Drying Temperature Effect on Water Vapor Permeability and Mechanical Properties of Whey Protein−Lipid Emulsion Films

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The water vapor permeability (WVP) and mechanical properties of whey protein isolate (WPI) and WPI-lipid emulsion films dried at different conditions were investigated. As drying temperature increased, WVPs decreased significantly. Significantly lower WVP was observed for emulsion films compared to WPI films. WPI−Beeswax (BW) and WPI-anhydrous milkfat fraction emulsion films dried at 80 °C and 40% RH gave the lowest WVP compared to 25 °C, 40% RH and 40 °C, 40% RH. A large drop in WVP of WPI-BW emulsion films was observed at 20% BW content. The decrease in WVP for emulsion films as drying temperature increased could be due to change in the lipid crystalline morphology and/or lipid distribution within the matrix. Mechanical properties of WPI and WPI-lipid emulsion films, on the other hand, were not modified by drying conditions.

INTRODUCTION

Edible films and coatings have received great interest in recent years because they can extend the shelf life and improve the quality of the food by providing a barrier to mass transfer, carrying food ingredients and/ or improving the mechanical integrity or handling characteristics of the food. Development has been focused upon barriers containing proteins, polysaccharides and lipids. Polysaccharides and proteins are good film-former materials, but they are poor moisture barriers. Lipids, on the other hand, provide a better moisture barrier, but they form brittle films and require solvents or high temperatures for casting (Krochta, 1997). The properties of edible films and coatings have been more extensively reviewed by Kester and Fennema (1986), Guilbert (1986), Krochta et al. (1994), Krochta and De Mulder-Johnston (1997), Debeaufort et al. (1998), and Baldwin, (1999).

Addition of hydrophobic lipids to hydrophilic protein- or polysacharide-based films, by forming a stable emulsion of the lipid (McHugh and Krochta, 1994; Debeaufort and Voilley, 1995) or by laminating the film with a lipid layer (Martin-Polo et al., 1992; Park et al., 1994), greatly improves water vapor barrier properties. The barrier efficiency of such composite films strongly depends on the polarity of the film components and the uniform or dispersed distribution of the hydrophobic substances in the component films (Kamper and Fennema, 1985; Debeaufort et al., 1993). Some investigations have shown that when the lipid is laminating the hydrophilic film, forming a bilayer, the barrier against water vapor transfer is greater than when a stable emulsion film is formed (Kamper and Fennema, 1984; Kester and Fennema, 1989a). However, lamination requires more steps than forming a composite film in which both hydrophilic

and hydrophobic components are dispersed in the film-forming solution. Furthermore, delamination of these films due to the high surface energy existing between the polar and nonpolar materials can be a problem (Kamper and Fennema, 1984).

Whey proteins produce transparent, bland, flexible water-based edible films with excellent oxygen, aroma, and lipid barrier properties at low relative humidity (McHugh and Krochta, 1994a; Miller and Krochta, 1997; De Mulder, 1999). Incorporation of various lipids into whey protein-based edible films results in films with reduced water vapor permeability (WVP) (McHugh and Krochta, 1994a; Shellhammer and Krochta, 1997; Pérez-Gago and Krochta, 1999a). In these films, the lipid is emulsified by whey protein forming an emulsion film. Factors affecting water vapor permeability of the whey protein−lipid emulsion films are lipid type, amount, and particle size (McHugh and Krochta, 1994a; McHugh and Krochta, 1994b; Shellhammer and Krochta, 1997). McHugh and Krochta (1994b) found that decreasing mean particle diameter of film-forming emulsion correlated well with linear decrease in film WVP. These films also revealed some emulsion separation during drying, since the range of lipid particle sizes was large enough to give unstable emulsions. Shellhammer and Krochta (1997) examined the effect on permeability and mechanical properties of a range of lipid types and amounts. The effect of lipid type on WVP correlated with the viscoelastic properties of the lipids and not with the WVPs of the pure lipid components, suggesting that the more viscoelastic lipid particles may have deformed to produce an internal interconnecting lipid network.

