Effect of Molecular Weight and Concentration of Polyethylene Glycol on Physicochemical Properties and Stability of Shellac Film

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The effects of molecular weight and concentration of plasticizer on physicochemical properties and stability of shellac films were investigated. Type of plasticizer was previously reported to have some effects on the stability of shellac films, and polyethylene glycol (PEG) was the plasticizer of choice for plasticizing shellac films. In this study, different molecular weights of PEG (200, 400 and 4000) were chosen at a concentration of 10% w/w of shellac films. Shellac in alcohol was prepared in a free film. The stability of shellac film was then performed at 75% RH, 40 °C for 3 months. The comparison was made between the film with and without plasticizer. Shellac films were then determined for acid value, insoluble solid, mechanical properties and water vapor permeability coefficient. It was reported that different molecular weights of PEG had some influence on physicochemical properties of the shellac films. Among different molecular weights of PEG, PEG 400 showed a suitable molecular weight that could protect the shellac chain at the carboxylic and hydroxyl groups. Therefore, the molecular weight of plasticizer played a crucial role for the protective ability at active sites. Further study was performed to investigate the effect of concentrations of PEG 400 on the stability. The results demonstrated that PEG 400 at a concentration of 10% (w/w) could prevent the polymerization process for only 4 months and a significant change of all parameters was then reported. However, a higher concentration, 20% (w/w) of PEG 400, could prolong the stability of shellac for 6 months of study. Therefore, the drawback of shellac as a natural polymer in pharmaceutical and food industries could be tackled by the appropriate size and concentration of plasticizer.

KEYWORDS: Shellac; polyethylene glycol; plasticizer; polymerization; and stability

INTRODUCTION

Shellac is a purified resinous secretion of lac insects, *Laccifer lacca*, which are mostly cultivated in host trees from India and Thailand (*1*). Shellac was reported as a complex mixture of polyesters and single esters, which contain hydroxyl and carboxyl (*2*, *3*). Due to excellent film forming and protective properties, it is widely used in food, agriculture and paint industries, to some extent still in the pharmaceutical industry and a market of growing interest in nutritional and health supplements and nutriceuticals (*1*). However, problems such as variation in batches, less stability and mechanical brittleness restricted its wide applications. In the pharmaceutical industry, shellac has been used for moisture protection and glossing, while the use for enteric coating of pharmaceutical products has been greatly declined (*4*−*7*).

Plasticizing is one of the effective methods for providing new materials for a variety of applications. Plasticizers are generally essential to overcome the brittleness of biopolymer films. Brittleness is an inherent quality attributed to the complex/branched primary structure and weak intermolecular forces of natural polymers. Plasticizers, by reducing the intermolecular forces, soften the rigidity of the film structure and increase the mobility of the biopolymer chains, thus improving the mechanical properties (*8*). There are some published studies in the literature related to the effect of plasticizers on the properties of film. Polyethylene glycol (PEG), which is a nonionic water-soluble polymer consisting of polar ethylene glycol units, has been used in methylcellulose-based films (*9*). Glycerol was shown to improve film flexibility, reduce film puncture strength, and affect elasticity and water vapor barrier properties of wheat gluten films (*10*). The composition, size and shape of a plasticizer as well as its compatibility with the polymer could affect the interactions between the plasticizer and the polymer, including its ability to attract water to the plasticized protein films (*11*). Plasticizers with characteristics such as small size, high polarity, more polar groups per molecule, and greater distance between polar groups within a molecule generally impart greater plasticizing effects on a polymeric system. The selection of a plasticizer for a specified system is normally based on the compatibility and permanence of the plasticizer, the amount of necessity for plasticization, and the desired physical

