Brazilian Journal
of Chemical
Engineering

ISSN 0104-6632
Printed in Brazil

Vol. 21, No. 02 pp. 261 - 264, April - June 2004

CHARACTERIZATION OF COMPOSITE BIOFILMS OF WHEAT GLUTEN AND CELLULOSE ACETATE PHTHALATE

F. M. Fakhouri, P. S. Tanada-Palmu and C. R. F. Grosso

Department of Food and Nutrition, Faculty of Food Engineering,
State University of Campinas, UNICAMP, Phone +(55) (19) 3788-4079,
Fax +(55) (19) 3788-4060, 13083-970, Campinas - SP, Brazil.
E-mail:grosso@fea.unicamp.br

(Received: February 13, 2003 ; Accepted: November10, 2003)

Abstract - The objective of this research was to develop and characterize composite biofilms produced using wheat gluten and cellulose acetate phthalate. Biofilms act as barriers to moisture and oxygen diffusion through the film. The films were prepared with different thicknesses and component concentrations and were analyzed for water vapor and oxygen permeabilities, water and acid solubilities and mechanical properties. Results showed that the mixture improved film characteristics more than each of the individual components alone. The 1:1 mixture had properties of better permeability to water and oxygen. The composite films were completely soluble in water and acid, with the exception of the film with the highest gluten concentration, which was 50% soluble in water and acid. An increase in gluten concentration in the composite films resulted in a decrease in tensile strength. There was no significant difference in elongation at break between the composite films. No difference in thickness was detected either. Results showed that the mixture improved the characteristics more than of the individual components alone.

Key words: biofilm properties, gluten, cellulose acetate phthalate.

INTRODUCTION

Edible films are viewed as an alternative for increasing the shelf life of fruits and vegetables, protecting them from the effects of humidity and oxygen, and thus delaying their deterioration. Use of these films is not recent. In China, in the eleventh and thirteenth centuries, wax coating was used on oranges and lemons to avoid moisture loss (Hardenburg, 1997). Edible films of carnauba wax have been commercialized since 1930 for the covering of fresh fruits and vegetables (Kaplan, 1986). Edible films are used to control oxygen permeability and the migration of water vapor, carbon dioxide and lipids in a food. Moreover, they can introduce additives such as antioxidants, antimicrobials and flavors into the food, thus improving the mechanical integrity, facilitating handling and preserving the quality of the food (Krochta and Johnson, 1997). Biofilms are produced from proteins, hydrocolloids, lipids or a combination of these. Composite films offer the advantage of combining the positive aspects of all components used. The objective of this work was to compare films of wheat gluten, of cellulose acetate phthalate and of a mixture of the two on water vapor and oxygen permeability, mechanical properties, and solubility in water and acid.

MATERIALS AND METHODS

Materials

Vital wheat gluten (Rhodia, Campinas, Brazil) and cellulose acetate phthalate (Eastman Kodak, USA) were used. The reactants were glycerol (Merck, Darmstadt, Germany), ammonium hydroxide (Synth, São Paulo, Brazil), calcium chloride (Ecibra, São Paulo, Brazil), sodium chloride (Synth, São Paulo, Brazil), magnesium nitrate (Ecibra, São Paulo, Brazil), monobasic sodium phosphate (Synth, São Paulo), tribasic sodium phosphate (Synth, São Paulo) and solid paraffin (Chemco, São Paulo, Brazil).

Preparation of Gluten Film

The film was prepared from a solution of gluten (9.0 g/100 mL solution), absolute ethanol (32.5 mL/100 mL solution), glycerol (1.50 g/100 mL solution), distilled water and ammonium hydroxide to adjust to pH 10. All components were mixed under magnetic stirring until the temperature of the mixture reached 70 C and the solution was centrifuged at 5856 g for 6 min at room temperature. The film-forming solution was poured and spread evenly over a Teflon-covered glass surface and dried at room temperature for 24 h (modified method from Gontard et al., 1993). All films used for experiments were equilibrated at 52% RH at 25 C for 48 h before being tested.

Preparation of Cellulose Acetate Phthalate (CAP) Film

The CAP film was prepared by solubilizing 10 g of CAP in a buffer solution of sodium phosphate at pH 8.0 in a mechanical shaker and then adding 0.5 g of glycerol. Aliquots of 8 mL and 16 mL were spread evenly over 15 cm Plexiglas plates and left to dry at room temperature for 24h.

Preparation of Composite Films

The composite films were prepared by mixing the CAP film-forming solution with the wheat gluten film-forming solution at ratios of 1:1, 1:4 and 4:1 under magnetic stirring, then the mixtures were spread evenly over Plexiglas plates in aliquots of 8 mL, 12 mL and 16 mL.