Common practice in film formation, both for hydrophilic and composite films, has mainly involved drying of the films at ambient conditions. The drying behavior of thin films has been most extensively studied for synthetic polymers, whereas edible films have received little attention (Alcantara et al., 1998). The barrier and mechanical properties of methycellulose films, whey protein isolate (WPI) films, and peanut protein concen-

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Table 1. Melting Point and Rheological Properties of Selected Lipids

	AMFF	beeswax	candelilla wax
melting point (°C)	~45[a]	62−64[b]	66−71[b]
hardness[c] (N)	5	25	170

[a] Kaylegian, 1993. [b] Bennett, 1975. [c] Hardness (Newtons) was measured as the resistance of the lipid sample to 3 mm cone penetration (Shellhammer and Krochta, 1997).

trate films have been shown to improve when drying temperatures were increased (Donhowe and Fennema, 1993; Alcantara et al., 1998; Jangchud and Chinnan, 1999). In all the situations, results were attributed to changes in film morphology. Higher drying rates induced crystallinity of methylcellulose, formed denser films with WPI, and induced cross-linking of peanut protein concentrate. A similar study was performed on the effect of drying rate on the barrier and mechanical properties of methylcellulose-lipid emulsion films (Debeaufort and Voilley, 1995). Films were prepared by dispersing the lipid in a methylcellulose solution with the aid of various emulsifiers and then drying at several temperatures, relative humidities and air velocities. Results showed that longer drying times of film-forming emulsion, produced by lower temperatures and higher relative humidities, provided better barrier and mechanical properties. Digitalized Imagining Analysis Microscopy showed that drying rate affected the lipid location within the polymer matrix and thus the macroscopic structure of the film, which could explain the differences found in permeability and mechanical properties.

No research has been published examining the barrier and mechanical properties of whey protein−lipid emulsion films dried at various temperatures. Results obtained at room temperature showed that lipid viscoelasticity had a significant effect on barrier properties of WPI-lipid emulsion films (Shellhammer and Krochta, 1997). Changing the drying temperatures could enhance or reduce lipid deformability and ability to yield an internal interconnecting lipid network. The behavior of hard waxes such as candelilla wax is expected to be different from softer and more viscoelastic lipids such as beeswax and milkfat fraction when dried at different temperatures. Drying at temperatures above or below the melting point of the lipid would also influence lipid deformability and distribution within the polymer matrix. In addition, emulsion stability can be affected by changes in the viscosity of the continuous phase as temperature increases or decreases. The objective of this investigation was to study the effect of drying conditions on water vapor permeability and mechanical properties of whey protein−lipid emulsion films.

MATERIALS AND METHODS

Materials. Beeswax (BW) (Strahl & Pitsch Inc., West Babylon, NY), Candelilla wax (CW) (Strahl & Pitsch Inc., West Babylon, NY), and a high-melting anhydrous milkfat fraction (AMFF) (VH-66, Center for Dairy Research, University of Wisconsin, Madison) were selected as the lipid phase of the WPI emulsion films. Melting points and hardness of BW, CW, and AMFF are shown in Table 1. Lipids were emulsified with 10% whey protein solution made from whey protein isolate (WPI) (Davisco Foods International, Le Sueur, MN). Glycerol (Fisher Scientific Inc., Fair Lawn, NJ) was added to all film-forming solutions as a plasticizer.

Film Preparation. WPI was heat denatured to allow formation of films with greater strength than films from native

WPI (Pérez-Gago and Krochta, 1999b). Aqueous solutions of 10% (w/w) WPI were prepared and heated at 90 °C for 30 min in a water bath to denature the whey protein. The lipid was melted in the hot protein solution, and the plasticizer was added in the amounts required to get the final film compositions. The protein-plasticizer ratio selected was 3 parts WPI to 1 part glycerol (dry basis), and this ratio was kept constant throughout the study. Both 10% WPI films (no lipid present) and emulsion films containing 40% lipid were prepared to study the effect of drying on the physical properties of the films. An additional experiment was conducted to study the effect of lipid content on WPI:BW emulsion films dried at high temperature. Addition of BW to the whey protein-glycerol solution ranged from 0 to 70% (dry basis). Each solution was homogenized with a high-shear probe mixer (Ultra-Turrax, model T25, IKA-Works, Inc., Cincinnati, OH). Homogenization conditions were optimized such that particle sizes of the different emulsions were similar. BW emulsions were emulsified for 1 min at 15 000 rpm followed by 3 min at 22 000 rpm, AMFF emulsions were emulsified for 2 min at 15 000 rpm followed by 3 min at 22 000 rpm, and CW emulsions were emulsified for 3 min at 15 000 rpm. All the emulsions were in a water bath at 80 °C during the homogenization process. After homogenization, the emulsions were placed in an ice bath to prevent further denaturation of the whey protein and to crystallize the lipid particles. The film-forming emulsions were degassed at room temperature with a vacuum pump. The films were prepared by pipetting an appropriate volume of the degassed emulsion to maintain a constant mass of solids (3 g of total solids/plate) on a smooth high-density polyethylene (HDPE) casting plate resting on a leveled granite surface. Films were dried in a temperature- and relative-humidity-controlled environmental chamber (Tenney-10 model TTUFR-40240, Tenney Engineering, Inc., Union, NJ) at three temperatures (25, 40, and 80 °C) and constant relative humidity (40% RH). Drying times at these drying conditions were 15, 4.25, and 2.23 h, respectively. The chamber was equipped with a one-setting fan that circulated interior air at approximately 20 m/min. Drying temperatures were chosen above and below the melting point of the lipids so that lipids stayed in the liquid or crystalline state during drying.

