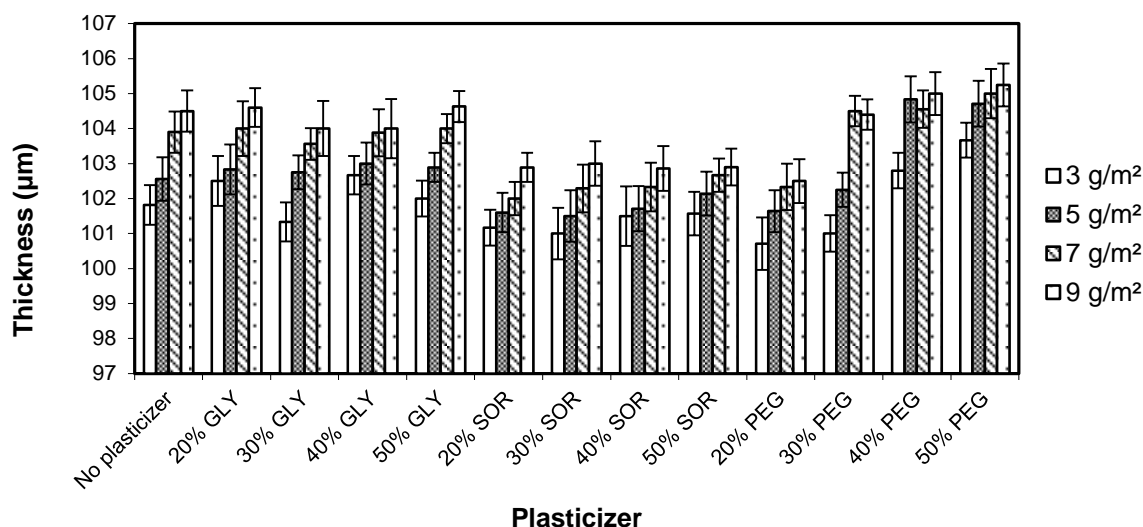

Fig. 1. Thickness of HPMC-coated paper as affected by coating weight and plasticizer type and amount. Mean values and LSD intervals are shown.

Water Vapor Permeability (WVP)

Transport properties of coated packaging materials are affected by coating composition (Tihminlioglu *et al.* 2010). Therefore, the detailed study of the water-vapor–barrier properties of the HPMC-coated papers as influenced by coating composition is of great importance for practical and commercial purposes.

Figure 2 shows WVP of the HPMC-paper films without and with various plasticizers and plasticizer contents at different coating weights. Multifactor ANOVA reveals that coating weight and coating composition (plasticizer type and amount) led to differences in the WVP response, the latter being more significant. The WVP of HPMC-coated papers plasticized with polyols increased slightly with increasing coating weight from 3 to 9 g/m^2. The WVP of uncoated papers was 581 ± 21 g $\mu m/m^2/d/kPa$. So, with the exception of the coatings formulated without plasticizers, which reduced WVP by 25% for 3 g/m^2, the rest did not provide any barrier against water vapor. Indeed, the WVP values of paper coated with formulations containing plasticizers were greater than

