

Edible Arabinoxylan-Based Films. 1. Effects of Lipid Type on Water Vapor Permeability, Film Structure, and Other Physical Characteristics

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Arabinoxylans (AX) are natural fibers extracted from maize bran, an industrial byproduct. To promote this polymer as a food ingredient, development of edible coatings and films had been proposed. Indeed, composite arabinoxylan-based films were prepared by emulsifying a fat: palmitic acid, oleic acid, triolein, or a hydrogenated palm oil (OK35). Lipid effects on water vapor permeability (WVP), surface hydrophobicity (contact angles), lipid particle size, and mechanical properties were investigated. Results showed that OK35–AX emulsion films had the lowest WVP. Emulsified films presented a bimodal particle size distribution; however, the smallest particle mean diameter (0.54 μm) was observed in OK35–AX emulsion films. Contact angles of water comparable to those observed for LDPE films ($>90^\circ$) are measured on the OK35–AX film surface. Finally, only triolein–AX emulsion films had elongation higher than films without lipid. These results suggest that OK35 enhances functional properties of AX-based films and should be retained for further research.

KEYWORDS: Arabinoxylans; edible film(s) and coating(s); water vapor permeability; mechanical properties; surface hydrophobicity; emulsion

INTRODUCTION

The potential of edible films to control water transfer and improve food quality and shelf life has been extensively reviewed (1–3). In general, polysaccharides and proteins have good mechanical properties and are excellent gas, aroma, and lipid barriers, but they are inefficient against water transfer. Lipids, on the other hand, offer high water barrier properties, but they form brittle films. Several research groups have attempted to improve the water vapor barrier properties of protein- or polysaccharide-based films by adding a lipid as an emulsion (4–6) or by laminating a lipid layer onto a previously formed film (7–9). Compared to bilayer films, emulsified films are less efficient water vapor barriers, but they require only a single step, exhibit good mechanical and adhesive properties, and can be made at room temperature (6, 7).

Arabinoxylans (AX) constitute a major fraction of cereal cell wall polysaccharides. They consist of a linear $\beta(1\rightarrow4)$ -linked xylan backbone to which α -L-arabinofuranose units are attached as side residues via $\alpha(1\rightarrow3)$ and/or $\alpha(1\rightarrow2)$ linkages. A number of studies focus on the extraction and structural characteristics

of these polysaccharides (10–12). The great functional role of arabinoxylans in the bread-making process has been largely reported (13, 14). However, the contribution of arabinoxylans to the malting and brewing qualities of barley grains has not yet been well elucidated (12, 14). Chanliaud (10) has developed arabinoxylan-based films. They have good tensile resistance and are an effective barrier against oxygen or carbon dioxide. Furthermore, AX films offer interesting water vapor permeability. Nevertheless, these functional properties are lower than those observed for plastic packaging. To enhance these performances, lipid addition has been suggested.

Many studies have reported the impact of lipid type on water vapor permeability (WVP) of emulsified films. Generally, the water transmission rate of a film increases as the length of the lipid hydrocarbon chain decreases and the unsaturation degree increases (4, 7, 15). Additionally, a film's water vapor resistance is inversely related to lipid polarity. Hydrophobic alkanes and waxes, such as paraffin and beeswax, are the most effective barriers (5, 15, 17). More specific fat characteristics such as physical state, conformation, polymorphism, and viscoelastic properties may also influence the WVP of the films.

Lipid particle size has an effect on the water vapor permeability of emulsified films. In fact, it was found that the decrease of lipid particle size correlates well with the reduction of WVP (4, 18). According to the tortuosity model (19), this result could be explained by the presence of a large number of spherical

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particles uniformly dispersed that increases the distance traveled by water molecules diffused through the film. In the case of a protein/beeswax emulsified film, this tendency was attributed to the immobilization of protein chains at the lipid interface, with the resulting formation of a more ordered and tightly cross-linked structure with a lower permeability (20, 21).

To better understand interfacial interactions in emulsions, a low molecular weight anionic surfactant, sodium dodecyl sulfate (SDS), has been used. SDS is one of the most strongly competitive surfactants approved for food use. Research studies describe the interactions between SDS and an uncharged water soluble macromolecule, ethyl(hydroxyethyl)cellulose (22, 23). The initial step in the surfactant-polymer interaction consists of a redistribution of the surfactant with a preference for the polymer coil regions over the bulk solution. Next, surfactant molecules cluster around the hydrophobic sites on the polymer. Finally, polymer molecules wrap around the surfactant micelles with other hydrophobic parts inserted between the surfactant molecules. Accordingly, in whey protein-based emulsions, particle size distribution has been shown to strongly depend on the surfactant/protein ratio (24, 25) as well as pH.

