

Characterisation of composite films made of konjac glucomannan (KGM), carboxymethyl cellulose (CMC) and lipid

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Received 7 June 2007; received in revised form 19 July 2007; accepted 16 August 2007

Abstract

Emulsion films prepared from konjac glucomannan (KGM), carboxymethylcellulose (CMC), with and without alkali (KOH), were characterised for selected properties such as morphological characteristics, water vapour permeability (WVP), water sorptive capacity and tensile properties. It was found that smaller lipid globules, with a homogeneous distribution, increased the apparent hydrophobicity and “tortuosity” of an emulsion film for water molecule transmission, thereby resulting in reduced WVP. This was achieved when alkaline deacetylated KGM and CMC were served as the base polymers for the emulsion, with the latter functioning as an emulsifier. Interactions between deacetylated KGM and CMC resulted in a stable emulsion.

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Keywords: Konjac glucomannan; Carboxymethylcellulose; Edible films; Water vapour permeability; Sorption isotherm; Tensile properties

1. Introduction

Most polysaccharide and protein films have poor moisture barrier properties. This is due to the presence of free hydroxyl groups in the matrix, which interact strongly with migrating water molecules. Poor moisture barrier properties can be improved by the addition of hydrophobic materials, either by laminating a hydrophilic film with a lipid layer or by forming a composite film in which both hydrophilic and hydrophobic components are dispersed in a co-solvent and then dried. The pros and cons of the bilayer and emulsion techniques have been discussed in previous works (Debeaufort, Martin-Polo, & Voilley, 1993; Debeaufort & Voilley, 1995; Donhowe & Fennema, 1994; Greener & Fennema, 1989; Guilbert, 2000; Kamper & Fennema, 1984a; Kamper & Fennema, 1984b; Martin-Polo, Mauguin, & Voilley, 1992; McHugh, 2000; Park, Testin, Park,

Vergano, & Weller, 1994; Park, Testin, Vergano, Park, & Weller, 1996).

Although it has been proven that bilayer films are good barriers to water vapour transmission, an emulsion composite film appears to be more favourable in terms of simple and feasible preparation procedures. Emulsion composite films may also possess good structural cohesion, imparted by the long-chain polymer and hydrorepellancy imparted by the lipid. Thus, research has advanced towards the development of emulsion-based edible films of low water vapour permeability (Debeaufort & Voilley, 1995; Garcia, Martino, & Zaritzky, 2000; Gontard, Ducheze, Cuq, & Guilbert, 1994; McHugh & Krochta, 1994; Wong, Gastineau, Gregorski, Tillin, & Pavlath, 1992; Yang & Paulson, 2000).

Ukai, Ishibashi, Tsutsumi, and Marakami (1976) and Krochta (1990) have described the mass transfer through an emulsion film. The former researchers proposed the “microvoid model” and suggested that mass transfer of gases and vapours occurs through microvoids, which are formed between the microparticles of the hydrophobic

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material and the hydrocolloid matrix. The latter researcher suggested the “micropathway model”, with mass transfer occurring through the high polymer matrix itself. The barrier properties of emulsion films were studied and reported to be dependent on preparation techniques, component compatibility, type and quantity of lipids used and microstructural, heterogeneity and surface energy of the film formed (Debeaufort & Voilley, 1993, 1995; Hagenmeier & Shaw, 1990; Kester & Fennema, 1989; Martin-Polo et al., 1992; Wong et al., 1992).

Debeaufort and Voilley (1995) reported that the effectiveness of emulsion films in reducing WVP strongly depended on the behaviour and stability of the emulsion during drying. It was suggested that a slow drying rate, small globule diameter and the presence of emulsifier, gave a better water vapour barrier and mechanical efficiencies. Little research has been devoted to study the effect of base polymer orientation, in influencing the stability and efficiency of an emulsion film.

This work is a continuation from a previous study reported by Cheng, Abd Karim, Norziah, and Seow (2002). The reported konjac glucomannan (KGM) and carboxymethylcellulose (CMC) films were found to show different microstructural properties, due to the interactive effect between KGM and CMC, in the presence or absence of alkali. It would be interesting to find out whether or not such a difference in microstructural properties has an influence on the properties of the emulsion films.

2. Materials and methods

2.1. Materials

Purified KGM (Propol A) was generously provided by Shimizu Chemical Corporation, Japan. Analytical grade low-viscosity carboxymethylcellulose (CMC) sodium salt (Sigma), palm olein (PO) lipid (Golden Agricultural Oils Sdn. Bhd., Malaysia) and potassium hydroxide (KOH) pellets (Merk) were used in this study.