those of uncoated paper. This result was contradictory to the decrease in WVP of NaCAS-coated paper, which was attributed to a clogging by the coating material of the pores of the cellulose structure of paper (Khwaldia 2010). Han and Krochta (1999) reported that whey protein isolate coating on paper reduced WVP by 44.8% for 10 g/m^2 coating weight compared with that of the uncoated paper. The different effect of biopolymer coating on WVP of paper is dependent on the type of coating material used, which reflected the difference of interaction between the biopolymer and the structural cellulose fiber of the paper (Rhim *et al.* 2006; Sothornvit 2009).
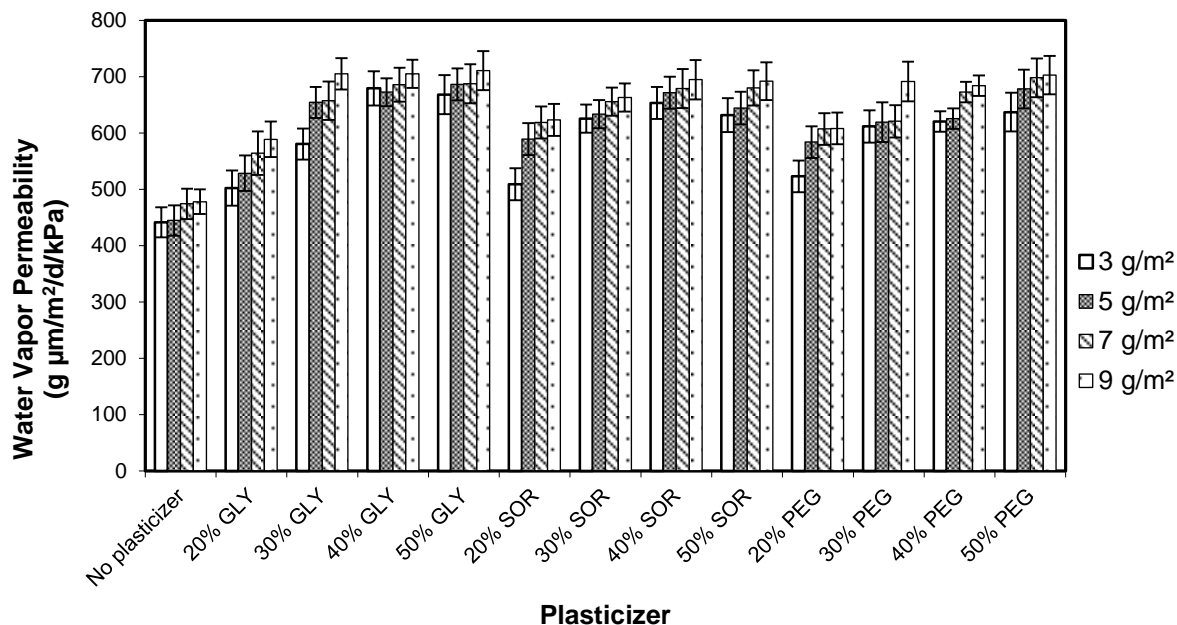

Fig. 2. Water vapor permeability of HPMC-coated paper as affected by coating weight and plasticizer type and amount. Mean values and LSD intervals are shown.

On the other hand, WVP was lower for HPMC-paper films without plasticizers compared with HPMC-paper films plasticized with polyols at all coating weights ($p < 0.05$). It was found that the WVP of coated papers increased with increasing plasticizer content (Fig. 2). The addition of a plasticizer modifies the properties of the coating by increasing the free volume of chain segments and the mobility of polymer chains, which may promote water-vapor diffusivity through the film and, hence, accelerate the water-vapor transmission (Orliac *et al.* 2003). As all the plasticizers used were hydrophilic, an increase in their concentration favored the adsorption of more water by the network and, thus, a higher level of vapor transfer. For all studied plasticizers, the lowest WVP values of coated papers with polyols were observed for 20% of GLY. The extent of the increase in WVP produced by increased GLY concentration was greater than those for the other two plasticizers.

Donhowe and Fennema (1993) studied the effects of various plasticizers on the WVP of methylcellulose films. Water-vapor–barrier properties were the best for unplasticized methylcellulose films, and incorporation of plasticizers, PEG, propylene glycol, and GLY into the methylcellulose films resulted in significant increase in WVP values. GLY is reported to have the greatest effect on WVP. Ayranci *et al.* (1997) studied the influence of the molecular weight of plasticizer on water-vapor–barrier performance of

HPMC-based edible films, and they showed that WVP values decrease sharply with increasing molecular weight of PEG.