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properties of the films (*12*). The flexibility of starch films can be improved by the incorporation of plasticizers (*13*). The major T_g values of the plasticized starch/decolorized hsian-tsao leaf gum films were relatively independent of the percentage of the added plasticizer (*14*). From our previous study, the aging of shellac was reported due to the polymerization among hydroxyl groups and carboxyl groups, resulting in the instability of shellac. The instability caused changes in the various properties such as acid value, insoluble solid, water vapor permeability coefficient (WVPC), and mechanical properties (*15*−*20*). The decrease in acid value and the increase in insoluble solid indicates that the polymerization of shellac film could be due to the loss of plasticizers. The stability of shellac could be improved when shellac was in an ammonium salt form which was a result of the protection of ammonium at the carboxylic groups of shellac, resulting in less polymerization (*19*−*23*). The effect of various plasticizers, such as diethyl phthalate, triacetin and polyethylene glycol 400 with the same concentration, on the mechanical properties and stability of shellac films was further studied. The percent insoluble solid of unplasticized film increased as the prolongation of storage time, confirming the instability because of the polymerization. A significant change in insoluble solid was found for plasticized shellac with DEP (diethyl phthalate) and TA (triacetin) whereas the addition of PEG 400 gave the highest stability. In addition, PEG 400 might still be retained in shellac as compared with other plasticizers because of the higher molecular weight and less volatilization. It was reported that using PEG 400 as plasticizer significantly improved the mechanical properties and the stability of film (*19*). This proved that the molecular weight of plasticizer played a crucial role in the stability of polymer film. Apart from the molecular weight, the concentration of plasticizers was also reported to affect the properties of film (*15*, *24*). The higher the glycerol content, the higher water vapor permeability and flexibility of gellan film were obtained, contributing to the higher hydrophilicity of a higher concentration of glycerol (*24*). An increase in the concentration of polyethylene glycol exhibited an enhancement of the flexibility of shellac film while reduction of the mechanical strength and glass transition temperature (T_g) of shellac film were obtained (*15*).

Therefore, the objective of the present study was to investigate the effect of different molecular weights and concentrations of PEG on the stability of shellac films. The plasticized and unplasticized shellac was prepared in a free film by the casting film method. The stability was then carried out at 40 °C, 75% RH. The evaluation parameters were acid value, insoluble solid, water vapor permeability, and mechanical properties.

EXPERIMENTAL DETAILS

2. Materials and Methods. *2.1. Materials.* Shellac was kindly supplied from Union Shellac Part., Ltd. (Bangkok, Thailand). The plasticizers used were polyethylene glycol (PEG) of molecular weight 200, 400, and 4000 kDa (Fluka, The Netherlands). The solvent used was ethanol obtained from Merk, Germany.

2.2. Methods

2.2.1. Preparation of Shellac Films. The 12% (w/w) shellac film was prepared by dissolving 6 g of shellac in 30 g of ethanol with constant stirring until a clear solution was obtained. Different molecular weights of PEG (200,400 and 4000) at the concentration of 10% w/w of shellac (0.6 g of PEG) were added to shellac solution, stirred for 30 min and finally adjusted to a total weight of 50 g with ethanol. These solutions were centrifuged for 10 min at 3,000 rpm, and insoluble solid was removed by paper filtration. An accurate weight of 50 g of filtrate was poured onto a Teflon plate and allowed to dry into film for 1−2 h at 50 °C in an oven. The dried film was peeled off from the Teflon plate and kept in a desiccator for 1 h prior to the stability study. The 3-month stability of unplasticized and plasticized shellac films for the effect of molecular weight was investigated

while the effect of concentration based on 10% and 20% of PEG 400 was studied for 6 months.

2.2.2. Acid Value and Insoluble Solid. The acid value was determined by the acid−base titration method. An accurately weighed 3 g of shellac film was dissolved in ethanol overnight and finally adjusted to a total accurate weight of 39 g with ethanol. The solution was centrifuged for 10 min at 400 rpm and filtered through filter paper. An accurate weight of 26 g of filtrate was titrated with 0.1 N sodium hydroxide. The end point was determined by pH meter. The acid value was expressed as milligrams of potassium hydroxide for neutralization of free carboxyl group in 1 g of shellac. The measurements were performed in duplicate. The insoluble solid on filter paper was dried at 70 °C until the dried weight was constant. The percentage of insoluble solid was calculated from the weight difference of filter paper that was dried at 70 °C before and after filtration.

2.2.3. Water Vapor Permeability Coefficient (WVPC). An adapted permeation cell for water vapor permeation study was used according to the method described in the Annual Book of ASTM Standards (*25*). The cell consisted of a glass bottle, filled with 30 g of dried calcium chloride granular and a cap with an opened circular hole in the area of 800−900 mm². Shellac film with a circular shape was placed inside the cap and then sealed tightly. The cell was then kept in a stability chamber at 40 °C and a relative humidity of 75% RH for 6 months. The weight change was recorded every 24 h. The WVPC of at least eight cells for all films was then calculated using the formula WVPC = $(W \times T)/(A \times \Delta P)$ where W is the amount of water permeated through the film (mg/h), T is the thickness of film (mm), A is the area of film, and ΔP is the pressure difference (mmHg).