2.2. KGM-based emulsion films preparation

A completely randomised design, with 2×2 factorial set of CMC (with and without) alkali (with and without) addition, was adopted.

Four types of emulsion films were prepared: KGM–PO, KGM–KOH–PO, KGM–CMC–PO and KGM–CMC–KOH–PO. Film compositions and film preparation procedures for these films were almost similar to those reported by Cheng et al. (2002), except that palm olein (PO) was added into each formulation. Prior to the corresponding film preparation procedures for each film type as described previously, an amount (2.1 g) of PO was homogenised in 100 ml of distilled water, at 20,500 rpm for 5 min, using an Ultra Turrax (IKA® – LaBortechnik) homogeniser. The PO emulsion was then quantitatively transferred into a National® home blender (Model MX-595N) and the final

volume of each formulation was controlled to approximately 1 l with distilled water.

To produce films with KGM only, KGM (7 g on dry basis) was added before blending for 15 min. The mixture was then left to stand for 1 h at 30 °C, after which 90 g portions were poured and spread onto a level, square perspex plate fitted with rims around the edge to give a 16×16 cm film-forming area. The solutions were allowed to dry at room temperature for approximately 20 h. Films that formed were peeled off and kept in zippered HDPE plastic bags.

To produce blended KGM–CMC films, a clear CMC solution was first prepared by dispersing 2.1 g of CMC (dry basis) into 100 ml of warm (~ 50 °C), vigorously stirred, distilled water in a beaker. The weight of CMC which was added was arbitrarily set at 30% of the weight of KGM used. The CMC solution was then quantitatively transferred into the blender, using distilled water to rinse the beaker. KGM was then blended in and the films were cast as described earlier.

Where required, 1.4 ml of 0.5 M KOH was added prior to blending, to give solutions an initial pH of about 10. For those treatments where alkali was excluded, the pH remained constant at about 6.5, before and after standing. On the other hand, pH of alkalisated solutions dropped from an initial value of ~ 10 –7 after standing for 1 h.

2.3. Films characterisation

Films prepared were characterised by scanning electron microscopy (SEM), sorption isotherm determination, water vapour permeability and tensile testing.

2.3.1. Scanning electron microscopy (SEM)

Films were “conditioned” at 30 °C and 69% RH (over a saturated strontium chloride solution) for 7 days prior to analysis. Each film was mounted on a specimen stub and coated with 100–200 Å thickness of gold, in a Polaron SC515 SEM coating system. The specimen was scanned using a Leica Cambridge S-360 electron microscope. Images were recorded on Kodak Plus-X pan films. Samples were photographed at tilt angles of 30–60° to the electron beam, for views in the cross-section.

2.3.2. Determination of water vapour permeability (WVP)

WVP tests were conducted following the ASTM (1981a) Method E96-80, with some modifications. The test film was sealed as a patch onto a glass permeation cell containing silica gel (RVP = 0), with a 1.5-cm-deep headspace. The cell was then placed in a desiccator, maintained at 30 °C and RVP = 0.22, using a saturated salt solution of potassium acetate. The dull side of the cast film was oriented towards the higher RH compartment and water vapour transport was determined from the weight gain of the cell. The cell was weighed daily over a 6-day period. Changes in the weight of the cell were plotted as a function of time. The constant rate of weight gain was obtained by linear

regression with $r^2 \geq 0.99$. The water vapour transmission rate (WVTR) was calculated from the slope of the straight line divided by the test area. WVTR ($\text{kg s}^{-1} \text{m}^{-2}$) for each type of film was determined with two individually prepared films as the replicated experimental units and three sub-samples were tested from each film. WVP ($\text{kg Pa}^{-1} \text{s}^{-1} \text{m}^{-1}$) was calculated as $\text{WVP} = [\text{WVTR}/S(R_1 - R_2)] \times d$, where S = saturation vapour pressure (Pa) of water at test temperature, R_1 = RVP in the desiccator, R_2 = RVP in the permeation cell, and d = film thickness (m). Film thickness was measured using a micrometer (Mitutoyo, Japan).

