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Structural Properties of Gelatin-Chitosan Composite Film Modified by Polyol

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The gelatin-chitosan composite films modified by different polyol including glycerol, sorbitol and polyethylene glycol 400 (PEG 400) were prepared using casting molding. The effects of polyol on the moisture sorption, water vapor permeability, tensile properties and transparency of the gelatin-chitosan composite films were investigated. The sorption isothermal data of the films were fitted by Guggenheim-Anderson-de Boer (GAB) model. The cross-sectional morphology of the composite films was investigated by scanning electron microscopy. The results indicated that the monolayer sorption value and tensile strength of the film decreased by the introduction of polyol, while the water vapor permeability and elongation at break increased. Glycerol and sorbitol exerted negligible effect on the transparency of the films. However, the transparency of the films plasticized with PEG 400 decreased with the ratio of PEG 400 increasing. Glycerol was a more effective plasticizer for gelatin-chitosan composite films.

Keywords: Gelatin, Chitosan, Composite film, Polyol, Property.

INTRODUCTION

The use of renewable resources to produce edible and biodegradable films that can extend the shelf life of food are a research focus of resource, material and food science and engineering. Using biodegradable material rather than conventional synthetic film can solve the problem of plastic waste disposal resulted by the lack of biodegradability[1-3]. Chitosan is an *N*-deacetylation product of natural chitin, the second abundant natural polymer on earth. It is a heteropolysaccharide, which is composed mainly of β-(1,4)-2-deoxy-2-amino-D-glucopyranose and partially of β-(1,4)-2-deoxy-2-acetamido-D-glucopyranose units [4]. As a kind of food packaging, chitosan is biodegradable, renewable and antibacterial[5,6]. Gelatin is obtained by partial hydrolysis of collagen, the most abundant structural protein in mammals. The advantages of gelatin as food packaging material include low-cost, biodegradable property, biocompatibility and abundant source[7]. The mixture of gelatin and chitosan is a good candidate for edible and biodegradable packaging materials. However, the application of gelatin-chitosan composite films has been limited due to their poor mechanical properties considering production process and end-use application, *i.e.*, fragility and brittleness[8]. Therefore, plasticizer plays an important role in gelatin-chitosan composite to determine its final structure and properties. Plasticizer could soften the film and increase the mobility of the biopolymer chains by decreasing the intermolecular forces[4], which avoid the cracking of the film during handling and storage. Current studies indicate that polyols are effective plasticizers for many natural biopolymers.

Gong *et al.*[9] studied the effects of glycerol, ethylene glycol and PEG 400 on the properties of gelatin-chitosan composite films. It showed that plasticizers could decrease the tensile strength, while increase the elongation and water vapor permeability of the composite films. When applied as packaging materials, the composite film will be exposed various external environment, for example, changes in humidity. Generally, the natural polymer material is sensitive to humidity changes. Arvanitoyannis *et al.*[10] found that the elongation and gas/water permeability of the composite films were increased with the content of water and polyol (glycerol or sorbitol) increasing, while the elasticity modulus and tensile strength were decreased.

Decreasing the water exchange between environment and the product is an important function of edible and biodegradable packaging films. The barrier properties of packing films depend on both water solubility and molecular diffusion coefficient in the matrix. The moisture sorption isotherm can be used to characterize the water absorption property of films. Some of the absorbed water would be transferred to products. Therefore, the knowledge of moisture sorption isotherm is important for predicting the stability of product quality during storage[4]. Several mathematical models can be used to describe the moisture sorption isotherms of foodstuff materials. Available models could be divided into several categories: kinetic models including Brunauer-Emmett-Teller (BET) model and Guggenheim-Anderson-de Boer (GAB) model, semi-empirical and empirical models. In recent years, the Guggenheim-Anderson-de Boer model has been widely applied to describe the water absorption behaviour[11]. According to Al-Muhtaseb *et al.*[12], Guggenheim-Anderson-de Boer parameters are more representative to explain the hydration process of food materials and natural biopolymers.