Water Absorption

The water-absorption capacity of coated paper, controlled by the type of cellulosic substrate and the nature of coating materials, reflects the paper resistance to water. The water resistance of the biopolymer-coated papers was measured through direct contact with the surface of the papers with water. Figure 3 shows water absorption of the HPMC-paper films without and with various plasticizers and plasticizer contents at different coating weights. An ANOVA reveals that coating weight and coating composition significantly affected ($p < 0.05$) water absorption values. Water absorption of HPMC-coated papers increased with increasing coating weight from 3 to 9 g/m^2. A similar effect of coating weight also has been reported for pulp papers coated with whey protein isolate (Han and Krochta 1999). The water absorption of uncoated papers was 25.44 ± 0.42 g/m^2. So, HPMC coatings formulated without plasticizers reduced water absorption by 25% for 3 g/m^2. This water-absorption reduction obtained in this study was comparable to that obtained by Reis *et al.* (2011) after chitosan coating with a coat weight of 3.5 g/m^2.
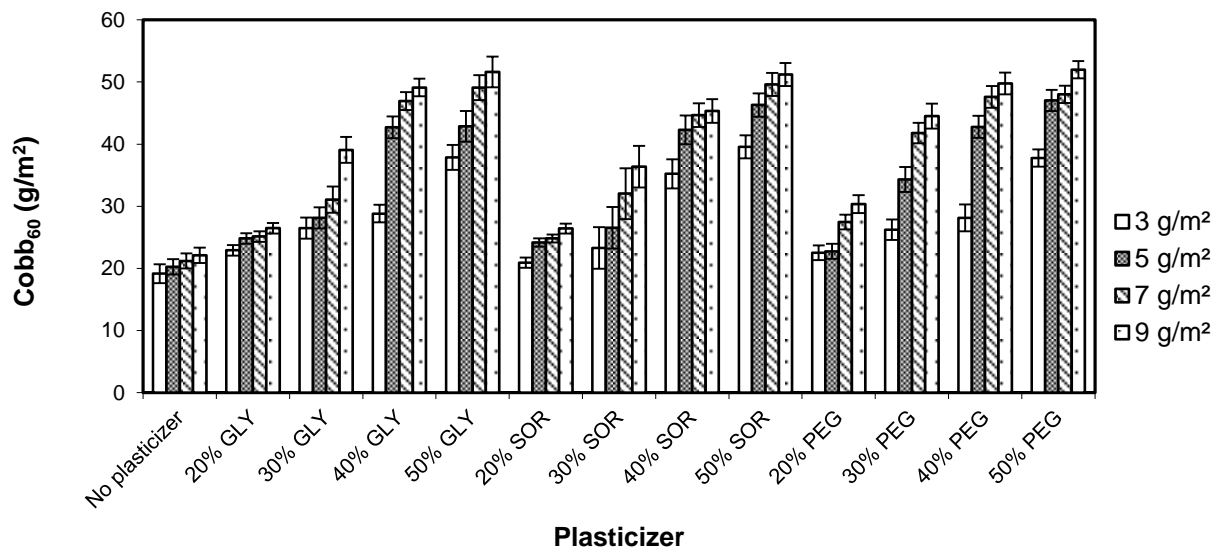

Fig. 3. Water absorption of HPMC-coated paper as affected by coating weight and plasticizer type and amount. Mean values and LSD intervals are shown.

On the other hand, water absorption was lower for HPMC-paper films without plasticizers compared with HPMC-paper films plasticized with polyols at all coating weights ($p < 0.05$). The water absorption of coated papers increased with increasing plasticizer content (Fig. 3). It can be noted that the highest $Cobb_{60}$ values, in the 38 to 52 g/m^2 range, were obtained for papers coated with the highest plasticizer concentration. The increase in water absorption with plasticizer concentration is associated with the hydrophilic nature of plasticizers used. As all the plasticizers tested were hydrophilic, an increase in their amount favored the absorption of more water by the network and, thus, a lower resistance to water. In addition, HPMC-coated papers with PEG as a plasticizer showed significantly higher Cobb60 values compared with the other plasticizers at 20% and 30% plasticizers ($p < 0.05$). However, the plasticizer type did not significantly affect

the water absorption of HPMC-paper films at high plasticizer contents (40% to 50%).