2.2.4. Mechanical Properties. The mechanical properties of shellac film were expressed by stress and strain value at break. The prepared films were measured for thickness and cut into a dumbbell shape with length of 25 mm and width of 6.045 mm. Measurement by the texture analyzer (TA . XT Plus, England) with a speed of 10 mm/min and a 100 N load cell was applied. The parameters were calculated as follows: stress = $P/(T \times W)$, where P is the maximum applied force (N), T is the thickness of film (mm), and W is the width of film (mm); % strain = $(\Delta L/L) \times 100$, where ΔL is the length difference (mm) and L is the original length of film (mm). At least 10 measurements were conducted.

2.2.5. Statistical Analysis. The data of this research was analyzed by means and standard deviation (means ± SD).The statistical analysis of data was determined by the analysis of variance (one-way ANOVA) at the 0.01 significance level.

RESULTS AND DISCUSSION

3.1. The Effect of Different Molecular Weight of PEG (200, 400, 4000) on Physicochemical Properties and Stability of Shellac Film. From our previous study, the instability of shellac was due to the polymerization of shellac at carboxylic groups and hydroxyl groups. One means to improve the stability was to protect at these functional groups (*10*−*19*, *23*). Plasticizers were used to protect at these groups. It was reported that appropriate plasticizer could protect shellac from polymerization. Among the studied plasticizers, PEG 400 was the only plasticizer that could protect shellac from esterification. The protection capability of PEG was due to the larger molecular weight and unlost plasticizer during storage (*19*). Therefore, different molecular weights of PEG, i.e., 200, 400, and 4000 were chosen to protect shellac from esterification and were investigated as shown in **Figures 1**−**5**.

3.1.1. Acid Value and Percentage of Insoluble Solid of Shellac Film. The physicochemical properties, including acid value and insoluble solid, were measured to study the effects of different molecular weights of PEG on the stability of shellac films. The acid value is expressed as the total number of carboxyl groups, which can indicate the solubility of enteric polymers such as shellac (*21*). Shellac films with various molecular weights of PEG at 10% w/w were compared with unplasticized shellac film.

As shown in **Figure 1**, the acid value of all films slightly decreased after storage for 90 days. The slight decrease in acid value was due to the component of shellac consisting of large numbers of carboxyl groups from polyester and single ester. The acid
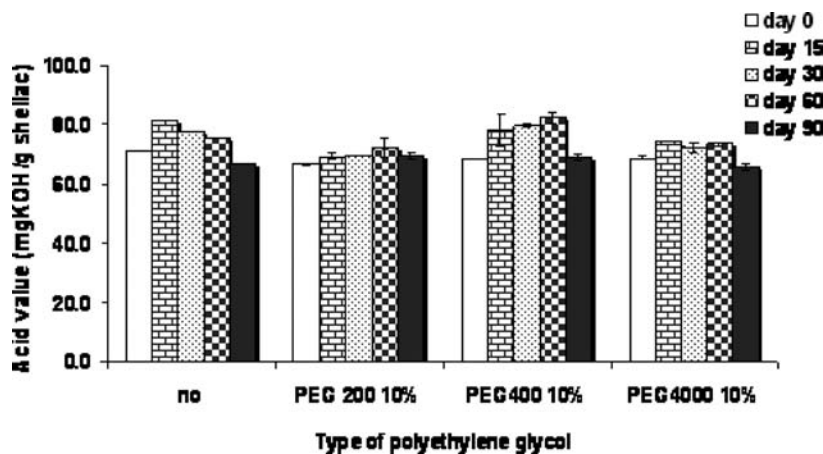
Figure 1. Effect of various polyethylene glycols on acid value of shellac films after storage at 40 °C, 75% RH for 3 months.
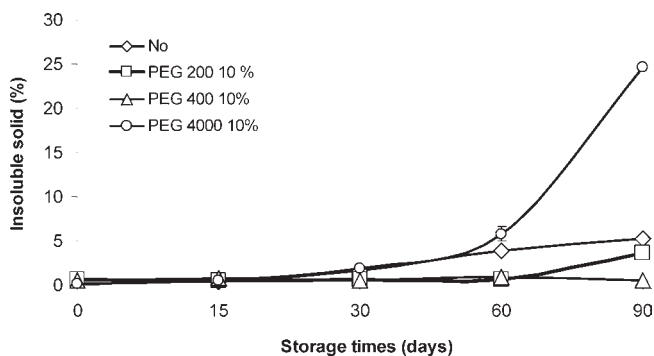

Figure 2. Effect of various polyethylene glycols on insoluble solid (IS) of shellac films after storage at 40 °C, 75% RH for 3 months.

value of shellac is the average value of polyesters, which have a lower acid value, and single esters, which have a higher acid value. The polyester tended to polymerize first such that the average acid value decreased slightly in the first three months of storage (*26*). However, increased acid value initially was noticed due to the temperature and moisture effect of the stability chamber causing the hydrolysis of shellac prior to the occurrence of polymerization. The hydrolyzed shellac resulted in an increase in hydroxyl group, and a higher acid value was obtained. The higher acid value due to the hydrolysis effect was also reported in our previous study (*20*). From this study, the influence of different type of PEG had less effect on the change of acid value.