2.3.3. Sorption isotherm determination

Sorption isotherms of KGM films were determined at 30 °C according to the procedure described by Spiess and Wolf (1983), with some modifications. Films were cut into small pieces and pre-dried in a vacuum desiccator over P_2O_5 , at room temperature (25–28 °C) for 7 days, to obtain “zero” water content. The dried films (200 mg each) were weighed, to the nearest 0.0001 g, into pre-weighed weighing bottles. The dried samples, in quadruplicate, were equilibrated in air-tight 1-l Kilner jars containing different saturated salt solutions of known relative vapour pressure (RVP), at 30 °C (Greenspan, 1977). The saturated salt solutions used were lithium chloride, potassium acetate, magnesium chloride, potassium carbonate, sodium bromide, strontium chloride, sodium chloride and potassium chloride, with RVP of 0.11, 0.22, 0.32, 0.43, 0.56, 0.69, 0.75 and 0.84, respectively. Samples were weighed daily. “Equilibrium” was assumed to have been achieved when the change in weight did not exceed 0.1% for three consecutive weightings. Moisture content (dry basis) was calculated from the weight gained at this point.

2.3.4. Tensile tests

Tensile tests were performed according to the ASTM (1981b) Method D882-80a, with some modification. Film strips (each measuring 14.0×1.5 cm) were stretched at 0.2 mm s^{-1} using a TA.XT2™ Texture Analyser (Stable Micro System, Surrey, UK). Before measurement, films were equilibrated under vacuum over saturated salt solutions of known RVP (0.22, 0.43, 0.69 and 0.84), at 30 °C for at least 4 days. The reported mean value for each type of film was determined, with three individually prepared films, as the replicated experimental units and four sub-samples tested for each film.

Tensile strength (TS, Pa) was calculated by dividing the maximum load by the original cross-sectional area of the specimen. Tensile modulus (TM, Pa) was calculated from the slope of the initial linear portion of the stress–strain curve. Tensile elongation- or strain-at-break (TE, %) was calculated by dividing the elongation at the moment of rupture of the specimen by the initial gauge length (10.0 cm) of the specimen and multiplying by 100.

3. Results and discussion

3.1. Morphology

The scanning electron micrographs of KGM–PO, KGM–CMC–PO, KGM–KOH–PO and KGM–CMC–KOH–PO are shown in Fig. 1a, b, c and d, respectively. Irrespective of the film-forming formulation, destabilisation of the emulsion was observed to occur during drying. This was evident by the heterogeneous structures and rough surfaces observed in all the emulsion films studied. Generally, emulsion creaming during film dehydration would result in emulsion films of a nonisotropic nature (McHugh & Krochta, 1994). After drying, the stripped films showed two surface finishings: the smooth and shiny film side facing the casting board and the dull and rough side exposed to the air.

KGM–PO films. As reported by Cheng et al. (2002), pure KGM films are compact with smooth contours without pores or cracks. However, when PO was added into the KGM film, the microstructure was changed dramatically. The surface contour of KGM–PO films was “bumpy” and the cross-section revealed a stacked layer-structure, as shown by the formation of channels (Fig. 1a). According to Anker, Bernsten, Hermansson, and Stading (2002), different layers of an emulsion films experienced different drying rates during the drying process. At the top, evaporation and diffusion of water are extensive, whereas the lower mass transfer of water in the center results in a slower drying rate (Anker et al., 2002). Thus, the top layer gelled and set faster than the inner center layer. When PO globules creamed to the surface, they were retained and embedded under the set upper layer and gave a protruded spherical outlook. On the other hand, weak interaction between the KGM polymer chains and PO globules gave rise to extensive flocculation and coalescence between PO globules – the formation of greater channelings in the matrix were then observed (Debeaufort & Voilley, 1995; Martin-Polo et al., 1992).

KGM–KOH–PO films. Fig. 1b shows increased surface irregularity and defects, with the addition of PO into KGM–KOH matrix, as compared to KGM–PO films. From the rough surface contour, it is obvious that many spherical masses were protruding from the film surface and many crater-like pits were evident. This suggests that PO globules creamed to the surface, at a faster rate, before the KGM–KOH–PO upper layer gelled and set on time. So when the creamed PO globules reached a point that had insufficient strength to resist stress, the surface would break and craters would form. From the cross-section view, the KGM–KOH–PO matrix was compact and dense. In addition, cracks appear to propagate easily in KGM–KOH–PO films – this is a good indication of a rigid and dense matrix that was unable to tolerate the disturbance brought about by the presence of PO globules. This is in line with the previous observation, that alkaline deacetylated KGM polymer chains interact extensively to give a distinct crystalline domain with long-range order.