So far, the research related moisture sorption isotherm of the gelatin-chitosan composite films is still scarce. Based on the above-referred background and the drive of fundamental research and industrial application of the gelatin-chitosan composite films, the aim of the present study was to evaluate the effects of glycerol, sorbitol and PEG 400 on the properties of gelatin-chitosan composite films. This was achieved by the application of the information from GAB moisture sorption isotherm, water vapor permeability, tensile property, light transmittance and morphology.

EXPERIMENTAL

Chitosan, food grade with the *N*-deacetylation degree of 95 % from β-chitin, was purchased from Shandong AK Biotech Co. Ltd., China. Other reagents are all analytically pure. Gelatin, glycerol, sorbitol, PEG 400 and phosphorous pentoxide were procured from Tianjin Kermel Chemical Reagent Co. Ltd., China. Glacial acetic acid, sodium hydroxide, lithium chloride, potassium acetate, magnesium chloride, potassium carbonate, sodium bromide, sodium chloride, potassium chloride and potassium nitrate were obtained from Tianjin Deen Chemical Reagent Co. Ltd., China.

Preparation of gelatin-chitosan composite films with different polyols: Certain amount of chitosan was dissolved in 2 % (w/v) acetic acid solution at room temperature to obtain a 2 % (w/v) solution. 10 % (w/v) gelatin solution was obtained by dissolving gelatin in deionized water at 50 °C. Then calculated amounts of the two solutions were mixed and stirred at 50 °C for 0.5 h to get a homogeneous 50/50 gelatin/chitosan (dry basis) blending solution. Three types of polyols including glycerol, sorbitol and PEG 400 were added in different ratios (0, 10, 20 and 30 %) of the total gelatin and chitosan (dry basis). Then the solutions were stirred for 0.5 h at 50 °C. After the solutions were degassed, the mixture solutions were cast in a mould, dried at room temperature to obtain composite films with the thickness of about 0.2 mm. The composite films were neutralized by 0.25 mol/L NaOH and rinsed with deionized water for several times. Finnaly, the neutralized composite films were dried at room temperature. The dried transparent composite films were stored in a sealed container for subsequent use.

Moisture sorption: Composite films were cut into samples of 20 mm × 20 mm and were adequately dried in desiccators over P_2O_5 for 7 days. The dried samples were weighed to obtain the initial weight (M_0) and then were placed in sealed container at 25 ± 2 °C over saturated salt solution of LiCl, CH_3COOK, $MgCl_2$, K_2CO_3, NaBr, NaCl, KCl and KNO_3 to give RH 11, 23, 33, 43, 58, 75, 84 and 92 % having water activity (a_w) values of 0.11, 0.23, 0.33, 0.43, 0.58, 0.75, 0.84 and 0.92, respectively. The samples were weighed periodically until equilibrium was reached. The moisture content (M) was defined as:

$$M = \frac{M_t - M_0}{M_0} \tag{1}$$

where M_t is the sample weight at time t. All the tested values were the averages of three measurements for each composite film.

Guggenheim-Anderson-de Boer model was used to fit the sorption isotherm data of gelatin-chitosan composite film. The model can be expressed as:

$$M_e = \frac{m_0 C_{GAB} K a_w}{(1 - K a_w)(1 - K a_w + C_{GAB} K a_w)} \tag{2}$$

where M_e is the equilibrium moisture content (g water/g solids) at a_w, m_0 is the monolayer moisture content (g water/g solids), C_{GAB} and K are the GAB constants.

The fitting accuracy of GAB model was evaluated from coefficient of determination (R^2) computed from the plot of observed (M_i) and corresponding predicted (M_{pi}) moisture content at equilibrium[4] and the mean relative deviation modulus (E):

$$E = \frac{100}{n} \sum_{i=1}^{n} \frac{|M_i - M_{pi}|}{M_i} \tag{3}$$

where n is the number of observations. The E value lower than 10 is regarded to accurately represent the experimental data[13].