Tensile Properties

High mechanical properties are usually required to maintain packaging integrity during shipping, handling, and storage. Desirable packaging materials should possess both adequate mechanical strength and flexibility. Tensile strength (TS) and elongation at break (%E) are the most commonly reported responses to describe mechanical properties of paper-based packaging materials. TS is a measure of the ability of a film to resist breaking under tension, which is dependent on the strength of fibers, their surface area, and length, and also on the bonding strength between them. %E shows the ability of a film to stretch before it breaks (Rabinovitch 2003).
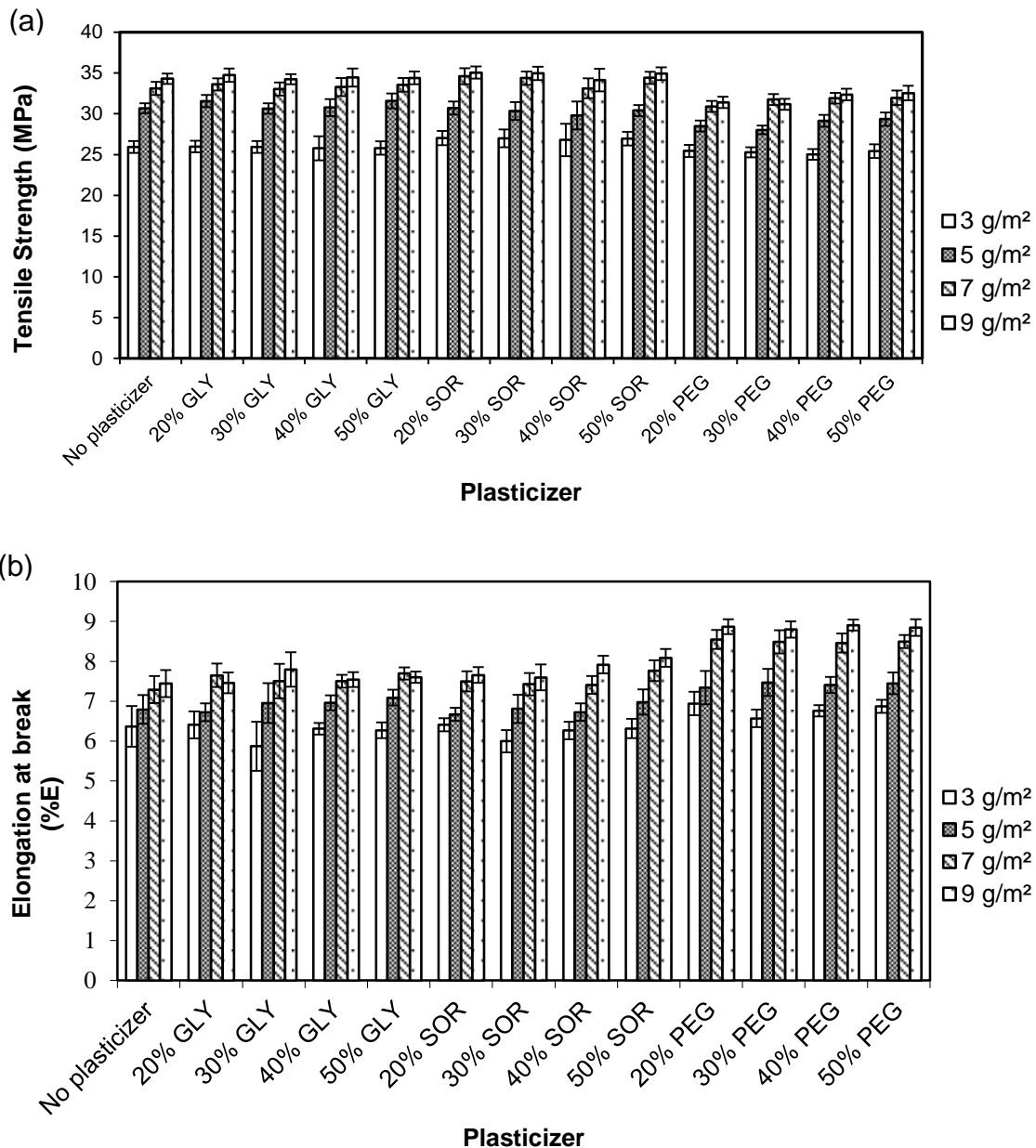
Fig. 4. Tensile strength (a) and elongation at break (b) of HPMC-coated paper as affected by coating weight and plasticizer type and amount. Mean values and LSD intervals are shown.