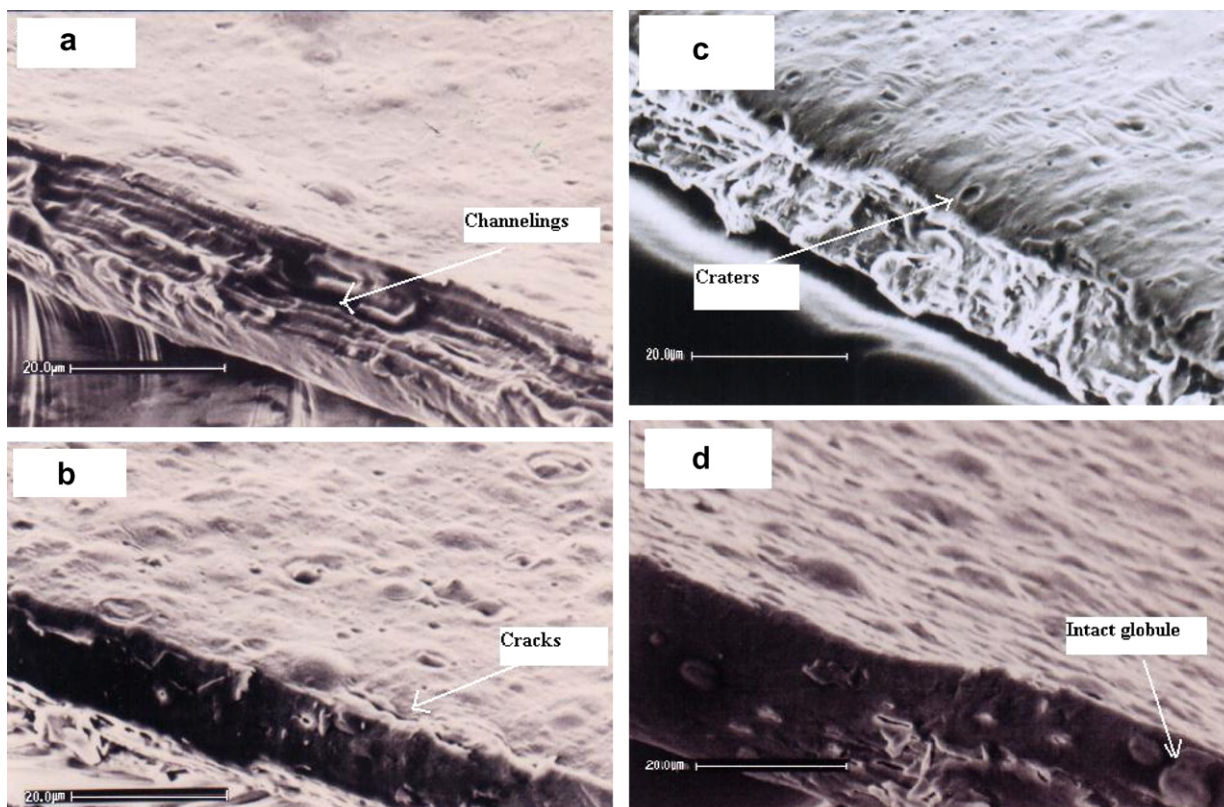


Fig. 1. Scanning electron micrographs of konjac glucomannan-based emulsion films: (a) KGM-PO, (b) KGM-KOH-PO, (c) KGM-CMC-PO and (d) KGM-CMC-KOH-PO.

KGM-CMC-PO films. Fig. 1c shows the morphology of the KGM-CMC-PO films. The surface of KGM-CMC-PO films was covered with a lot of crater-like pits. The edge view of KGM-CMC-PO films was found to possess a sponge-like and loosely filled structure. The greater formation of crater-like pits revealed a weak interaction between KGM and CMC, that failed to retain surface integrity when the surface was subjected to stress by the PO globules. This weak network is clearly evident and supported by previous X-ray diffractometry (Cheng et al., 2002), in which the X-ray diffraction pattern of the KGM-CMC film showed a much lower and broader peak. This suggests that the matrix of KGM-CMC is highly amorphous. On the other hand, the sponge-like matrix suggested that PO globules were dispersed homogeneously throughout the matrix, thereby disrupting the KGM-CMC network. The capability of the KGM-CMC-PO matrix in stabilising the emulsion during drying was enhanced in the presence of CMC, which can inherently function as an emulsifier. The possible mechanisms involved could be one or a combination of the following effects (Dickinson, 1992): (1) CMC adsorbed at the surface of PO globules and/or bridge them to contribute to a state of particle aggregation and/or (2) interaction between KGM-CMC forming a macromolecular network that entrapped PO globules in between.