Water vapor permeability (WVP): The samples were dried in a desiccator for more than 2 weeks before testing. The cups filled with 10 mL distilled water were covered with a test sample at 25 ± 2 °C. The whole assembly was weighted and placed in a sealed container with silica gel. The weight loss of the cup assembly with time was measured periodically and it was plotted with respect to time. The linear regression method ($R^2 > 0.99$) was used to obtain the slope of the line, the water vapor transmission rate (WVTR) was defined as: WVTR = slope / film area

Water vapor permeability (WVP) can be expressed as:

$$WVP = \frac{WVTR \cdot L}{\Delta P} \tag{4}$$

where L is the average film thickness; ΔP is the water vapor pressure difference between inside and outside cup assembly; ΔP is 3.169 kPa at 25 °C. All the tested values were the averages of three measurements for each sample.

Tensile properties: The tensile properties of the films were tested by a microcomputer-controlled electronic testing machine (CMT6104, Shenzhen SANS Materials Detection Co. Ltd., China) according to People Republic of China, GB/T 1040.3-2006. The composite films were cut into desired shape according to the standard. The samples were conditioned at RH 43 % for 7 days before testing. The stretching speed was set at 50 mm/min. All the tested values were the averages of five measurements for each sample.

Light transmittance: The transmittance of the films was measured at selected wavelengths between 200 and 780 nm, using an ultraviolet and visible spectrophotometer (TU-1810SPC, Beijing Persee General Instrument Co. Ltd., China). Composite films were cut into samples of 50 mm × 10 mm and were conditioned at RH 43 % for 7 days before testing. An empty spectrophotometer cell was used as blank. All the thickness of samples was about 0.2 mm. The transmittance (T) of samples was calculated by eqn. 5:

$$-\log T = \frac{0.2\times A}{d} \qquad (5)$$

where A is the measured absorbance and d is the thickness (mm) of sample. Higher T value represents higher transparency.

Morphology: Composite films were frozen in liquid nitrogen and immediately fractured. The cross section of the specimen was sputtered with gold and characterized by a scanning electron microscopy (Quanta 250, FEI Co. Ltd., Czech Republic).

RESULTS AND DISCUSSION

Moisture sorption isotherms of gelatin-chitosan composite films: The moisture sorption isotherms of gelatin-chitosan composite films are shown in Fig. 1. The slope of the moisture sorption isotherm is less at lower a_w and the slope increases rapidly with the increase of a_w ($a_w > 0.58$). The polyols contribute to increase moisture content of the films when $a_w > 0.75$. This result indicates that polyol increases the molecular mobility of the film and the interaction with water for their favourable hydrophilicity at high a_w[13]. For $a_w < 0.58$, the polyols did not increase the moisture content of the composites compared with the one without polyol. Moreover, it seems that higher ratio of polyols reduced the equlibrium moisture content at low a_w. Similar behaviour was reported in amylose and amylopectin films plasticized with glycerol at $a_w < 0.5$[14] and starch films plasticized with sorbitol stored at $a_w = 0.57$[15]. This phenomenon could be explained by the fact that hydrogen bonding is the main intermolecular interaction in the adsorption mechanism at low a_w and the hydrogen bonding between polyol and polymer is stronger than that between water and polymer. Therefore the strongly immobilized structural water is replaced by polyol[14].

The parameters, coefficients of determination and E of modeling the sorption isotherms by GAB are listed in Table-1. The coefficients of determination obtained are bigger than 0.99 and the E values are less than 10. This result shows in good agreement with the fitting curves.