Lipid Particle Size Analysis. McHugh and Krochta (1994b) confirmed that lipid particle sizes in films measured by scanning electron microscopy were similar to those in emulsions. Thus, laser light scattering was utilized to examine the particle size distribution of the emulsions prior to film formation, and results were assumed to be similar for the dried emulsion films. Lipid particle size distribution of the film-forming emulsions was determined using a Malvern Master-sizer (model MS 20, Malvern Instruments Ltd., Malvern, England). Three measurements of each emulsion were made. The relative refractive index of the lipid phase to water was set at 1.08, calculated from the refractive indices of the lipid and water phases, and the absorption was set at 1.0. A 45 mm focal length and a 2.2 mm beam length were used. The obscuration in all the measurements was 0.15 ± 0.02. The mean particle size was recorded as the D(3,2) diameter.

Water Vapor Permeability Measurements. A modification of the ASTM E96−95 gravimetric method for measuring WVP was employed (McHugh et al., 1993). Three films were cast from each solution, which corresponded to three different replicates. From each film, two samples with lack of defects were cut and mounted on Plexiglas test cups containing 6 mL of water, with the film surface which had been exposed to air during drying facing either the low relative humidity (RH) environment ("facing up") or the high RH environment ("facing down"), allowing detection of any phase separation within the film. The cups were placed in desiccator cabinets containing fans and held at 0% RH using anhydrous calcium sulfate (W. A. Hammond Drierite Co., Xenia, OH). Weights taken periodically after steady state was achieved were used to calculate the % RH at the film underside and the resulting WVP.

Film Thickness Measurements. Film thickness was measured after the WVP test using a caliper micrometer (no. 7326, Mitutoyo Mfg. Co. Ltd., Japan), taking measurements

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at six random positions of the film. Averaged values of the six thickness measurements per film were used in all the WVP calculations.

Tensile Properties Determination. Film solutions were cast onto rectangular 23 cm × 30 cm, rimmed, smooth HDPE plates, by applying 3 g of total solids/plate to minimize thickness variations between treatments. After drying at controlled conditions, the films were conditioned at 58% RH in a chamber that contained saturated sodium bromide (Fisher Scientifics Inc., Fair Lawn, NJ) solution for more than 2 days. This preconditioning enabled ease of handling and cutting of the films. Test pieces of film were cut using a striking die (The Right Image, Sacramento, CA). The film samples presented a rectangular center section measuring 15 mm wide by 100 mm long, flaring to 25 mm by 35 mm square grips on each end, which provided a greater grip area. Before testing, all film strips were equilibrated for 36 h to 53% RH in a cabinet using magnesium nitrate (Fisher Scientifics Inc., Fair Lawn, NJ) saturated solution at room temperature (23 ± 2 °C).

Tensile measurements were performed following the procedure outlined in ASTM method D882-97. The ends of the equilibrated film strips were mounted and clamped with pneumatic grips on a Universal Testing Machine (Model 1122, Instron Corp., Canton, MA) with a 500 kg load cell. The initial gauge length was set to 115 mm, and films were stretched using a crosshead speed of 50 mm/min. Testing conditions were controlled throughout the measurements and held constant at 50 ± 5% RH and 23 ± 2 °C. Tensile properties were calculated from the curve of stress (tensile force/initial cross-sectional area) versus strain (extension as a fraction of the original length), using Series IX Automated Materials Testing System Software (Instron). Mechanical properties reported are maximum tensile stress (MPa), elongation at break (%), and elastic modulus (MPa).

Statistical Analysis. The effects of drying conditions, lipid type, and amount on the final film WVP and mechanical properties were determined by using a split-plot design with drying conditions being the blocked variable. Statistical analysis was performed using SAS software (SAS Institute Inc., 1989). Duncan's multiple comparison test was used to determine significance of differences between means.