KGM-CMC-KOH-PO films. The morphological characteristics of KGM-CMC-KOH-PO films are shown in Fig. 1d. Unlike the rest of the emulsion films, no signs of

channels, cracks or craters were seen in the KGM-CMC-KOH-PO films. From the side view, intact PO globules were observed and the matrix structure was compact and dense. The PO globules were dispersed homogeneously throughout the matrix. Although PO globules creamed during drying, the surface integrity was retained and PO globules were embedded under the surface. This suggests that PO globules were much more able to be “frozen” and stabilised within the KGM-CMC-KOH-PO matrix during drying, as compared with the KGM-CMC-PO matrix. The enhanced resistance to creaming and coalescence may be attributed to the extensive interaction between CMC molecules with deacetylated KGM molecules, in addition to the emulsifying properties of CMC. The organisation of the deacetylated KGM-CMC network would increase viscosity, thereby reducing mobility and the coalescence of PO globules was reduced (Debeaufort & Voilley, 1995). In other words, PO may become trapped within the compact network of deacetylated KGM with CMC.

3.2. Water vapour permeability

Table 1 shows that among the emulsion films, KGM-PO films showed the highest WVP value followed by KGM-KOH-PO, KGM-CMC-PO and KGM-CMC-KOH-PO. This does not follow the base polymer WVP pattern (when no PO was added), which decreased in the

Table 1
Water vapour permeability of KGM-based films with and without PO

Film type	WVP $\times 10^{14}$ (kg Pa ⁻¹ s ⁻¹ m ⁻¹) ^A	Film type ^B	WVP $\times 10^{14}$ (kg Pa ⁻¹ s ⁻¹ m ⁻¹) ^A
KGM-PO	2.15 \pm 0.05a	KGM	1.37 \pm 0.24bc
KGM-KOH-PO	1.82 \pm 0.08b	KGM-KOH	1.15 \pm 0.10c
KGM-CMC-PO	1.76 \pm 0.15b	KGM-CMC	1.92 \pm 0.13a
KGM-CMC-KOH-PO	1.19 \pm 0.20c	KGM-CMC-KOH	1.53 \pm 0.30b

WVP, water vapour permeability; KGM, konjac glucomannan; KOH, potassium hydroxide; CMC, sodium carboxymethylcellulose; PO, palm olein.

^A Mean \pm 1 SD ($n = 6$). Means within a column with the same letter are not significantly different at the 5% probability level.

^B Data extracted from Cheng et al. (2002).

order: KGM-CMC > KGM-CMC-KOH > KGM > KGM-KOH. This suggested that the PO added had an interactive effect with the base polymers. Comparing all film types (with and without PO addition), KGM-KOH films showed the lowest WVP value, irrespective of the film type. This was attributed to the highly structured matrix formed through hydrogen bonding, that diminished the number of available -OH groups for interaction with migrating water molecules.

When compared to their counterparts which contained no PO, KGM-PO and KGM-KOH-PO showed relatively higher WVP than KGM and KGM-KOH, respectively. The reverse was true in the case of KGM-CMC-PO and KGM-CMC-KOH-PO films, where lower WVP values were observed when compared with KGM-CMC and KGM-CMC-KOH, respectively. This is an interesting phenomenon, where the addition of a hydrophobic material (PO) into an emulsion film does not guarantee reduced WVP. To find a plausible explanation for such anomalous behaviour, one has to examine the microstructures of both the surfaces and the cross-sections of the various emulsion films studied. This is because permeability of emulsion films is influenced by the existence of steric hindrance and “tortuosity” for diffusion of water molecules (Holton, ASP, & Zottola, 1994) and also the existence of pores, voids, cracks and channelings (Torres, 1994; Wong et al., 1992).

In the case of KGM-PO films, the flocculated and/or coalesced PO globules did not help to increase the diffusion path for the water molecules. Conversely, the nonhomogeneous repartition of hydrophobic substance in the network had induced a lower efficiency in limiting water vapour transfer. McHugh and Krochta (1994) found that decreasing emulsion-particle diameters correlate well with linear decreases in WVP. Furthermore, water molecules can still permeate through the nonlipid phase. For KGM-KOH-PO films, the heterogeneous film structure and presence of cracks explain the relatively high WVP values. A moderately low WVP value given by KGM-CMC-PO films was attributed to the homogenous distribution of hydrophobic materials throughout the matrix. This increased the “tortuosity” for diffusion of water molecules through the KGM-CMC-PO film. However, its water vapour barrier properties were inferior to the KGM-CMC-KOH-PO films owing to the presence of surface defects that enhanced the absorption of water molecules.