The monolayer value m_0 represents that the maximum amount of water can be absorbed by dry film per gram in a single layer and it is a measure of number of sorbing sites[11]. The monolayer value is of great practical interest in the physical and chemical stability of foods and natural biopolymers. Studies about the rate of chemical reactions occurring in foods have indicated that water absorbed on monolayer are not available for metamorphic reaction, therefore the rate of quality loss can be negligible for many foods in the case of monolayer water absorption[16]. Table-1 shows that m_0 decreases with the addition of polyol. The result agrees with that of Enrione *et al.*[13]. They found a similar tendency for starch films plasticized with glycerol. As mentioned, hydrogen bonding is the main intermolecular interaction while water is absorbed on monolayer and the hydrogen bonding between polyol and polymer is stronger than that between water and polymer, therefore the immobilized stable structural water is replaced by polyol[14]. The composite films plasticized with sorbitol present the lowest m_0 value at the same plasticizer ratio. The reason may be that

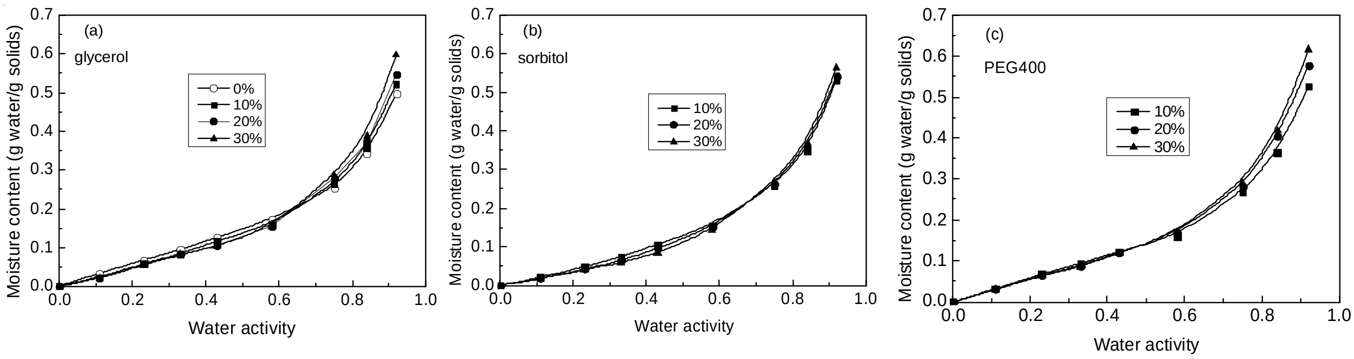

Fig. 1. Effect of polyols on the sorption isotherms for gelatin-chitosan composite films

TABLE-1
PARAMETERS, COEFFICIENTS OF DETERMINATION AND E OF GAB MODEL FOR GELATIN-CHITOSAN COMPOSITE FILMS

Plasticizer type	Ratio (%)	m_0	C_{GAB}	k	R^2	E
Glycerol	0	0.09628	5.374	0.8828	0.9989	3.476
	10	0.09475	3.926	0.8997	0.9994	5.443
	20	0.09269	3.817	0.9130	0.9984	6.258
	30	0.09137	3.538	0.9288	0.9983	7.119
Sorbitol	10	0.09089	3.325	0.9109	0.9991	6.088
	20	0.08976	2.825	0.9189	0.9988	8.238
	30	0.08786	2.546	0.9298	0.9977	8.482
PEG 400	10	0.09546	4.814	0.8980	0.9996	3.101
	20	0.09378	4.519	0.9196	0.9987	2.895
	30	0.09313	4.321	0.9308	0.9987	3.374

sorbitol is similar to the molecular structure units of chitosan and there are stronger intermolecular forces in the composite films plasticized with sorbitol, resulting in decreased available sites of water absorbing[11]. The m_0 values for the composite films plasticized with PEG 400 are higher than those of glycerol and sorbitol and close to the composite films without polyol. This phenomenon could be due to the fact that PEG 400 contains the minimum hydroxyl at the same amount of plasticizer and there are the least chances for PEG 400 molecules interacting with gelatin and chitosan molecules, resulting in the smallest reduction in sorption sites.