Insoluble solid parameter is one of the indicators that show the stability of shellac. Upon storage, shellac films were polymerized and a percentage of insoluble solid was formed. **Figure 2** demonstrates the percent insoluble solid of shellac films in the presence of different molecular weights of PEG. The insoluble solid values were 6%, 3%, 0.3% and 25% for unplasticized and plasticized shellac with PEG 200, 400 and 4000, respectively, after 90 days of storage. A slight change was observed for unplasticized and plasticized shellac with PEG 200 while a profound increase ($p < 0.01$) was shown for plasticized shellac with PEG 4000 after 90 days of storage. An extremely low amount of insoluble solid was reported for shellac plasticized with PEG 400. The impact of molecular weight of PEG could be explained by a polymerization diagram as indicated in **Figure 3**. The polymerization of shellac could be protected by hydrogen bond formation between carboxyl or hydroxyl group of shellac and hydroxyl group of PEG. PEG 200 and 400 could prevent the polymer chains of shellac from the cross-linking formation of the esterification process, and hence lower insoluble solid was shown compared with unplasticized

shellac. However, the PEG 4000 could not perform the protective capability efficiently due to the longer chain of PEG 4000 causing difficulty in insertion between the polymer networks. The higher protection ability of PEG 400 could possibly be due to the proper length of the PEG 400 chain in preventing the esterification. Therefore, the length of the PEG chain played a crucial role for the protection ability. In addition, PEG 4000—the high polarity indicated by the high surface free energy—tended to accelerate the polymerization process to a high extent compared with unplasticized shellac. The surface free energy of PEG 200, 400, and 4000 was 35.3, 42.6, and 87.2 mJ/m^2, respectively. (Data was measured in this study.) The high polarity was due to the higher amount of OH group per molecule, contributing to stronger hydrogen bonding such that the closer shellac chain network formed, leading to enhancement in polymerization. PEG 200 could protect shellac up to 90 days, and further storage gave higher insoluble solid (data was not shown) resulting from the loss of plasticizer upon storage. Therefore, PEG 400 was the only plasticizer that could protect shellac from polymerization, indicating that the molecular weight of plasticizer contibuted to the stability of shellac and hence the optimum molecular weight was required to protect shellac from polymerization. The result was in agreement with our previous study (*19*). The stabilization effect of plasticizer was reported due to the protection of plasticizer at active sites and hence the lower polymerization of the shellac chain. PEG 400 was the only plasticizer that could protect at active sites whereas triacetin (TA) and diethyl phthalate (DEP) could not, attributed to the lower molecular weight and the loss of plasticizers.