The effects of lipid type on mechanical properties have also been examined. Generally, lipid addition has a negative impact on mechanical properties because of low interactions between fats and polysaccharides or proteins (5, 17, 26), and lipids are unable to form an internal interconnecting lipid network.

The major objectives of this study were to develop arabinoxylan-lipid films from emulsions and to determine the effects of fat type (palmitic acid, stearic acid, triolein, and hydrogenated palm oil) on water vapor barrier properties, film structure, and surface and mechanical properties.

MATERIALS AND METHODS

Materials. Food grade AX extracted from maize bran were donated by ULICE (Riom, France). Anhydrous glycerol for plasticizing was purchased from Fluka (Sigma-Aldrich Chimie, Saint Quentin Fallavier, France; 98% purity). Glycerol monostearate (GMS) employed as emulsifier was purchased from Prolabo (Merck eurolab, Fontenay-sous-Bois, France; 99% purity). The following fats were used to prepare edible composite films: palmitic acid (C_{16}) (Prolabo Merck eurolab, Fontenay-sous-Bois, France; 97% purity), stearic acid (C_{18}) (Prolabo Merck eurolab; 96% purity), triolein (Sigma-Aldrich Chimie; 65% purity), and hydrogenated palm oil (OK35) (SIO, Saint Laurent Blangy, France). SDS (Prolabo Merck eurolab; 98% purity) and the hydrophobic dye Sudan III red (Sigma-Aldrich Chimie) were used for the characterization of film structure. Two commercial synthetic films, cellophane 300P (Courthauld, U.K.) and a low-density polyethylene Riblène FF30 (LDPE) (EniChem Polymères, Mazingarbe, France; 37% crystallinity), were used as hydrophilic and hydrophobic models for this study.

Arabinoxylan Film Preparation. An AX film-forming solution was prepared by dispersing 16 g of AX powder in 100 mL of osmosed water at 75 °C for 40 min with constant stirring. During the heating and stirring, anhydrous glycerol was then added at a concentration of 15% (w/w total dry matter). A vacuum was applied with a vacuum pump to remove dissolved air bubbles. Then, the film-forming solution was poured into a thin layer chromatography spreader and cast onto glass plates previously covered by an adhesive PVC sheet to prevent the sticking of dried films. To evaporate the solvent (water) and thereby form a network, the solution was dried in a ventilated chamber (WTB Binder, Labortechnik, GmbH) for ~4 h at 40% relative humidity and 30 °C. These drying conditions allowed reasonably quick formation of homogeneous AX films that could be peeled intact from their supports. To prepare emulsified films, blends of lipids and GMS (90:10 w/w) were made. The emulsifier concentration of ~10% (w/w) was chosen in agreement with the results found by Debeaufort (18, 27), who showed that, at this concentration, fine and stable emulsions with a unimodal

particle size distribution are obtained. The GMS-lipid mixture was added at a concentration of 30% (w/w total solid basis) to the plasticized film-forming solution composed of arabinoxylans and glycerol. After the lipids had been melted, the hot solution was emulsified with a homogenizer (Ultra-Turrax model T25 IKA, Labortechnik, GmbH) for 2 min at 24000 rpm. These emulsifying parameters, chosen in agreement with those of Debeaufort (18, 27), were checked to ascertain that no change occurred in particle size after 2 min of homogenization. Afterward, the film-forming emulsion was degassed and dried in the same conditions previously specified. For each film formulation a minimum of three films were made and completely characterized in order to validate film preparation procedures.

Film Thickness Measurement. The thickness of conditioned (22% relative humidity, 25 °C, and 48 h) films was determined by employing an electronic gauge (Multi Check FE, List-Magnetik, GmbH) with a precision of 1 μm . The average values of 30 thickness measurements per type of film were used in all WVP calculations.