RESULTS AND DISCUSSION

Water Vapor Permeability of the Emulsion Films. Drying conditions affected the final appearance of the films. In general, films had homogeneous lipid distribution when dried at 25 °C and 40% RH. However, when dried at 40 and 80 °C, migration of the lipid toward the edges was observed, with greater migration at 80 °C. This migration could be attributed to the fact that as the film started drying from the edges of the casting plates to the center, the lipid phase tends to migrate toward the more hydrophobic (lower moisture) area. Differences on film appearance were also found depending on lipid type. CW emulsion films were more brittle than BW and AMFF emulsion films, and when dried at higher temperature, the films tended to shrink. However, samples with uniform lipid distribution and lack of defects were obtained to measure WVP. AMFF gave emulsion films that were greasy on the surface. All these differences could be attributed to the diverse chemical composition of these lipids, with resulting different melting points and rheological properties (Table 1).

Lipid type, drying temperature, and film orientation had statistically significant effect on the WVP of the films. Figure 1 shows the effects that lipid type, phase separation, and drying conditions have on WVP of the WPI-lipid emulsion films. Lipid separation within the film was determined by measuring WVP as a function of film orientation on the test cups. As expected, films formed from 10% WPI with no lipid present showed no
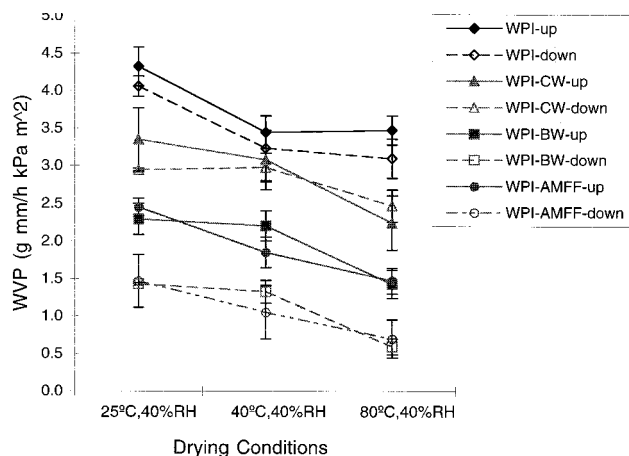

Figure 1. Drying conditions effect on water vapor permeability of WPI:Gly:Lipid (45:15:40) emulsion films.

Table 2. Average Lipid Particle Sizes in Film-Forming Emulsions

emulsion	mean particle diameter (μm)
candelilla wax	1.34 ± 0.02
beeswax	1.57 ± 0.06
AMFF	1.83 ± 0.14

difference in WVP when film orientation on the test cups was changed. However, phase separation occurred in all the emulsion films except for CW emulsion films. These results can be attributed to lipid particle size. Emulsions prepared with BW and AMFF had a larger particle size than emulsions prepared with CW (Table 2). Phase separation occurred in BW and AMFF films since the particle size was larger, producing less stable emulsions. Preliminary experiments were performed to establish homogenization conditions to obtain emulsions with similar lipid particle sizes. Increasing the degree of high-shear mixing homogenization, by increasing homogenization time and/or speed, did not further decrease particle size. CW emulsions gave films with the lowest particle size using lower homogenization conditions compared to BW and AMFF emulsions. The different chemical composition of these lipids could be responsible for the difference in particle sizes. Differences in particle size distribution as a function of fatty acid chain length have also been reported. Particle size increased with increased chain length of fatty acid in whey protein-fatty acid emulsion films (Craig et al., 1998). The fact that higher temperature did not produce phase separation in CW emulsion films leads to the conclusion that the drying conditions used did not affect emulsion stability. However, it would be of great interest to further study the effect of lipid particle size on WVP as the emulsion films are dried at different conditions.

It is clear from results that CW emulsion films gave the highest WVPs among the emulsion films studied, while BW and AMFF emulsion films gave similar WVPs at all drying conditions (Figure 1). These results are consistent with those found by Shellhammer and Krochta (1997), where CW formed emulsion films with the highest WVP at all the lipid contents.