In general, the mechanical properties of the coated/laminated films in composite structure are controlled by the substrate or base film (Hong *et al.* 2005). However, tensile properties of HPMC-coated papers were shown to be mainly dependent on the coating weight (Fig. 4). The TS of coated papers increased with increasing coating weight from 3 to 9 g/m^2. At 30% GLY concentration, HPMC coating on paper enhanced TS by 43% for 9 g/m^2, 38% for 7 g/m^2, and 28% for 5 g/m^2, compared with that of the uncoated paper (23.94 $\pm$ 0.60 MPa). A similar effect of coating weight has been also reported for paper sheets coated with chitosan, whey protein isolate, whey protein concentrate, and wheat gluten protein (Gällstedt *et al.* 2005). This seems to be consistent with our previous findings on TS values of HPMC- and chitosan-coated papers (Aloui *et al.* 2011). Conversely, Rhim *et al.* (2006) and Sothornvit (2009) reported a decrease in TS after biopolymer coating because of the interference of coating materials with molecular interactions among the fibers.

On the other hand, the increase in %E as coating level increased, as shown in Fig. 4(b), was probably caused by a stress relaxation in the base paper during the coating process when the base paper was exposed to the water in the coating solution (Kjellgren *et al.* 2006; Khwaldia 2010).

It was found that the tensile parameters (TS, %E) of papers coated with formulations containing plasticizers were not affected by plasticizer concentration. Moreover, no effect was found with GLY and SOR on TS and %E of coated papers compared with those without plasticizer (Fig. 4). However, as reflected in Fig. 4, PEG had a plasticizing effect on HPMC coatings. Indeed, incorporation of PEG caused a decrease in TS and an increase in %E of the HPMC-paper films. This implied that PEG could make HPMC coating flexible. Park *et al.* (1993) and Cao *et al.* (2009) also found that the addition of PEG led to a decrease in TS and elastic modulus, which led to an increase in %E of hydroxypropyl cellulose and gelatin films, respectively.

HPMC-coated papers with PEG as a plasticizer showed significantly lower TS and higher %E, compared with the other plasticizers ($p < 0.05$). Nevertheless, no statistically significant difference was found between TS and %E of coated papers containing GLY and SOR. GLY and SOR having similar chemical structures of straight-chain molecules did not produce any significant difference in tensile properties of HPMC-coated papers.

Plasticizer physiochemical properties, such as chemical structure, shape, polarity, chain length, physical state, and number of active functional groups determine its ability to plasticize a polymer network. Although PEG is also considered in the same polyol plasticizer category as GLY and SOR, it showed higher plasticizer efficiency as evidenced by the greater %E than the other plasticizers. The differences in plasticizing effect between plasticizers were possibly due to the different availability of oxygen atoms for hydrogen bonding. The spacing of O atoms in PEG 200 may have allowed more room for the formation of H-bonding with biopolymer chains (Sothornvit and Krochta 2000).

With all the plasticizers used in this study, the range of TS and %E values of HPMC-coated papers was approximately 25.80 MPa to 35.06 MPa and 5.87% to 8.87%, respectively. The TS and %E of coated papers were greater than that of uncoated paper (23.94 $\pm$ 0.60 MPa; 4.52 $\pm$ 0.32%), which means that HPMC coating improved paper strength and ductility. Our results are in agreement with those of Vartiainen *et al.* (2004), who reported that chitosan coating improved the tensile properties of paper and could be regarded as a reinforcement layer. The TS and %E values for HPMC-coated papers in this study are similar to those obtained by Rhim *et al.* (2006) for alginate- or soy protein-

coated paperboards. HPMC-coated papers exhibited similar TS and higher %E compared with chitosan-coated papers (Aloui *et al.* 2011).

Tearing Resistance

The tearing resistance corresponds to the average force applied during the tearing operation; it is likely that it relates to the fracture stress and/or fracture resistance or toughness of the material (Rabinovitch 2003).