Water Vapor Transfer Rate (WVTR) and Permeability Measurement. The WVTR was gravimetrically determined at 25 °C using an AFNOR modified procedure (8). Before WVTR determination, all films were equilibrated at 22% relative humidity using a saturated salt solution of potassium acetate, at 25 °C for 48 h. Four samples, 4.2 cm diameter disks, were cut from each film. Each sample was placed between two Teflon rings on the top of a glass cell containing a saturated salt solution of potassium chloride with a water activity of ~0.84 at 25 °C. Test cells were weighed and introduced into a climate-controlled chamber (WTB Binder, Labortechnik, GmbH) regulated at a relative humidity of 22% and 25 °C. Cells were weighed three times per day for 7 days. At the steady state, the WVTR ($\text{g}/\text{m}^2\cdot\text{s}$) and WVP ($\text{g}/\text{m}\cdot\text{s}\cdot\text{Pa}$) coefficients were defined as follows:

$$\text{WVTR} = (\Delta M/\Delta t)A \quad (1)$$

$$\text{WVP} = (\text{WVTR} \times e)/\Delta p \quad (2)$$

where $\Delta M/\Delta t$ is the amount of moisture lost per unit of time (g/s), A is the film area exposed to moisture transfer ($8 \times 10^{-4} \text{ m}^2$), e is the film thickness (m), and Δp is the water vapor pressure difference (Pa) between the two sides of the film. For each type of film, a minimum of three water vapor determinations, each one including the four test cells, were made.

Surface Hydrophobicity. The surface hydrophobicity of the films was evaluated from contact angle measurement using a goniometer (Krüss GmbH). A droplet of osmosed water was placed on the film, and the angle of the tangent to the basis of the droplet (contact angle) was measured and expressed in degrees. A minimum of five measurements per film were carried out.

Film Structure Analysis. Laser light scattering and optical microscopy were used to study AX film structure. For each emulsified film, the droplet size distribution, the weight mean diameter ($D_{4,3}$, De-Broucker mean), and the volume-surface mean diameter ($D_{3,2}$, Sauter mean) were determined by laser light scattering using a Malvern Mastersizer S2-01 (Malvern Instruments Ltd.). An average diameter is the diameter of a hypothetical particle that represents the total number of particles in the sample (28, 29). The weight mean diameter ($D_{4,3}$) is the average size based on the unit weight of the particles; meanwhile, the volume-surface diameter ($D_{3,2}$) represents the average size based on the specific surface per unit volume. These mean particle diameters were defined as follows:

$$D_{4,3} = \frac{\sum n_i d_i^4}{\sum n_i d_i^3} \quad (3)$$

$$D_{3,2} = \frac{\sum n_i d_i^3}{\sum n_i d_i^2} \quad (4)$$

In eqs 3 and 4, n_i is the number of droplets in each size class and d_i is the droplet diameter. The particle size distribution is displayed by using a frequency curve that allows us to show peaks in the graph and to compare results from different measurements. The x -axis represents

the logarithm of the particle size. The y -axis is a linear scale and represents the relative volume fraction in percent of particle that belongs to specified size classes on the particle size axis. The relative volume fraction (φ) was defined as follows:

$$\varphi(\%) = (v_i/V) \times 100 \quad (5)$$

In eq 5 v_i is the volume of the droplets on the i th size class and V is the total volume of all the droplets in the emulsion. For instance, a relative volume fraction of $\sim 18.2\%$ means that the total volume of all particles with diameters in this specified range represents 18.2% of the total volume of all particles in the distribution.

Before analysis, the samples were prepared as follows: first, 1 g of dried film was dispersed in 50 mL of osmosed water at room temperature with moderate magnetic stirring. To study the incidence of lipid aggregation, films were also dispersed in 50 mL of a 0.1% SDS solution. The refractive indices of the lipid phase to water were 1.0751 and 1.0724 for palmitic acid and stearic acid, respectively, and 1.0966 for both triolein and hydrogenated palm oil. The optical model 0405 was used for fatty acid emulsions, and model 0505 was used for triolein and hydrogenated palm oil. Focal lengths of 45 and 300 mm were used to measure mean diameters from 0.1 to 600 μm . The absorbance was measured at 633 nm laser light. Lipid droplets were sized using water as a dispersant, and all measurements were carried out at room temperature. Three measurements per film were carried out.

Lipid particles, previously marked with Sudan III red, were observed with an inverted-light contrasted binocular microscope (500 \times objective, Leitz Labovert). The images were recorded with a CCD camera (model 6710, Cohu) associated with an image analysis system (Imaging Technology Inc., computer type PC 386, software Visilog 3.6, Noesis). Four samples of each film were prepared by dispersion in cold water. Samples, held between two glass plates, were observed and analyzed.