3.3. Sorption isotherm

Statistical analysis showed that KOH and CMC, and their interaction, have significant ($P < 0.01$) effects on the sorption isotherms of the emulsion films studied. As shown in Fig. 2, KGM-KOH-PO film gave relatively lower water sorptive capacity (WSC) when compared to KGM-PO films. Conversely, KGM-CMC-PO and KGM-CMC-KOH-PO films show substantially higher water-binding capacity relative to KGM-PO films. Alkaline deacetylation

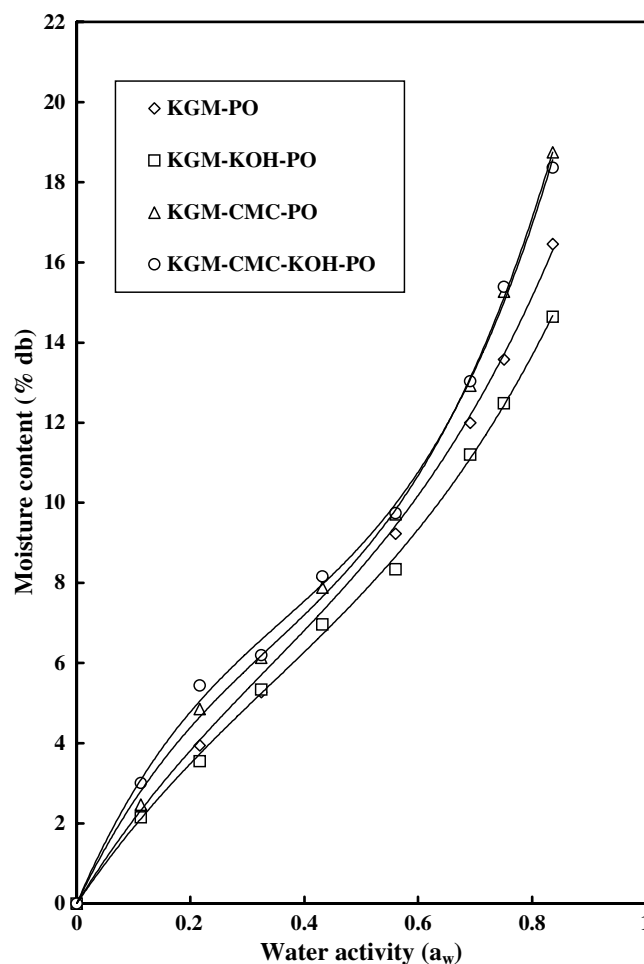


Fig. 2. Moisture sorption isotherms of konjac glucomannan-based emulsion films at 30 °C. (Typical coefficient of variation (C.V.) for quadruplicate measurements did not exceed 12%.)

promoted polymer chain association and reduced the number of active sites ($-OH$) for water-binding. On the other hand, CMC enhanced the WSC of KGM-based emulsion films, irrespective of alkaline treatment.

3.4. Tensile properties

Research on lipid/hydrocolloid composite films has focused mainly on their moisture barrier properties. The influence of lipid incorporation on the mechanical properties of hydrocolloid films has not been extensively investigated (Yang & Paulson, 2000). Furthermore, research interest was normally focused on the film properties as a function of lipid concentration (Ayranci & Tunc, 2001; Garcia et al., 2000; Yang & Paulson, 2000).

Variations in tensile modulus (TM), tensile strength (TS), and *tensile elongation* (TE) of KGM-based emulsion films, as a function of a_w , are shown in Figs. 3–5, respectively.

Effect of a_w . Tensile properties did not appear to respond linearly to changes in a_w . As the films were hydrated from 0.22 to 0.43, TM and TS were significantly