The constant C_{GAB} decreases while k value increases with the polyol ratio increasing. The C_{GAB} is related to the heat of sorption of monolayer. The decrease in this value suggests that water molecules are bounded to the sorption sites of the matrix less strongly when polyol is present, indicating that polyol may occupy some sorption sites of the matrix[13]. The composite films plasticized with sorbitol present the lowest C_{GAB} value at the same ratio of plasticizer. As mentioned above, sorbitol is similar to the molecular structure units of chitosan. Therefore, there are stronger intermolecular forces in the composite films plasticized with sorbitol, which may result in some available sites to absorb water occupied by sorbitol[11]. The C_{GAB} values for the composite films plasticized with PEG 400 are higher than that of glycerol and sorbitol and close to the composite film without plasticizer. It is possible that PEG 400 contains the minimum hydroxyl groups at the same amount of plasticizer and there are the least chances for PEG 400 interacting with gelatin and chitosan molecules, which results in the slightest effect on the available sites to absorb water. The k value is related to the difference between the heat of sorption of the multilayer and the heat of condensation of pure water. As the latter remains constant, the increase in this value toward a value of 1 would indicate a reduction in sorption energy of the multilayer[13] and a decrease of the sorbed water in multilayer when the polyol is present.

Water vapor permeability (WVP): The water vapor permeability and mechanical properties of gelatin-chitosan composite films are shown in Table-2. The WVP increases with the ratio of polyol increasing. Similar behaviour was reported for gelatin films plasticized with glycerol[17], chitosan films plasticized with glycerol[1] and gelatin-chitosan composite films with glycerol and sorbitol[10]. The reason might be that the addition of polyols increases moisture adsorbability of the composite films and makes the absorbed water diffuse through the films and desorb from the film surface more easily. Martelli *et al.*[2] stated that water vapor permeating through polymer films proceeds through four steps: (I) absorption of water vapor on to the polymer surface; (II) solution of water vapor into the polymer matrix; (III) diffusion of water vapor through the polymer; and (IV) desorption of water vapor from the polymer surface. Fig. 1 shows that moisture content of the composite films increases with the increase of plasticizer content (that is, higher adsorbability and solubility) at higher water activity ($a_w > 0.75$). Table-1 shows that the increase of the plasticizer content results in the decrease of the m_0 and C_{GAB} value and the increase of the k value, which indicates that monolayer adsorbed water, multilayer adsorbed water and adsorption strength decrease with the increase of plasticizer content, so free water content increases and composite film adsorbed water diffuse through the film and desorb from the film surface more easily. Comparing with the composite film plasticized with glycerol, the composite film plasticized with sorbitol shows lower m_0 and C_{GAB} value and higher k value, so the WVP of the latter is higher. While comparing with the composite film plasticized with glycerol, the composite film plasticized with PEG 400 shows higher m_0 and C_{GAB} and lower k value and the WVP of the composite film plasticized with PEG 400 is higher. This phenomenon may be due to the destabilization of gelatin-chitosan matrix by long-chain PEG 400 molecules, which lead to widening the interstitial space in the gelatin-chitosan matrix and allowing for an increased diffusion rate of water vapor through the films[4].

An important function of food packaging is to avoid or decrease the water molecules migration between the product and the surrounding environment, so WVP should be as small as possible. According to the above comparison, it could be concluded that glycerol might be a more suitable plasticizer for gelatin-chitosan composite films.