Tensile Tests. Determination of tensile strength (TS), percentage of elongation at break (TE), and elastic modulus (EM) of the films was performed with an Instron Universal Testing Machine (model 1122, Instron Engineering Corp., Canton, MA) with a 5 kN load cell. A total of 20 samples, 6 \times 2 cm strips, were cut from each type of film and conditioned at 54% relative humidity and 25 $^{\circ}\text{C}$ for 10 days. Cross-head speed was set at 100 mm/min, and all tests were made at room temperature.

Statistical Analysis. All data were analyzed and compared by using variance analysis and Student–Newmans–Keuls t tests ($\alpha = 0.05$) on SAS software (SAS Institute Inc., version 6.02, Cary, NC).

RESULTS AND DISCUSSION

Water Vapor Permeability. AX-based films have a range of thickness values from 65 to 100 μm . In a previous study (results not published), when film thickness increased was from 50 to 150 μm , the WVP of AX-based films increased, while the WVTR decreased and tended to reach a plateau. For this reason, WVTR and WVP are both expressed and given in **Table 1**.

Homogeneous AX-based films are smooth, brown, translucent, and 91 μm in thickness. The WVP of this film is $\sim 1.77 \times 10^{-10}$ g/m \cdot s \cdot Pa, as shown in **Table 1**. This high permeability is due to the hydrophilic nature of AX polymers, which are natural fibers with a great hydration capacity (14). Consequently, AX films are soluble in water at 30 $^{\circ}\text{C}$. However, our WVP value is twice higher than that of the AX film tested by Chanliaud (10). This difference is probably due to, first, the purity of the AX powder; second, the differences in the preparation of the films; and, finally, the experimental conditions employed for WVP determination, which are not the same. In comparison with other edible films, the WVP of AX films is comparable to the value obtained for methyl cellulose films (30, 31) and less than that of amylose films (32). Moreover, AX films are more permeable than protein films (26, 33, 34). Finally,

Table 1. Thickness, Water Vapor Transmission Rate, Permeability, and Contact Angle of Different Arabinoxylan-Based Films and Plastic Packagings^a

film	thickness (μm)	WVTR ($\times 10^{-3}$ g/m 2 \cdot s)	WVP ^b ($\times 10^{-10}$ g/m \cdot s \cdot Pa)	contact angle ^c (deg)
arabinoxylans	90.8 \pm 6.6a	3.92 \pm 0.14b	1.77 \pm 0.06a	70.8 \pm 5.1b
AX–C ₁₆	89.9 \pm 9.8a	3.37 \pm 0.09c	1.52 \pm 0.04b	64 \pm 4.6b
AX–C ₁₈	81.6 \pm 5.4b	2.91 \pm 0.12d	1.19 \pm 0.05c	68.6 \pm 3.6b
AX–triolein	66.6 \pm 6.7c	3.52 \pm 0.15c	1.18 \pm 0.05c	39 \pm 8.5c
AX–OK35	89.8 \pm 9.1a	2.71 \pm 0.17d	1.24 \pm 0.06c	94.4 \pm 2.1a
cellophane	20	6.17 \pm 0.63a	0.69 \pm 0.07d	
LDPE	25	0.13 \pm 0.01e	0.019 \pm 0.001e	100.7 \pm 11.4a

^a Values in a column followed by the same letter are not significantly different at the risk of 5%. ^b Means of at least three replicates \pm standard deviation for WVP determination. ^c Mean of a minimum of 15 measurements for contact angle determination. (Contact angle of water on cellophane film cannot be measured because of the highly hydrophilic nature of this polymer.)

the WVP of AX films is 3–100 times higher than that of commercial plastic films.

Effects of Lipid Type on Water Barrier Properties and Film Structure. Lipid addition changed the appearance of films, which became more opaque. To eliminate film orientation or phase separation effects, the film face that was in contact with the glass plate was always exposed to the atmosphere with the highest relative humidity (84%) during WVP measurement. In all cases, the addition of fats significantly ($P < 0.05$) decreased the WVTR of AX-based films (**Table 1**). The WVTR and WVP of AX–C₁₈ films are lower than those of AX–C₁₆ films. These results are also described by other researchers (4, 7, 15) who have observed that WVP decreases as the chain length of fatty acid increases from lauric acid (C₁₂) to stearic acid (C₁₈). Even though all of these fatty acids are solid at 25 $^{\circ}\text{C}$, the fatty acids with a shorter chain length have a greater chain mobility, are more polar, and may not possess the essential extensibility for the organization of an interlocking network (4). The AX–triolein film presents the highest WVTR. This could be due to the presence of double bonds (unsaturations) in the lipid structure, which enhances the polar properties of this triglyceride. The WVTR in AX–OK35 film is not significantly ($P < 0.05$) different from that in the AX–C₁₈ film. This result could be explained by the OK35 composition (35). In fact, this fat is a blend of triglycerides mainly composed of lauric acid (35%), stearic acid (25%), myristic acid (18.3%), and palmitic acid (11.8%). Kamper and Fennema (7) showed that emulsified film made from the blend of stearic acid and palmitic acid has WVP similar to that of the stearic acid emulsified film, and, C₁₈–C₁₆ emulsified films are easier to prepare and have a more consistent appearance resulting from the lowering of lipid melting point.