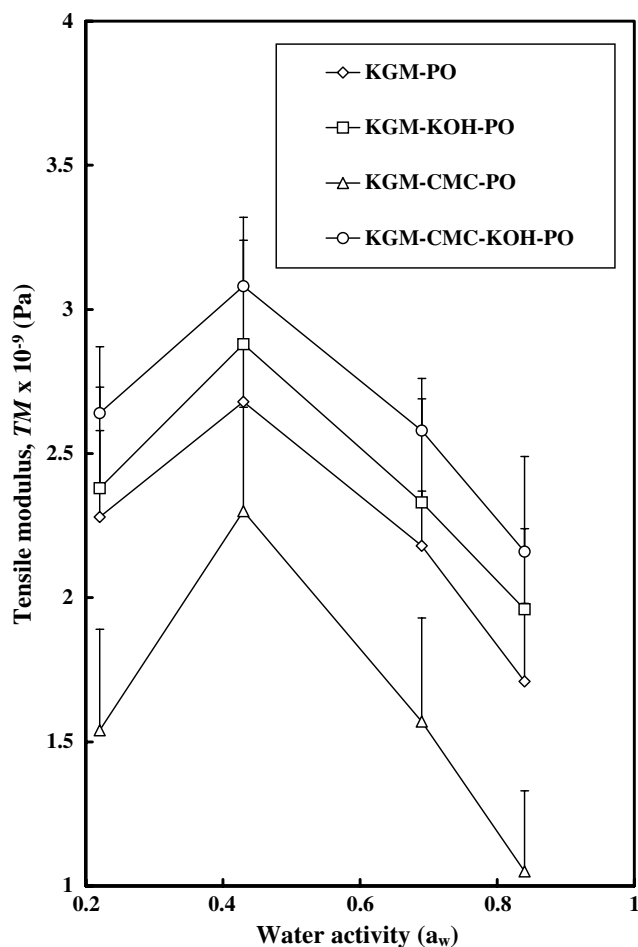


Fig. 3. Tensile modulus of konjac glucomannan-based emulsion films as a function of a_w . (Vertical bar represents plus one standard deviation from the mean.)

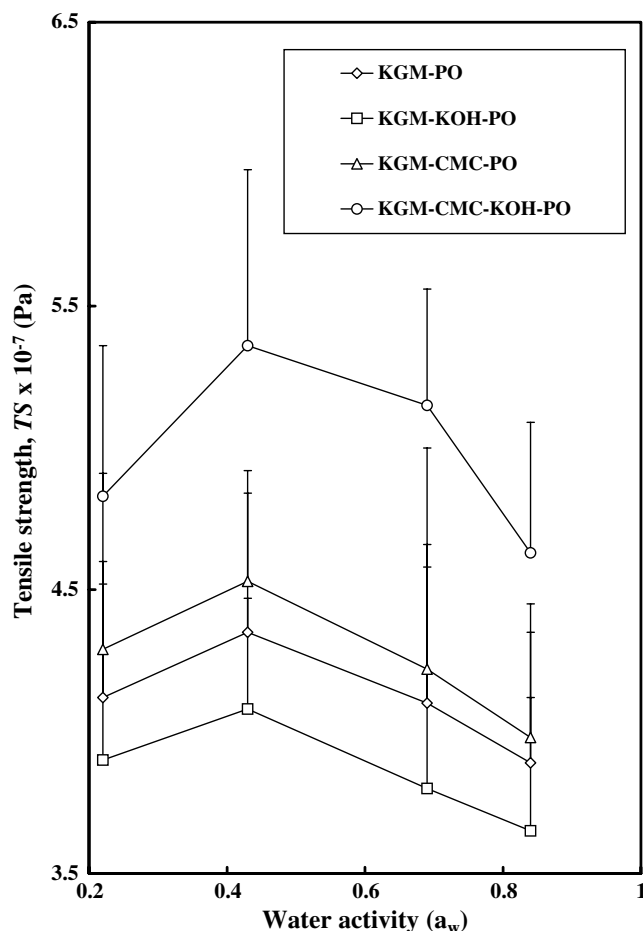


Fig. 4. Tensile strength of konjac glucomannan-based emulsion films as a function of a_w . (Vertical bar represents plus one standard deviation from the mean.)

($P < 0.01$) increased but TE was reduced. Above 0.43 a_w , tensile properties became increasingly plasticised – TM and TS were reduced and TE enhanced. Similar trends were reported by Cheng et al. (2002) for KGM-based films without the addition of PO.

Effect of PO. In comparing the tensile properties displayed in Figs. 3–5 with the corresponding results reported by Cheng et al. (2002), significant differences were observed for the following pairs of films: KGM vs KGM-PO; KGM-KOH vs KGM-KOH-PO; KGM-CMC vs KGM-CMC-PO; and KGM-CMC-KOH vs KGM-CMC-KOH-PO. Upon incorporation of PO, the overall average values of TM and TS were reduced considerably. Conversely, TE increased somewhat with the presence of PO. This appeared to be due to the plasticising effect caused by incorporation of hydrophobic materials into a hydrophilic matrix, which lead to a decrease in mechanical properties (Quezada Gallo, Debeaufort, Callegarin, & Voilley, 2000). When functioning as a plasticiser, PO globules disrupt the intermolecular hydrogen bonding and hence reduced TM and TS. On the other hand, PO could also serve as a lubricant that facilitated polymer chain mobility especially when a film piece is under stress.