Tensile property: Tensile property is one of the most important properties of biodegradable films considering production process and end-use application. Tensile strength shows the maximum tensile stress that the film can sustain, elongation at break is the maximum change in length of a test sample before breaking. The tensile properties of gelatin-chitosan composite films plasticized with different polyols are shown in Table-2. The results indicate that the tensile strength and elongation at break of the films vary with the ratio and nature of polyol used. With the ratio of plasticizer increasing, the tensile strength of the samples decreases, while the elongation at break of the

TABLE-2
WATER VAPOR PERMEABILITY (WVP) AND MECHANICAL PROPERTIES OF GELATIN-CHITOSAN COMPOSITE FILMS

Plasticizer type	Ratio (%)	WVP (10^{-10} gms^{-1}m^{-2}Pa^{-1})	Tensile strength (MPa)	Elongation at break (%)
Glycerol	0	1.45 ± 0.07	85.6 ± 3.3	1.7 ± 0.5
	10	1.75 ± 0.12	83.9 ± 3.8	3.7 ± 0.5
	20	1.82 ± 0.07	60.4 ± 3.2	12.3 ± 0.6
	30	1.96 ± 0.17	45.6 ± 2.0	31.3 ± 2.8
Sorbitol	10	2.23 ± 0.03	81.3 ± 3.4	2.1 ± 0.3
	20	2.28 ± 0.04	79.8 ± 2.0	8.8 ± 0.4
	30	2.37 ± 0.03	65.1 ± 3.9	16.7 ± 1.8
PEG 400	10	2.23 ± 0.12	66.4 ± 1.2	6.2 ± 0.4
	20	2.34 ± 0.07	43.1 ± 1.0	18.6 ± 0.6
	30	2.43 ± 0.23	39.88 ± 1.62	18.2 ± 2.4

samples increases. Therefore the flexibility of the composite films increases with the ratio of plasticizer increasing. The result is similar to the previously reported work in which the flexibility of hydrophilic films increased with the increase of the plasticizer ratio[11,18-20]. It could be related to the structural changes of the gelatin-chitosan network when plasticizer was added. Plasticizer is able to insert itself between two polymer chains and form polymer-plasticizer hydrogen bonds which replace the polymer-polymer hydrogen bonds, to reduce the internal hydrogen bonding among polymers, decrease the energy required for molecular motion and increase molecular mobility and as a result, the flexibility of the films is increased. However, small increase was found in the elongation at break at 10 % of plasticizer. These results agree with Ghasemlou *et al.*[21] that kefiran films plasticized with glycerol and Cao *et al.*[22] that gelatin films plasticized with PEG, respectively. Ghasemlou suggested that the lowest effective glycerol ratio was 10 % (w/w). Below this ratio, the films tend to be brittle. Cao mentioned that the addition of 10 % PEG could not improve the flexibility of gelatin film effectively. When the ratio of plasticizer increased to 20 % (w/w), the composite films plasticized with PEG 400 showed lower tensile strength and higher elongation at break comparing with films plasticized by glycerol or sorbitol. This phenomenon could be attributed to the fact that the interstitial space in the matrix is widened with the addition of long-chain PEG 400 molecules and the internal hydrogen bonding among the matrix is reduced, therefore the relative movement among the macromolecular chains is increased. When the ratio of glycerol increased from 20 % (w/w) to 30 % (w/w), the elongation at break of the composite film increases significantly and this behaviour could be due to the fact that adding a large amount of glycerol could effectively reduce the internal hydrogen bonding among polymers, which lead the relative movement between the macromolecular chains increase significantly, thus the flexibility of the composite films is increased markedly. At the ratio of PEG 400 increased from 20 % (w/w) to 30 % (w/w), the elongation at break of the composite film decrease slightly. The reason may be that the amount of hydroxyl groups in PEG 400 is less comparing with glycerol and sorbitol at the same mass and the polarity of PEG 400 molecules is lower, therefore the intermolecular interaction between PEG 400 with gelatin and chitosan molecules are weaker[22]. When the plasticizer content exceeds its compatibility limit in the polymer, phase separation would occur and free (excess) plasticizer would separate from the film matrix and migrate to the film surface. Compared with the composite films plasticized with glycerol, the composite films plasticized with sorbitol show higher tensile strength and lower elongation at break. This phenomenon could be due to lower moisture content of the latter at the same plasticizer ratio. It is well known that water could act as an effective plasticizer to reduce the internal hydrogen bonding among polymers and increase the relative movement between the macromolecular chains, so sorbitol exhibited a less efficient plasticizing effect. It could be concluded that glycerol might be the most suitable plasticizer for gelatin-chitosan composite films. Significant plasticizing effect for glycerol could be found at the ratio higher than 30 %.