3.1.2. The Water Vapor Permeability Coefficient of Shellac Film. The effects of PEG with different molecular weights on WVPC of shellac film are displayed in **Figure 4**. The effect of molecular weight had some effects on WVPC differently; the addition of PEG400 and 4000 caused higher WVPC whereas PEG 200 gave lower WVPC than unplasticized shellac and other MWs of PEG. The increase in WVPC was in accordance with some other works (*17, 27*). The addition of some plasticizers, e.g. glycerol, sorbitol, and polyethylene glycol, had increased the WVPC of films prepared from various polymers due to the high aqueous solubility of plasticizer (*28, 29*). The decrease in WVPC for PEG 200 might be due to the more compact form comparing to PEG 400 and PEG 4000, hence the lowest WVPC. The result was in agreement with the work of Cao et al., which reported a better plasticizing effect for gelatin film plasticized with the lower MW of PEG and the higher WVPC obtained with an increase in the molecular weight of PEG (*29*). The increased WVPC with the increase in MW of PEG complied with the hydrophilicity of shellac film plasticized with PEG, indicated by the higher surface
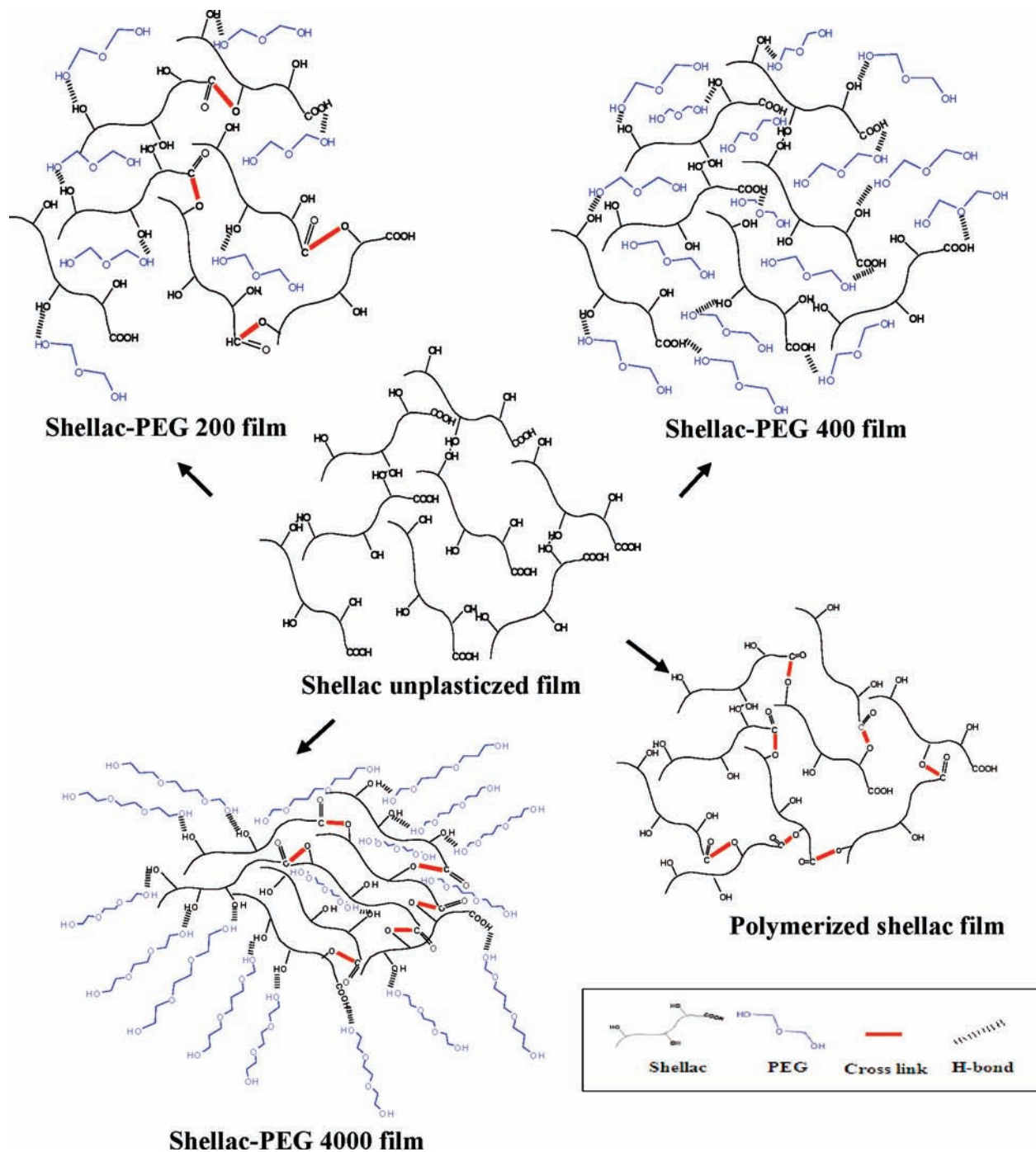
Figure 3. Polymerization diagram of unplasticized and plasticized shellac film with PEG (200, 400, 4000) after storage at 40 °C, 75% for 3 months.
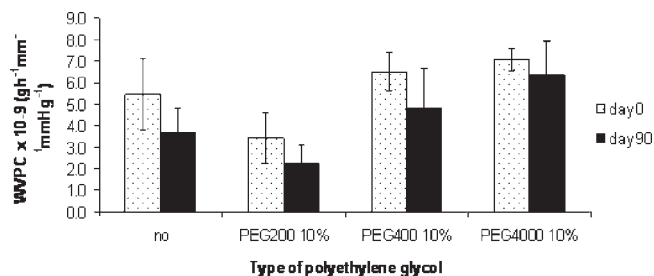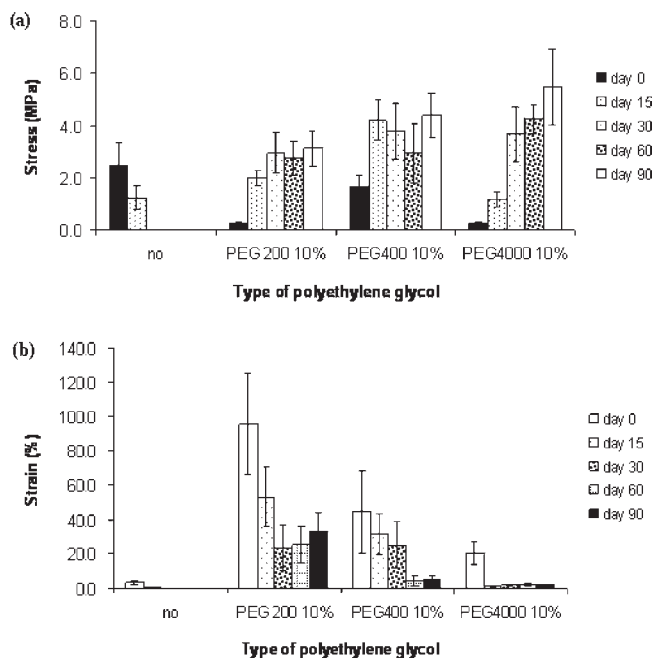