Water Vapor Permeability

The water vapor transmission rate of the films was determined gravimetrically at 25 C using the modified American Society for Testing and Materials (ASTM) Standard Method E-96 according to Tanada-Palmu et al. (2000). The samples, triplicates of each film, were conditioned for 2 d at 52% RH before measurement. Water vapor permeability was calculated according to the ASTM method.

Oxygen Permeability

The oxygen transmission rates were determined using a modification of ASTM Standard Method D 3985-81 with an Ox-Tran apparatus (Mocon, Inc., Minneapolis, USA) at 25 C. The samples, duplicates of each film, were conditioned for 2 d at 52% RH before measurement. Oxygen permeability (OP) was calculated by dividing ratio of oxygen transfer by the oxygen pressure and multiplying by mean thickness.

Tensile Strength and Percentage of Elongation at Break

Tensile strength of the film and percentage of elongation at break were determined using a TA.XT2 Texture Analyzer (Stable Micro System, Surrey, UK), operated according to ASTM Standard Method D 882-83 (25 C, initial grip separation = 50 mm and cross head speed = 100 mm/min). The peak loads and extension at break were recorded for film specimens tested (100 mm long and 25.4 mm wide). The films were conditioned for 2 d at 52% RH before measurement. Six specimens of each film were measured. Tensile strength and percentage of elongation at break were calculated according to the ASTM method.

Solubility in Water

The percent of solubility was the percentage of dry matter in the film solubilized after 24 h immersion in water. The percentage of initial dry matter in each film was determined at 105 C for 24 h. Two discs of film (2 cm diameter) were cut, weighed, immersed in 50 mL of distilled water and slowly and periodically shaken for 24 h at 25 C. The pieces of film were then taken out and dried (105 C for 24 h) to determine the weight of dry matter which was not solubilized in water. The weight of solubilized dry matter was calculated by subtracting the weight of dry matter not solubilized from the weight of the initial dry matter and was reported on an initial dry weight basis.

Solubility in Acid

The same procedure as for solubility in water was used, except that the films were immersed in HCl 1 N solution for 24 h at 25 C.

Statistical Analyses

The Statistica 5.5 (Statsoft, USA) program was used to determine the significant differences in all film properties at a confidence level of 95%.

RESULTS AND DISCUSSION

The composite films of wheat gluten and CAP were opaque due to cellulose and fragile, specially the thinner films. Due to their brittleness, it was not possible to characterize the composite films with the highest concentration of gluten. The composite films with 1:1 and 1:4 ratios of gluten to cellulose of all thicknesses had much better water vapor barriers (Table 1) than films of hydroxipropilmetilcellulose (HPMC) and metilcellulose (MC) with values of 1.92 and 1.48 gmm/m^{2}dkPa, respectively (Kamper and Fennema, 1984; Greener and Fennema, 1989). In the composite film with the highest concentration of gluten, water vapor permeability increased. The composite films as well as the cellulose films were completely soluble in water. The composite film with the highest concentration of gluten had 50% solubility in acid, and the composite films with the other concentrations of gluten and cellulose were completely soluble in acid. Tensile strength (Table 2) decreased with the increase in gluten concentration in the composite films. Addition of gluten up to a concentration of 25% increased tensile strength in the composite films; however at higher concentrations, strength decreased. For elongation at break, there was not a significant difference in the composite films; the film with the highest concentration of cellulose showed the highest elongation, which was still low in comparison with the gluten film. For thickness of the films, there was no significant difference in any property measured. Oxygen permeability could only be measured in the 1:1 (16 mL) cellulose to gluten composite film because the other films were very fragile and brittle. This permeability was low compared to that of the gluten film and the values from Park and Chinnan (1990) for zein:glycerol films (13.0-44.9 cm^{3}mm/m^{2}dkPa) and gluten:glycerol films (9.6-24.2 cm^{3}mm/m^{2}dkPa) at 30 C and 0% relative humidity.

Table 1: Water vapor permeability and solubility in water and acid of the films

Film	Water vapor permeability (gmm/m^{2}dkPa)*	Solubility in water (%)*	Solubility in acid (%)*
CAP:glu (1:4)/8ml	-	-	-
CAP:glu (1:4)/12ml	-	-	-
CAP:glu (1:4)/16ml	12.19 ± 0.25^a	100.00^a	50.69 ± 1.64^b
CAP:glu (1:1)/8ml	3.75 ± 0.17^b	100.00^a	100.00^a
CAP:glu (1:1)/12ml	3.83 ± 0.13^b	100.00^a	100.00^a
CAP:glu (1:1)/16ml	4.11 ± 0.20^b	100.00^a	100.00^a
CAP:glu (4:1)/8ml	3.81 ± 0.01^b	100.00^a	100.00^a
CAP:glu (4:1)/12ml	3.64 ± 0.0^b	100.00^a	100.00^a
CAP:glu (4:1)/16ml	3.76 ± 0.26^b	100.00^a	100.00^a
CAP/8ml	6.03 ± 0.35^{ab}	100.00^a	30.50 ± 1.56^c
CAP/16 ml	10.05 ± 0.47^a	100.00^a	33.14 ± 1.87^c
Gluten	8.61 ± 1.03^{ab}	22.70 ± 4.10^b	-