The HPMC-paper films showed an increase in tearing resistance with increasing coating weight from 3 to 9 g/m^2 (Fig. 5). At 30% PEG concentration, HPMC coating on paper enhanced tearing resistance by 31% for 9 g/m^2, 16% for 5 g/m^2, and 13% for 3 g/m^2, compared with that of the uncoated paper (433 ± 15 mN). The increases in tearing resistance arising from increases in coating weight reported in this study were consistent with the findings of Gällstedt *et al.* (2005) and Khwaldia (2010), who reported that increasing biopolymer coating weight increased tearing resistance and toughness of the paper. The tear index of paper, however, was not affected by HPMC coating (Sothornvit 2009).
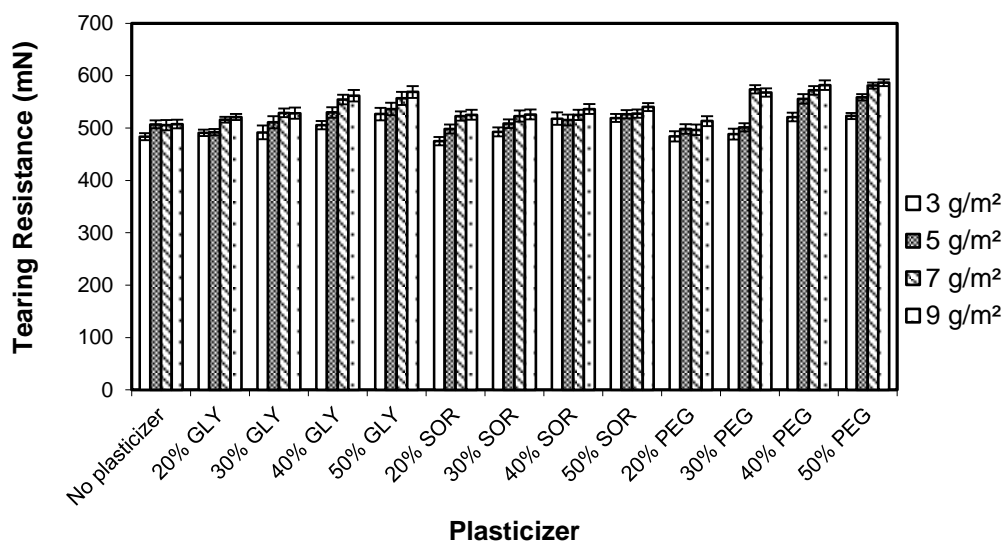

Fig. 5. Tearing resistance of HPMC-coated paper as affected by coating weight and plasticizer type and amount. Mean values and LSD intervals are shown.

As shown in Fig. 5, plasticizer concentration affects tearing resistance ($p < 0.05$). The tearing resistance of coated papers increased as the amount of plasticizer in the coating increased. However, no significant differences were observed between the tearing resistance of HPMC-paper films without plasticizer and those at 20% plasticizer. HPMC-coated papers with PEG as a plasticizer showed significantly higher tearing resistance compared with the other plasticizers at 30% to 50% plasticizers ($p < 0.05$).

Gloss

Gloss relates to the ability of a surface to direct reflected light. Specular gloss is used mainly as a measure of the shiny appearance of films and surfaces. Gloss is a relevant property of the coating since it has a direct impact on the appearance of the coated product.