Figure 4. Effect of polyethylene glycols on water vapor permeability coefficient (WVPC) of shellac film after storage at 40 °C, 75% RH for 3 months.

free energy. The surface free energy of shellac film was 26.24, 28.86, 31.98, and 35.86 mJ/m^2 for unplasticized and plasticized

with PEG 200, 400, and 4000 respectively. (Data was determined in this study.) Upon storage of all films for 3 months, WVPC decreased slightly for all plasticized films. The reduction of WVPC might be a result of the loss of free volume causing the tighter structure of film, contributing to the lowering of WVPC.

3.1.3. The Mechanical Properties of Shellac Film. The effects of PEG with different molecular weights (200, 400 and 4000) on the mechanical properties of shellac film are displayed in **Figure 5**. Initially, all plasticized shellac films had lower stress than the unplasticized shellac film as shown in **Figure 5a**. Initially, the addition of PEG at all molecular weights caused the reduction of polymer interaction, hence the lower stress obtained (*29*, *30*). The decrease in stress could be due to the effect of the reduction of intermolecular attraction inside the shellac network. The result was in agreement with Sothornvit and Krochta (*11*) and

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(a)

(b)

Figure 5. Effect of polyethylene glycols on mechanical properties (stress (**a**) and strain (**b**)) of shellac films after storage at 40 °C, 75% RH for 3 months.

Jongjareonrak et al. (*31*), which reported that PEG of a smaller molecular weight was more efficient in interaction with β-lacto-globulin and gelatin molecules. Similarly, Yang and Paulson (*24*) demonstrated that the effectiveness of glycerol in gellan films was most likely due to its smaller size contributing to the readiness of insertion between the polymer chains, and consequently exerting into a larger extent on the mechanical properties of the film than the larger PEG molecules. In addition, at equal concentration, the total number of glycerol molecules in the film-forming solution was greater than that of the higher molecular weight of PEG, and therefore glycerol had more functional groups (−OH) than PEG which could promote the plasticizer−polymer interactions in the films (*32*). The increase in MW of PEG from 200 to 400 gave higher stress due to the effect of higher OH bonding of PEG 400. This could be proved by the higher surface free energy of PEG 400 than PEG 200 indicating the higher hydrophilicity. However, the increase in MW from 400 to 4000 exhibited lower stress due to the steric hindrance effect of the large molecule causing difficulty in hydrogen bond formation and was in accordance with the high insoluble solid for PEG4000. The ability of hydrogen bonding of PEG was dependent on the number of hydroxyl groups (−OH) and molecular size, which was related to the change in stress. Turhan et al. (*9*) reported the increase in hydrogen bond forma-tion between methylcellulose and a different molecular weight of PEG as the molecular weight of PEG increased detected by FTIR spectroscopy. Upon storage, all plasticized shellac films tended to increase stress due to hydroxyl groups (−OH) of plasticizer chains contributing to the development of hydrogen bonds between shellac and plasticizer replacing the polymer to polymer interactions in natural polymer films (*32*). However, the unplasticized shellac film decreased such that the stress could not be detected after 15 days of storage and was in agreement with our previous work (*19*). The stress of plasticized shellac film after 90 days of storage was 3.12, 4.4, and 5.4 MPa for plasticized with 200, 400, and 4000 respectively. The change was statistically significant ($p < 0.01$).