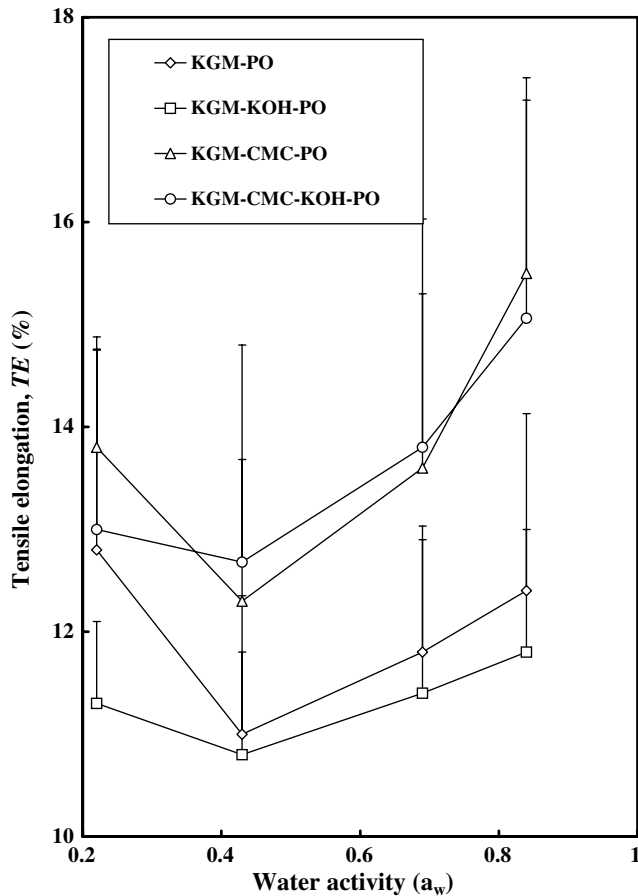


Fig. 5. Tensile elongation of konjac glucomannan-based emulsion films as a function of a_w . (Vertical bar represents plus one standard deviation from the mean.)

Effects of alkali treatment. KGM–KOH–PO and KGM–CMC–KOH–PO films show significantly ($P < 0.01$) higher TM values when compared with KGM–PO and KGM–CMC–PO films, respectively (Fig. 3). This is because alkali treatment was hypothesised to deacetylate KGM molecules and promote film strength and cohesiveness. Thus, the effect of alkaline deacetylation was manifested in TM, which gave an indication of film stiffness. However, alkali treatment generally reduced TS and TE of the films in the absence of CMC. KGM–KOH–PO films demonstrated the lowest TS and TE. As pointed out in the SEM studies, KGM–KOH–PO films possessed physical defects such as cracks. When KGM–KOH–PO films were strained, localised stress was easily triggered and propagated around the vicinity of the defects. KGM–KOH–PO film was thus unable to withstand higher stress (low TS) and failed at low strain (low TE).

Effects of CMC. When comparing TM values for KGM–PO and KGM–KOH–PO with KGM–CMC–PO and KGM–CMC–KOH–PO, respectively, it is interesting to find that at low deformation, the effect of CMC on film modulus was dependent on the presence or absence of alkali. Generally, CMC decreased film modulus. However, in the presence of alkali, a strong interaction between CMC

and deacetylated KGM resulted in an enhancement rather than a reduction in TM of the KGM-based emulsion films. However, at high deformation, the presence of CMC enhanced the TS and TE of the emulsion films, negating the effect of alkali. This could be a consequence of the interactive effect of CMC and PO in the KGM-based emulsion films, which produced a lubrication effect that made the KGM emulsion films more flexible, ductile and stretchable.

4. Conclusions

It was found that in order to produce emulsion films with higher barrier and mechanical efficiency, maintenance of a stable emulsion was a necessity during the film drying process. Films with smaller size fat globules and with a more homogeneous distribution throughout a strongly interactive film matrix is bound to give a better water vapour barrier and mechanical properties, in the absence of physical defects. This was attributed to an increase in apparent hydrophobicity and “tortuosity” of the film for water molecule transmission. This study revealed that the interaction of CMC with deacetylated KGM was able to enhance emulsion stability during drying and produced emulsion films with better barrier and mechanical efficiency.

Acknowledgements

L.H.C. is grateful to Universiti Sains Malaysia for the award of a fellowship under the Academic Staff Training Scheme. This work was supported by an 8th Malaysia Plan R&D grant under the Intensification of Research in Priority Areas (IRPA) Programme of the Ministry of Science, Technology and Environment, Malaysia.

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