Specular gloss of HPMC-paper films were found to depend significantly on coating weight and coating formulation (plasticizer type and amount), the latter being more significant (Fig. 6).
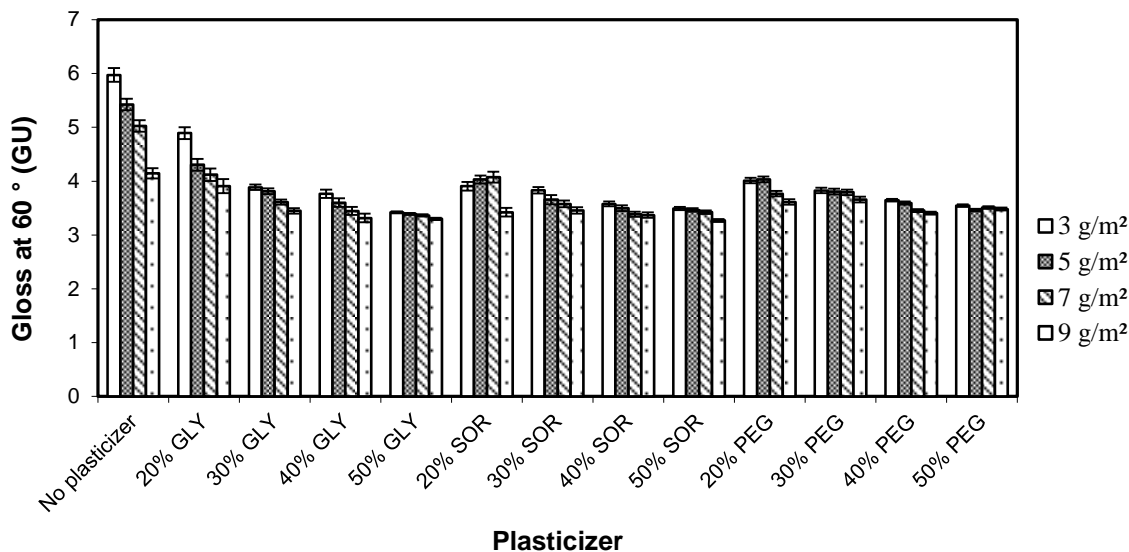

Fig. 6. Specular gloss of HPMC-coated paper as affected by coating weight and plasticizer type and amount. Mean values and LSD intervals are shown.

Gloss of HPMC-coated papers decreased with increasing coating weight from 3 to 9 g/m^2. The gloss values of HPMC-paper films without plasticizers were significantly greater than those for the uncoated paper (3.51 ± 0.04 GU). So, with the exception of the coatings containing high plasticizer content (40% to 50%), the rest showed a tendency to have a more glossy surfaces than the uncoated cellulosic substrate. This seems to be consistent with the finding of Han and Krochta (2001), who reported that the WPI coating increased surface smoothness and homogeneity, resulting in a glossier surface after coating. It was also reported that HPMC-coated polypropylene films had higher gloss than methylcellulose- and dextrin-coated films (Hong *et al.* 2005).

On the other hand, plasticizer-free HPMC coatings provided paper with the highest gloss, and incorporation of plasticizers PEG, SOR, and GLY into the HPMC coatings resulted in significant decreases in gloss values (Fig. 6).

Among the plasticizers applied, PEG conferred more glossy surfaces to HPMC-coated paper than the other plasticizers. The observed decrease of gloss as the plasticizer amount increases in the coating may be caused by the surface of the coated paper becoming more rough and heterogeneous.

Gloss is known to be strongly related to the surface morphology reached during the film drying. At higher plasticizer contents, coating heterogeneity due to polymer–plasticizer phase separation may result in lower gloss values (Trezza and Krochta 2001). Conversely, Hong and Krochta (2004) found no significant difference in variations of gloss as a function of GLY content. Villalobos *et al.* (2006) observed a decrease in gloss as the surfactant ratio increased in the HPMC film.

CONCLUSIONS

1. The barrier properties of the coated papers were shown to be more dependent on the coating formulation than on coating weight, while the mechanical properties of the coated papers were mainly dependent on the coating weight.

2. The application of plasticizer-free HPMC coatings provided paper with the highest barrier properties and gloss. WVP and water absorption capacity of coated papers increased with increasing plasticizer concentration and coating weight.

3. Incorporation of PEG caused a decrease in TS and an increase in %E of the HPMC-paper films. PEG could make HPMC coating flexible and is more suitable as HPMC plasticizer than GLY and SOR because it shows higher plasticization efficiency. Furthermore, the use of hydrophilic plasticizer has negative effects on barrier properties, as shown in the results of WVP and water absorption capacity. However, plasticizer is apparently required to obtain coatings with good flexibility and to keep their integrity once applied and subsequently formed.

4. HPMC coating provided paper with desirable mechanical properties, and the resulting HPMC-coated papers showed good appearance, flexibility, and adhesion between the coating and the cellulosic substrate. HPMC seemed to be an appropriate coating material for paper-based packaging materials.

ACKNOWLEDGMENTS

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