Figure 5b shows the effect of storage time on strain of all shellac films. The higher strain was observed for all plasticized shellac films compared with unplasticized film as a result of hydrophilicity of
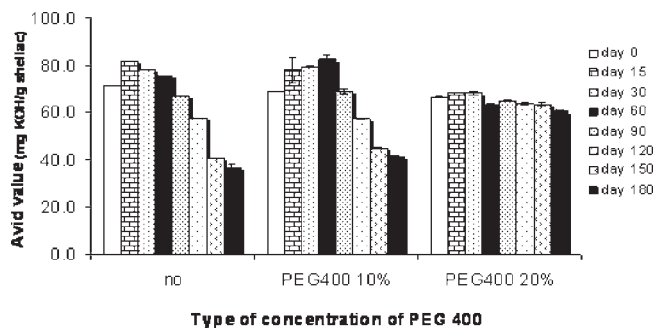
Figure 6. Effect of concentrations of PEG 400 on acid value of shellac films after storage at 40 °C, 75% RH for 6 months.
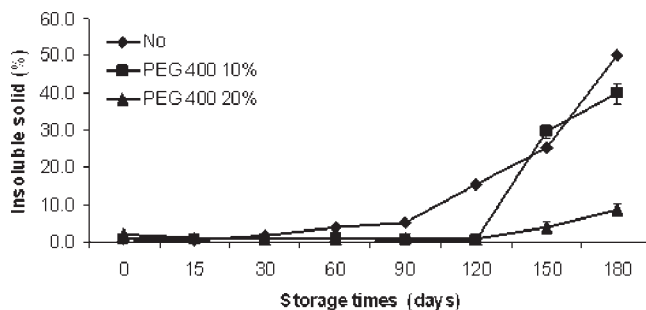
Figure 7. Effect of concentrations of PEG 400 on insoluble solid (%) of shellac films after storage at 40 °C, 75% RH for 6 months.

Table 1. Effect of Concentrations of PEG 400 on WVP Coefficient of Shellac Films after Storage at 40 0C, 75% RH for 6 Months

	water vapor permeability coefficient $\times 10^{-9}$ (g h^{-1} mm^{-1} mmHg^{-1})		
type of film	day 0	day 90	day 180
no plasticizers	5.43 ± 1.66	3.65 ± 1.18	6.51 ± 1.12
PEG 400 10%	6.51 ± 0.89	4.81 ± 0.82	6.31 ± 2.88
PEG 400 20%	25.3 ± 4.45	22.4 ± 2.15	22.3 ± 2.04

PEG. The work was in accordance with Qussi and Suess, which found that various plasticizers, including triethylcitrate and poly-ethylene glycol, could change the strain of shellac film (*15*). Initially, the percentage of strain of unplasticized shellac film and plasticized shellac films with PEG 200, 400, and 4000 were 3.4, 95.5, 44.8 and 20.2%, respectively. The higher the MW, the lower percentage of strain was obtained. The highest percentage of strain of plasticized shellac films with PEG 200 might be a result of a smaller molecule rendering to a higher diffusing and interacting with shellac molecules and hence higher plasticizing efficiency. The result complied with the lowest value of WVPC and was in agreement with Cao et al. (*29*). They found that the PEG of lower molecular weights evidenced the better plasticizing effect for gelatin films and then a better visual property (*29*). The effect of storage time caused reduction of the strain of plasticized shellac film significantly ($p < 0.01$), and the strain after 90 days of storage was 32.8, 4.4 and 1.6% for plasticized with PEG 200, 400 and 4000, respectively. The decrease in strain was a result of the loss of free volume of shellac molecule and was in accordance with the change in other physicochemical properties and agreed well with other studies (*19*, *32*). Luangtana-anan et al.(*19*) reported the decrease in flexibility and increased brittleness upon storage of all shellac films. Heng et al.(*32*) indicated that the change in mechanical properties of ethyl cellulose films was attributed to the loss of plasticizer during the storage.

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Table 2. Effect of Concentrations of PEG 400 on Mechanical Properties of Shellac Films after Storage at 40 °C, 75% RH for 3 Months

storage time (days)	no platicizers		PEG 400 10%		PEG 400 20%	
	stress (MPa)	strain (%)	stress (MPa)	strain (%)	stress (MPa)	strain (%)
day 0	2.48 ± 0.86	3.39 ± 0.84	1.63 ± 0.47	44.79 ± 23.95	0.72 ± 0.27	47.52 ± 9.37
day 15	1.22 ± 0.47	0.54 ± 0.04	4.21 ± 0.77	31.40 ± 11.71	0.90 ± 0.34	71.78 ± 7.62
day 30			3.78 ± 1.07	25.00 ± 13.65	0.91 ± 0.13	74.45 ± 21.48
day 60			2.94 ± 1.13	4.20 ± 2.85	0.71 ± 0.37	73.04 ± 12.19
day 90			4.40 ± 0.85	4.40 ± 2.87	0.58 ± 0.18	53.19 ± 15.91