Light transmittance: The light transmittance of gelatin-chitosan composite films is shown in Fig. 2. The transparency of materials is an advantage in food packaging, for it allows consumers to see the product before buying and the products with an attractive appearance could be better presented by sellers[23]. In general, the light transmittance of the composites with poor compatibility will be low due to light reflection or scattering at the two-phase interface. Fig. 2 shows that transparency in the visible region is high when the composite films are plasticized with glycerol, sorbitol and without plasticizer. Therefore, there is a good compatibility between the glycerol, sorbitol and the matrix. However, the transparency of the composite films plasticized with PEG 400 decreases with the increase of the PEG 400 ratio. As previously mentioned, the polarity of PEG 400 molecules is lower than glycerol and sorbitol and there are less chances for PEG 400 interacting with gelatin and chitosan molecules[22]. When the ratio of PEG 400 exceeds its compatibility limit in the polymer, phase separation would occur and the excess (free) PEG 400 would separate from the film matrix and migrate to the film surface. Phase separation would lead to a heterogeneous structure and as a result, the light transmittance of the samples would be decreased by the action of light scattering in the two phase interface.

Moreover, packaging materials should have the function of protecting food from the effects of UV radiation. The present study demonstrates that gelatin-chitosan composite films perfectly meet this requirement. All analyzed films, regardless of the ratios or types of polyols, reveal excellent UV rays barrier properties. UV rays can be divided into UV-A, UV-B and UV-C radiation rays, corresponding to wavelength range 400-315, 315-280 and 280-200 nm, respectively. The energy will be higher and the impact on food stability will be greater with shorter wavelength. Fig. 2 shows the transmittance of light in UV-B region does not exceed 5 % and that in UV-C region

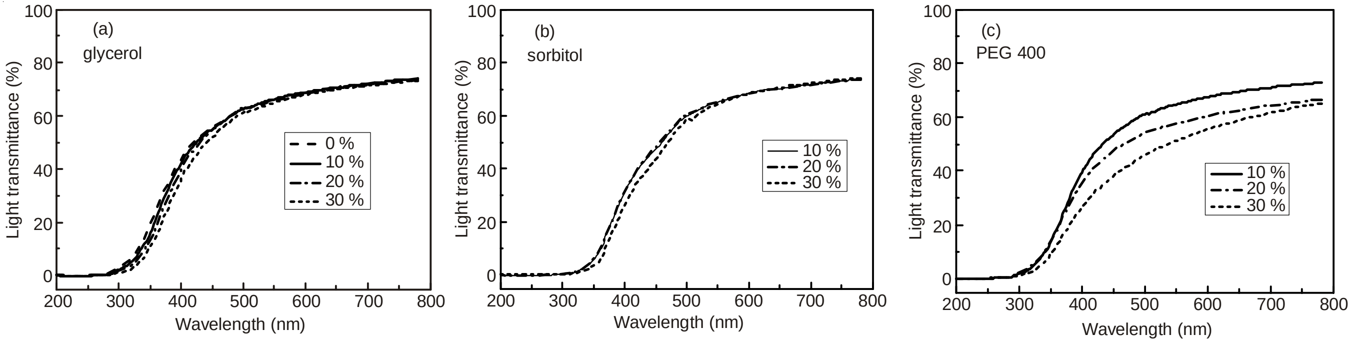

Fig. 2. Effect of polyols on the light transmittance of gelatin-chitosan composite films