Triolein and hydrogenated palm oil (OK35) have already been used as hydrophobic substances in emulsified films by Quezada-Gallo (35). He found that the WVP of wheat gluten–OK35 films (0.74×10^{-10} g/m \cdot s \cdot Pa) is lower than that of wheat gluten–triolein films (0.97×10^{-10} g/m \cdot s \cdot Pa). It has been shown in this work that the WVPs of methyl cellulose–OK35 films and methyl cellulose–triolein films are not significantly ($P < 0.05$) different. Such a tendency is also observed with the WVP of AX-based films. In the present study, wheat gluten and its components form films that are less permeable and more hydrophobic than methyl cellulose- and AX-based films. The most effective barriers are obtained when stearic acid, triolein, or hydrogenated palm oil is emulsified in the AX network. In comparison with synthetic films, the WVP of emulsified AX-based films remain 3–100 times higher. As was noticed by

Table 2. Weight Mean Diameter ($D_{4,3}$) and Volume-Surface Mean Diameter ($D_{3,2}$) of Lipid Particles Emulsified in the Arabinoxylan Matrix^a

emulsified lipid	water		SDS solution	
	$D_{3,2}$ (μm)	$D_{4,3}$ (μm)	$D_{3,2}$ (μm)	$D_{4,3}$ (μm)
C ₁₆	6.03 \pm 0.13	21.17 \pm 0.1	2.09 \pm 0.13	44.64 \pm 0.79
C ₁₈	9.46 \pm 0.18	19.77 \pm 0.34	2.35 \pm 0.06	33.63 \pm 0.39
triolein	2.38 \pm 0.08	15.49 \pm 0.54	5.35 \pm 0.11	64.74 \pm 1.6
hydrogenated palm oil	0.54 \pm 0.04	17.21 \pm 0.15	0.58 \pm 0.02	19.99 \pm 0.38

^a Mean of three measurements \pm standard deviation.

Krochta (19), these results suggest the WVP of emulsified films depends mostly on the nature of the continuous phase and, second, on the type of fat employed. In fact, a hydrophilic polymer matrix is quite compatible with water vapor and offers little resistance to water transfer.

Other parameters such as lipid particle size and distribution could influence the water resistance of the films. In emulsion systems, the particle size of the lipid droplets is usually reported as mean particle values (28, 29). These values represent different diameters of a hypothetical sphere that represents the total number of particles in the sample. However, emulsification gives a distribution of lipid particles, with lipid droplets having larger and smaller particle sizes than the mean particle value. In our study, to more easily interpret results, values are given in **Table 2**, first, as the volume-surface mean diameter ($D_{3,2}$), also known as the Sauter mean, which represents the average size based on the specific surface per unit volume and better characterizes small and spherical particles. The second mean diameter commonly used is the weight mean diameter ($D_{4,3}$), also called the DeBroucker mean, which is the average size based on the unit weight of the particles and represents larger particles with an irregular form such as aggregates. **Figure 1** shows the particle size distribution profiles of AX-lipid emulsified films. These frequency curves represent the relative volume ratio in percent versus the particle size. After film dissolution in water, palmitic acid and stearic acid give similar particle size distribution profiles (**Figure 1a,b**). These are bimodal with an important population characterized by a peak at 18.21 μm and 22.5% for palmitic acid and at 22.04 μm and 27.5% for stearic acid. The analysis of triolein particle size distribution (**Figure 1c**) shows that the first population overlaps the second one. The particle size distribution of the hydrogenated palm oil shows two separate populations (**Figure 1d**): the first peak at 0.158 μm and 1.7% and the second at 19.95 μm and 5.88%. The volume-surface mean diameter, $D_{3,2}$, of the hydrogenated palm oil is \sim 0.54 μm (**Table 2**) and represents the finest emulsion. The particle size of fatty acids is larger than that of the other fats. Nonetheless, all fats exhibit similar weight mean diameters ($D_{4,3}$) values.