3.2. Effect of Concentrations of PEG 400 on Physicochemical Properties and Stability of Shellac Film. *3.2.1. Acid Value and Percentage of Insoluble Solid of Shellac Films.* Polyethylene glycol 400 was chosen for the effect of concentration of PEG 400 at the concentration of 10% w/w and 20% w/w. The acid value of unplasticized shellac and plasticized with 10% w/w and 20% w/w PEG 400 after storage for 6 months was shown in Figure.**6**. The acid value of this film was reduced from 71, 68, and 66 to be 36, 41, and 60 for unplasticized and plasticized with 10% w/w and 20% w/w PEG 400, respectively, after 6 month of storage. The significance of change for unplasticized and plasticized with 10% w/w PEG 400 was reported ($p < 0.01$). The reduction of acid value was in agreement with the increase in insoluble solid, indicating the esterification at the carboxylic and hydroxyl groups as displayed in **Figure 7**. The drastic increase in insoluble solid was observed after 4 months and 5 months of storage for unplastized and plasticized with 10% w/w PEG ($p < 0.01$). After 6 months study the insoluble solid was 50, 40 and 8% for unplasticized and plasticized with 10% and 20% w/w respectively. PEG 400 at higher concentration had the lowest insoluble solid. Therefore, it could be proved that PEG400 at 20% w/w could be applied to protect shellac form polymerization.

3.2.2. Water Vapor Permeability Coefficient of Shellac Films. **Table 1** shows the effect of concentration of PEG 400 on the WVPC. The addition of plasticized with 20% w/w of PEG 400 caused higher WVPC whereas unplasticized and plasticized with 10% w/w of PEG 400 had lower value due to the hydrophilicity of the PEG 400 molecule, which is favorable for adsorption of water molecules, enhancing water vapor transmission through the films (*14*). The concentration of PEG 400 at 20% (w/w) exhibited higher moisture content and hence higher WVPC. The result was in agreement with the other studies (*33, 34*). Mali et al. reported the increased hydrophilicity of the starch films at higher glycerol concentration due to more active sites of hydrophilic hydroxyl groups for exposing to the water molecules (*33*). Cuq et al. indicated that an increase in plasticizer concentration resulted in an increase in the WVPC, due to the reorganization of the polymer network, with a consequent increase in the free volume (*34*). The effect of storage time had slight influence on the WVPC for all films after 6 months of storage.

3.2.3. Mechanical Properties of Shellac Film. **Table 2** shows the effect of concentration of PEG 400 on the mechanical properties (stress and strain). As the PEG 400 increased from 10% w/w to 20% w/w, the more incorporation of PEG 400 into shellac network caused the reduction of intermolecular interactions of shellac chains and the increase in mobility of polymer chains resulting in the decreased stress and increased strain with the increasing PEG 400 concentration (*14*). The result was in agreement with Qussi and Suess. They reported an increase in concentrations of PEG on the shellac film leading to an increase in the elongation and flexibility of the film (*15*). The increase in concentration was associated with the increase in the molecular volume and the number of hydroxyl groups leading to the higher accessibility of interaction with the chain of shellac and was in

accordance with the change in mechanical properties and water vapor permeability. The increase in concentration of PEG showed the efficiency for improving the flexibility and decrease in the brittleness upon storage time. After 90 days of storage, the addition of 20% w/w PEG 400 in shellac free film contributed to a slight decrease in stress and increase in strain whereas the slightly increased stress and significantly decreased strain ($p < 0.01$) of the lower concentration of PEG 400 indicating the prolonged stability of shellac by the higher concentration of PEG 400 during storage. The result was in agreement with the other studied parameters. However, the films were still very brittle to perform for 6 months.

3.3. Conclusion. The drawback of the use of shellac as film coating was due to the polymerization. The approach to increase the polymerization by protecting at carboxylic and hydroxyl groups of shellac chain was by the application of plasticizer. The protecting ability of plasticizer at active sites of shellac was a result of suitable molecular weight and concentration of plasticizer, acting by the formation of hydrogen bonding between the active sites of shellac and hydroxyl group of PEG. PEG 400 at the concentration of 20% w/w was the best plasticizer among the studied PEG. The stabilized shellac was reported with the minimum change of all physicochemical properties such as acid value, insoluble solid, mechanical properties and water vapor permeability coefficient. To choose the appropriate plasticizer and concentration is the requirement for the film coating solution to not only increase the flexibility but also enhance the stability. Therefore, this work could contribute to the wider application of shellac for the film coating protection of food and pharmaceutical products in food and pharmaceutical industries.

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