Analysis of variance confirmed that drying conditions had a significant effect on WVP ($p < 0.01$). Results showed that increasing the film drying temperature formed films with decreased WVP, for both 10% WPI and WPI-lipid emulsion films. Lower permeability in polymer films can be achieved by inducing a more

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compact polymer structure that retards permeant diffusion through the films (Miller and Krochta, 1997). In synthetic polymer films, it is well-known that an increase in temperature increases the cohesive strength between polymer chains, due to the increased thermal motion of the polymer chains leading to more extensive chain–chain interactions (Banker, 1966). From our results, it could be hypothesized that an increase in drying temperature of WPI films allowed for better polymer arrangement and cohesion within the film matrix that resulted in a tighter and more compact structure. Furthermore, hydrophobic interactions are increased at high temperatures (Kinsella and Phillips, 1989), which could also result in greater interactions between polymer chains.

Interestingly, the WVP was not different for WPI films dried at 40 and 80 °C. However, for all emulsion films, WVPs decreased as drying temperature increased to 80 °C. As 80 °C is above the lipid-melting points, this could be due to a change in the lipid crystalline morphology and/or lipid distribution within the matrix. Upon cooling, lipids can crystallize into one of the three principle polymorphic forms: α, β, or β'. The final crystalline form depends on the cooling rate and melting history. Kester and Fennema (1989b) found that the moisture permeability decreased following transition from the least stable α polymorph to the more dense hydrocarbon packing β' polymorph in a blend of fully hydrogenated soybean and rapeseed oil. Drying of our films at temperatures above the lipid-melting point could change the polymorphic form of the subsequently formed lipid crystals, increasing molecular density and modifying the barrier properties of the emulsion films. In addition, the distribution of the lipid phase within the matrix affects the barrier properties of the films (Kamper and Fennema, 1985; Debeaufort et al., 1993). Debeaufort and Voilley (1995) observed, through the use of Digitalized Imaging Microscopy, that drying rate strongly affected the paraffin location within a methylcellulose matrix. Destabilization of the emulsion was attributed to a decrease in viscosity of the continuous phase as temperature increased. Moreover, at temperatures above the melting point, the lipid is in the liquid state, and lipid particles might tend to deform, merge, and aggregate into networks as water evaporates.

To better understand the role of lipid content in emulsion films when dried at high temperatures, WPI-BW emulsion films at different BW content were prepared and dried at 80 °C and 40% RH. The WVP of these films is shown in Figure 2. Phase separation was observed at all BW contents, due to large particle size of the BW droplets. An apparent feature is the dramatic drop in the WVP of the WPI-BW emulsion films at 10 and 20% BW level when film orientation on the test cups was "down" and "up", respectively. Compared to the results of Shellhammer and Krochta (1997) which showed a large drop in WVP at 40% BW, the drop in WVP has been shifted to lower BW contents. This could be an indication of a change in BW distribution in the emulsion film, creating regions of higher BW content and as a consequence lower film permeability. Another interesting feature is the low WVP obtained for the films at concentrations above 20% BW. Values are comparable to the ones obtained by Shellhammer and Krochta (1997), even though plasticizer levels are 5 times larger than the ones used by these authors. This suggests that,
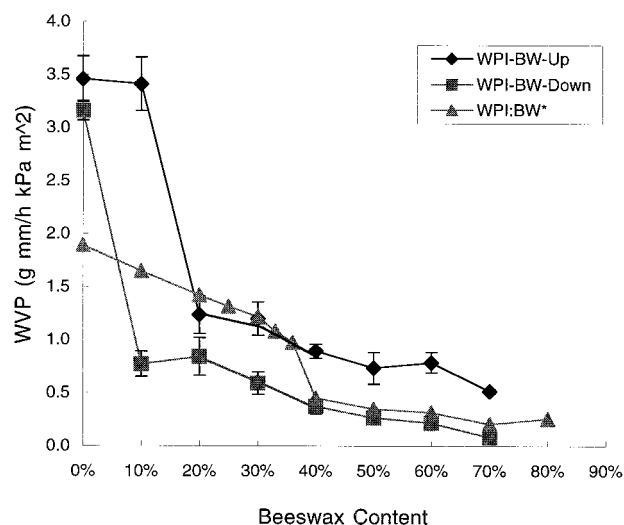