In this work, particle size distributions showed the presence of two populations. Furthermore, $D_{4,3}$ values are $> 10 \mu\text{m}$. These results suggest the existence of aggregated particles. To confirm this hypothesis, optical microscopy and a food grade surfactant (SDS) were used.

Microscopic observations of fatty acid emulsions (**Figure 2a,b**) confirm the presence of both single and flocculated particles. For triolein and the hydrogenated palm oil (**Figure 2c,d**), aggregates were not observed.

Emulsified films were also dissolved in an SDS solution and analyzed by laser light scattering. **Figure 1** illustrates the impact of SDS on lipid particles. In the case of triolein, a shift in particle size distribution (**Figure 1c**) and an increase in $D_{3,2}$ and $D_{4,3}$

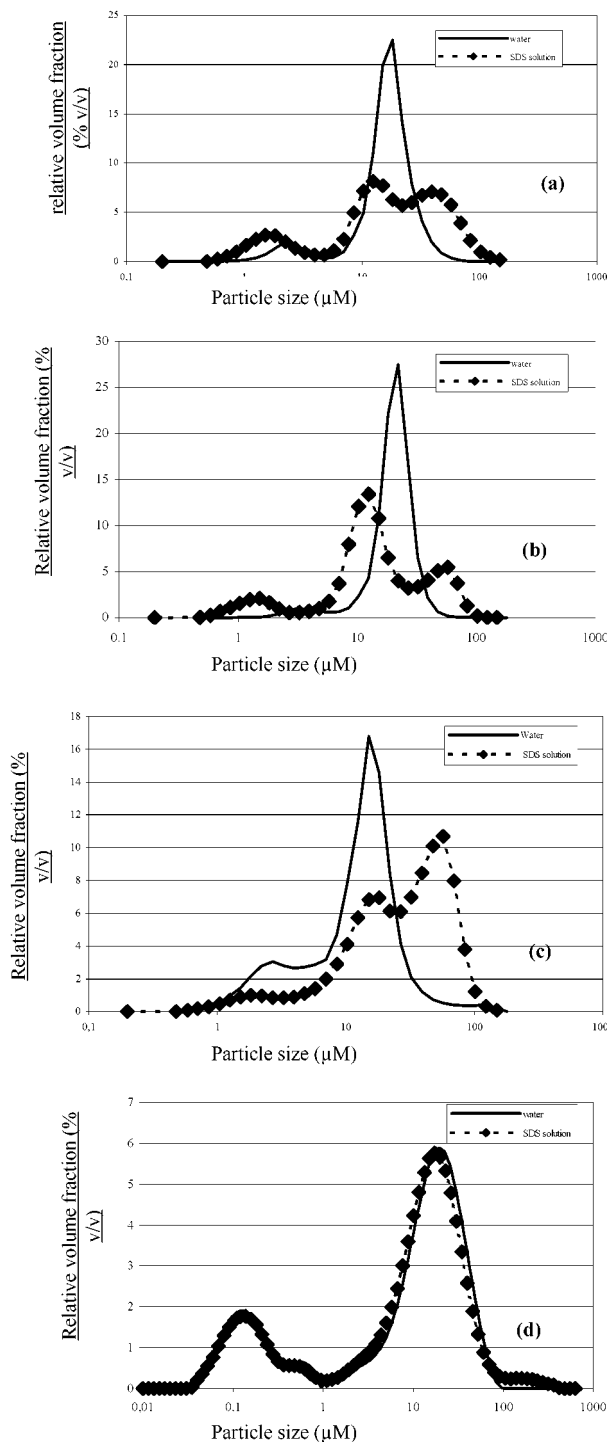


Figure 1. Lipid size distribution profile of arabinoxylan–lipid emulsified films dispersed in water or in a 0.1% SDS solution: (a) palmitic acid; (b) stearic acid; (c) triolein; (d) hydrogenated palm oil.