*Mean and standard deviation of replicates. $^{a\text{-}c}$ Means with different superscript letters in the same column are significantly different ($p<0.05$) according to the ANOVA and Tukey tests.

Table 2: Mechanical properties and oxygen permeability of the films

Film	Tensile strength (MPa)*	Elongation at break (%)*	Oxygen permeability ($cm^3 \mu m/m^2 dkPa$)*
CAP:glu (1:4)/8ml	-	-	-
CAP:glu (1:4)/12ml	-	-	-
CAP:glu (1:4)/16ml	1.32 ± 0.11^{e}	4.06 ± 0.78^{e}	-
CAP:glu (1:1)/8ml	7.57 ± 0.18^{de}	4.65 ± 1.63^{de}	-
CAP:glu (1:1)/12ml	10.89 ± 2.19^{cd}	5.27 ± 0.85^{cde}	-
CAP:glu (1:1)/16ml	16.18 ± 3.11^{bc}	4.91 ± 1.01^{de}	22.21 ± 1.23^{b}
CAP:glu (4:1)/8ml	12.87 ± 2.41^{cd}	4.79 ± 1.96^{de}	-
CAP:glu (4:1)/12ml	22.60 ± 1.63^{ab}	6.79 ± 0.98^{bd}	-
CAP:glu (4:1)/16ml	23.78 ± 5.12^{a}	8.82 ± 0.68^{b}	-
CAP/8ml	11.08 ± 3.29^{cd}	7.86 ± 1.23^{bc}	-
CAP/16 ml	11.76 ± 4.31^{cd}	8.60 ± 1.36^{b}	-
Gluten	5.25 ± 0.24^{de}	215.30 ± 12.20^{a}	41.02 ± 0.86^{a}

*Mean and standard deviation of replicates. $^{a\text{-}e}$ Means with different superscript letters in the same column are significantly different ($p<0.05$) according to the ANOVA and Tukey tests.

CONCLUSION

Composite films of wheat gluten and cellulose acetate phthalate had lower water vapor permeability than films of either gluten or cellulose, showing an excellent water vapor barrier. With an increase in gluten concentration in the composite films, the solubility in acid and water and the mechanical resistance of the films decreased.

REFERENCES

ASTM; Methods D 3985-81, D 882-83 and E 96. In Annual Book of Standards. American Society for Testing Materials. Philadelphia, PA (1980).

Gontard, N.; Guilbert, S. and Cuq, J.L.; Water and Glycerol as Plasticizer Affect Mechanical and Water Vapor Barrier Properties of an Edible Wheat Gluten Film. Journal of Food Science, 58, N$^{0.}$1, 206-211 (1993).

Greener, I.K. and Fennema, O.; Barrier Properties and Surface Characteristics of Edible Bilayer Films. Journal of Food Science, 54, N$^{0.}$6, 1393-1399 (1989).

Hardenburg R.E.; Wax and Related Coatings for Horticultural Products. A Bibliography. Agriculture Research Service Bulletin 51, 15, United States Department of Agriculture, Washington, DC (1997).

Kamper, S.L. and Fennema, O.; Water Vapor Permeability of Edible Bilayer Films. Journal of Food Science, 49, 1478-1481, 1485 (1984).

Kaplan H.J.; Washing, Waxing and Color-adding in Fresh Citrus Fruit*,* W.F. Wardowski*,* S. Nagy and W. Grierson, eds., p.379. A VI, Westport, CT (1986).

Krochta, J.M. and Johnson, C.M.; Edible and Biodegradable Polymer Films: Challenges and Opportunities. Food Technology, 51, N$^{0.}$2, 60-74 (1997).

Park, H.J. and Chinnan, M.S.; Properties of Edible Coatings for Fruits and Vegetables, ASAE Paper N$^{0.}$90-6510, presented at the 1990 International Winter Meeting, American Society of Agricultural Engineers, December 18-21, Chicago, IL (1990).

Tanada-Palmu, P.S.; Helén, H. and Hyvönen, L.; Preparation, Properties and Applications of Wheat Gluten Edible Films**.** Agricultural and Food Science in Finland, 9, 23-35 (2000).