Figure 2. Lipid content effect on water vapor permeability of WPI:Gly:BW emulsion films dried at 80 °C and 40% RH. (WPI:BW*) Shellhammer and Krochta (1997).
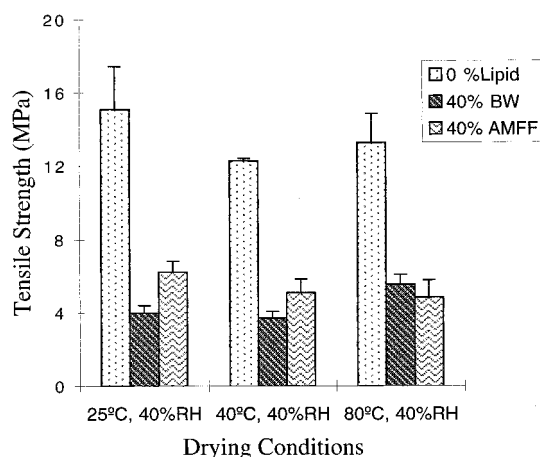

Figure 3. Tensile strength of WPI:Gly (75:25) and WPI:Gly:Lipid (45:15:40) emulsion films as affected by drying conditions.

in emulsion films containing high lipid levels, plasticizer content does not play an important role in affecting WVP.

Mechanical Properties of Emulsion Films. WPI–CW emulsion films were very brittle and curled after drying, giving no possibility of obtaining samples for mechanical testing. However, we were able to study the effect of drying conditions on mechanical properties for WPI-AMFF and WPI-BW emulsion films. Drying conditions had no effect on the tensile strengths, stretchabilities and elastic moduli of these films (Figures 3–5). Alcantara et al. (1998) also found no significant differences in tensile strength and stretchability for 10% WPI films dried at different temperatures. Elongation, however, increased for films dried at higher temperatures, although the differences were small.

For peanut protein films, drying conditions affected the mechanical properties of the films (Jangchud and Chinnan, 1999). Tensile strength and elongation increased when the film-forming temperature increased from 70 to 90 °C. These results were attributed to protein denaturation at the higher temperature, which led to tighter, more compact protein networks and structures. In their study, films were formed after drying at the desired temperature for 16 h. The longer
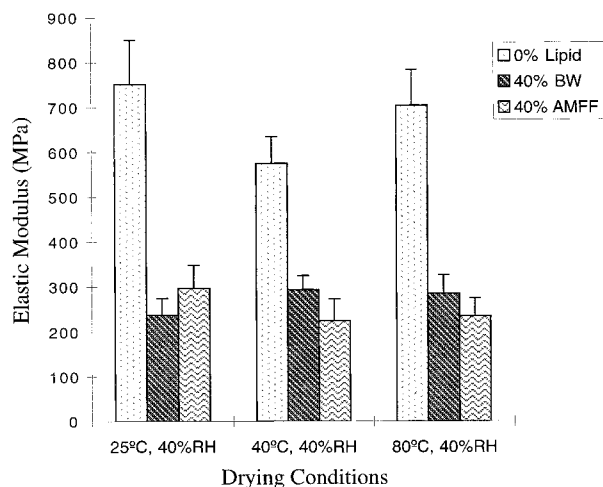
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Figure 4. Elastic modulus of WPI:Gly (75:25) and WPI:Gly: Lipid (45:15:40) emulsion films as affected by drying conditions.
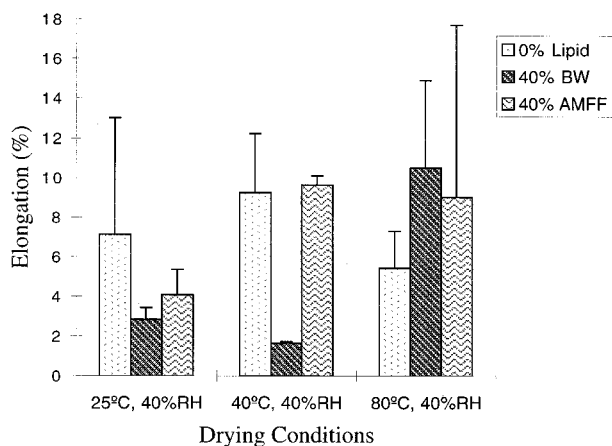

Figure 5. Elongation of WPI:Gly (75:25) and WPI:Gly:Lipid (45:15:40) emulsion films as affected by drying conditions.

drying times could have acted as a heat-curing time, inducing additional cross-linking of protein. Miller et al. (1997) observed that heat curing of whey protein films increased tensile strength and decreased elongation and Young's Modulus of the final film.

In our study, significant differences ($p > 0.001$) were observed on the tensile strength and Young's Modulus only between 10% WPI films and the emulsion films. This confirms the fact that the protein phase has much more tensile strength than the lipid phase. When comparing emulsion films, no differences were observed between WPI-BW and WPI-AMFF emulsion films. These results are consistent with the ones obtained by Shellhammer and Krochta (1997).

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