(**Table 2**) values were observed. These results suggest that SDS could act as a bridge between two droplets and thereby increase their tendency to flocculate. Such behavior has already been observed by several authors (24, 25) with whey protein emulsions. For stearic acid and palmitic acid, a drop in relative volume fraction associated with a decrease of $D_{3,2}$ values occurred, whereas the $D_{4,3}$ values increased. This could be attributed to the destruction of flocs resulting from the competitive SDS adsorption at the oil–water interface and the induced Coulombic repulsion. Moreover, SDS may form complexes with the polymer at the same time, increasing the weight mean

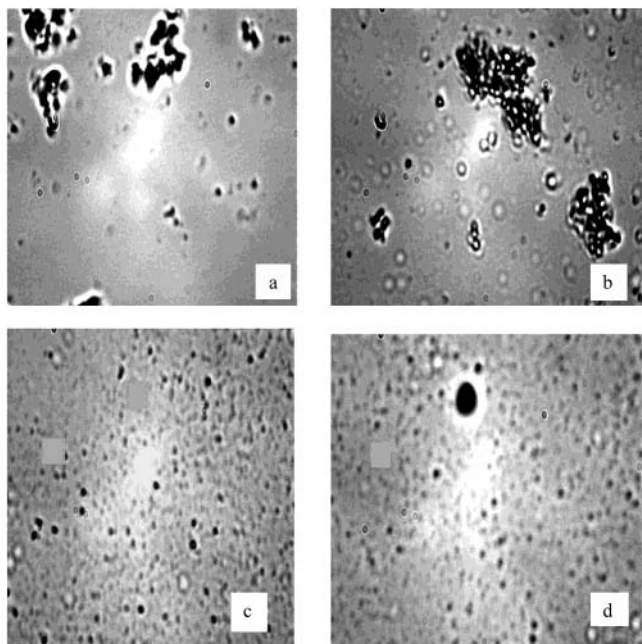


Figure 2. Optical microscopy images (500 \times) of arabinoxylan-based films containing palmitic acid (a), stearic acid (b), triolein (c), and hydrogenated palm oil (d) in emulsion. (Figure is reproduced here at 64% of its original size.)

diameter. The hydrogenated palm oil offers similar particle size distribution and $D_{3,2}$ and $D_{4,3}$ values after film dispersion in water or SDS solution. This assumes that SDS interfacial interactions are weak and coalescence more than flocculation occurred during film preparation.

Optical microscopy and SDS action confirm the presence of aggregates. Several authors have set out to correlate particle size and distribution with WVP (4, 18, 20, 21). Indeed, McHugh and Krochta (20) showed a linear decrease of WVP with a decrease in mean particle diameter for a given volume fraction of lipid. In our study, stearic acid obtained the highest mean diameter, followed by palmitic acid and triolein. The finest emulsion was obtained with the hydrogenated palm oil. Furthermore, all of them show a heterogeneous particle distribution. Nonetheless, these three emulsified films offer the lowest WVP, without significant difference. Thus, direct correlation cannot be established between WVP and particle size and distribution because lipid nature and particle size change at the same time.

Emulsion stability, particle size, and distribution are not the major factors that affect WVP. Other factors such as film orientation, drying conditions, and lipid hydrophobicity must be taken in account.

Surface Hydrophobicity. Contact angles are an indicator of the surface hydrophobicity or the wettability of polymers. In **Table 1**, the contact angles of different food packagings are presented. The contact angles of cellophane film cannot be measured because water droplets are instantaneously absorbed due to the highly hydrophilic nature of this polymer. The surface hydrophobicity of homogeneous AX films, characterized by a contact angle of $\sim 71^\circ$, is comparable to that of methyl cellulose films (27, 31). Gliadin films exhibit higher contact angles of $85\text{--}105^\circ$ (36). This means that gliadin films are less rapidly wetted, which explains the water insolubility of wheat gluten films. Conversely, AX films are water soluble. Additionally, AX films are less hydrophobic than low-density polyethylene films.

Table 3. Mechanical Properties of Arabinoxylan-Based Films and Commercial Plastic Films^a

film	elongation (%)	tensile strength (MPa)	elastic modulus (MPa)
arabinoxylans	7.4 \pm 2.9b	26.5 \pm 4.1a	72.4 \pm 35.2a
AX-C ₁₆	1.7 \pm 0.5c	7.8 \pm 1.2cb	59.2 \pm 19.3b
AX-C ₁₈	2.5 \pm 1.3c	7.1 \pm 1.9c	51.96 \pm 18.7b
AX-triolein	10.8 \pm 3.3a	8.8 \pm 1b	25.84 \pm 11.5c
AX-OK35	8.9 \pm 3.4b	6.4 \pm 1.1c	26.65 \pm 8.1c
cellophane	20 (39)	114 (39)	
LDPE	100–965 (38)	13–28 (38)	

^a Mean of 20 measurements \pm standard deviation. Values in a column followed by the same letter are not significantly different at the risk of 5%.