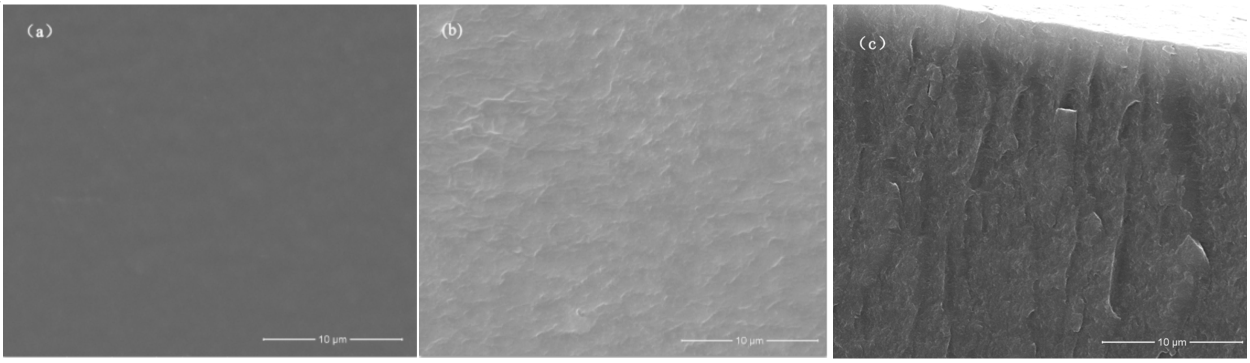

Fig. 3. SEM cross sectional images of gelatin-chitosan composite films

does not exceed 1 %. The results indicate that gelatin-chitosan composite film may have potential possibilities to retard lipid oxidation in foodstuffs induced by UV light. Similar results were found in pea protein isolate films plasticized with glycerol and sorbitol[23] and whey protein films plasticized with glycerol[24,25]. This related to the fact that gelatin and chitosan possess strong absorption in UV region. Barrier properties of gelatin against UV radiation relate to the presence of UV-absorbing chromophore, especially tryptophan and tyrosine and secondly, phenylalanine, disulfide bonds and peptide bonds[23].

Morphology: In order to understand the changes in the microstructure of the composite films, SEM was used to investigate the cross sectional morphology of the composite films plasticized with glycerol, sorbitol and PEG 400 at the ratio of 0, 10, 20 and 30 %, respectively. Fig. 3 shows the SEM cross sectional images of the gelatin-chitosan composite films containing 30 % glycerol, 20 % PEG 400 and 30 % PEG 400.

In general, the SEM images of the cross-sectional morphology of the gelatin-chitosan composite films show smooth and uniform cross sections without pores or cracks (as Fig. 3 (a)), regardless of the type and amount of plasticizers, indicating a high compatibility among components. However, heterogeneous cross section is observed in the films plasticized with 20 % PEG 400 (Fig. 3 (b)) and 30 % PEG 400 (Fig. 3 (c)). As previously mentioned, the ratio of PEG 400 exceeds its compatibility limit in the polymer, which makes phase separation occur and then leads to an uneven structure.

Conclusion

The moisture content of the composite films increased with the increase of water activity. Guggenheim-Anderson-de Boer model could accurately describe the adsorption isotherm of composite films. The monolayer values and binding strength between water molecule and active site of the matrix were decreased with the ratio of polyol increasing. Polyol could increase water vapor permeability of the composite films, among which glycerol showed the least effect at the same plasticizer ratio. The flexibility was increased with the ratio of plasticizers increasing. The highest elongation at break could be found when the ratio of glycerol is 30 %. While the ratio of PEG 400 increased from 20 % (w/w) to 30 % (w/w), elongation at break of the film decreased slightly. The effect of polyol on the transparency of the composite films was very little except composite films with 20 % PEG 400 and 30 % PEG 400. All tested films showed excellent function to absorb UV radiation. The decrease in elongation at break and transparence of the composite films plasticized with PEG 400 was caused by phase separation and this was confirmed by scanning electron microscopy. Glycerol was the most effective plasticizer in the plasticizers studied.

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