Surface Properties of Emulsified Films. Addition of fatty acids did not result in important changes in the films' surface hydrophobicity (**Table 1**). Contact angles were significantly lower ($P < 0.05$) for films made with triolein. The polar character of this lipid probably reinforces the water affinity of AX films. A hydrophobic surface showing contact angles superior to 90° is obtained with AX-OK35 films. These values are similar to those regularly observed with LDPE. In contrast with this synthetic film, the angles of AX-OK35 films decrease 10 times more quickly with time (data not shown), suggesting a rapid change in surface properties. In other work (37), it was shown that the highest WVTR and permeabilities correspond to the smallest contact angle values for methyl cellulose films. This is in contrast to the present study, where such a relationship is not shown. In fact, triolein is the most efficient water barrier and offers the lowest contact angles.

As observed for other edible films, AX-based films exhibit lower WVP and hydrophobicity than protein-based films and commercial plastic packaging. Other qualities, such as good mechanical properties, are required in edible coatings. Hence, the percentage of elongation and the tensile strength of AX films developed in this study were measured.

Mechanical properties of AX-based films are given in **Table 3**. These characteristics are similar to those observed for methyl cellulose (31, 38) and starch films (10). Although AX films seem to be less deformable than protein films (26, 39), they have better tensile strength. Cellophane films display a percentage of elongation comparable to that of edible films, but they are stronger. On the contrary, LDPE films have a very high percentage of elongation and a tensile strength similar to those measured for edible coatings.

Influence of Lipid Addition on Mechanical Properties. Elongation of AX films significantly ($P < 0.05$) decreases when fatty acids are incorporated into the AX matrix (**Table 3**). Such a tendency has already been reported by other authors (17, 26) and could be explained by the fact that lipids are unable to form a cohesive and continuous matrix. Contrary to the fatty acids tested in our study, oleic acid has been reported to increase elongation of soy protein, corn zein, and egg white films (40). Incorporation of hydrogenated palm oil does not significantly affect AX film extensibility. However, a significant ($P < 0.05$) increase in film elongation is observed when triolein is added. In a previous study, Shellhammer and Krochta (16) observed an increase in whey protein elongation with an increase in the milkfat fraction. They explained this as a plasticizing effect of unsaturated and lower molecular weight triglycerides present in the milkfat. Triolein, which is liquid at room temperature, has been said to induce lubrication of methyl cellulose chains

(6). All of these results suggested that triolein could modify the AX network.

Fat addition also affects the tensile strength and elastic modulus of the films. Indeed, both tensile strength and Young modulus are significantly ($P < 0.05$) lessened. These negative effects may result from the diminution of the cohesion between AX chains and the weak interactions between the apolar lipid molecules and the polar polymer. In comparison with cellophane and LDPE films, emulsified AX films have a lower percentage of strain and tensile strength at the breaking point. AX-based films and, more generally, food packaging must represent a good compromise between elongation and tensile strength. Food packaging has to withstand the packaging process and application, external stress (transport, handling), and, more specifically for edible coatings, they have to be thin and tender in order to be imperceptible during eating.

Conclusion and Perspectives. Fat addition is the most common means used to improve the water barrier efficiency of hydrocolloid-based films. In our study, lipid has been shown to slightly affect the WVP of AX-based films. In comparison with other hydrocolloid-based films, the WVP of emulsified films is suggested to depend mostly on the nature of the continuous phase and, second, on the lipid type. Then, the relationship between lipid particle size and WVP could not be established because lipid particle size and distribution changed with lipid type. However, OK35-AX emulsion films presented a heterogeneous, fine, and stable particle size distribution. On the subject of films' surface hydrophobicity, contact angles similar to those of LDPE have been observed on OK35-AX emulsion films. Meanwhile, OK35-AX emulsion films had extensibilities similar to those of AX-based films without lipid. This result could be attributed to weaker interactions between this hydrophobic fat and the hydrophilic polymer chains. All of these results suggest that OK35 could enhance the functional properties of AX-based films. For further research on composite AX-based films, this fat has been chosen. To obtain a narrow lipid particle size distribution with no overlapping, the use of sucroesters instead of GMS as an emulsifier has been proposed.

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