

Effect of Salts and Plasticizers on Stability of Shellac Film

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The aim of this study was to increase the stability of shellac because of the polymerization. A few approaches have been applied in this study. Shellac film was prepared in two salt forms, that is, ammonium and 2-methyl-2-amino 1-propanol salts, and a comparison was made with shellac film in free acid form. The other approach was by the application of plasticizers. These plasticizers were diethyl phthalate, triacetin, and polyethylene glycol 400 (PEG 400). Plasticized shellac and unplasticized shellac films in free acid form were then compared. All shellac films were kept in stability chamber at 40 °C, 75% RH for a period of 3 months. The studied parameters such as insoluble solid, acid value, mechanical properties, and water vapor permeability were detected every month. Analysis of variance (ANOVA) technique was used to analyze data. The applications of salt forms proved statistically significant ($p < 0.01$) to reduce the polymerization process whereas certain plasticizers could enhance the stability. PEG 400 was the only plasticizer that could show the increase in stability. The improvement of stability might be a result of the interference of a larger molecule of PEG 400 causing the difficulty in interaction among carboxyl or hydroxyl groups of shellac and the effect of lower loss of plasticizer.

KEYWORDS: Shellac; plasticizer; salt; stability

INTRODUCTION

Shellac is a natural polymer of animal origin secreted by the lac insect *Kerria lacca*, which parasitically grows on some specific types of trees in China, India, and Thailand. In Thailand, shellac can be found on *Samanea saman* and *Leucaena leucocephala* trees, family of Leguminosae. Shellac is composed of hard resin and soft resin of polyester and single ester with polyhydroxypolybasic acids such as aleuritic acid, jalaric acid, and laccijalaric acid as shown in **Figure 1** (1, 2). It is insoluble in water but soluble in alcohol and alkaline solution and possesses a very low water and acid permeability (3). The pH solubility profile of shellac was reported in our previous study and the profile could be modified by hydrolysis process (4). The ionization constant (pK_a) was found to be 6.7 (4). We found that shellac started to dissolve at pH above 7; therefore,

it has been widely used in pharmaceutical industry for enteric coating to protect from acid in gastrointestinal tract (5). In addition, because of the extremely low water permeability, it has been commonly used as a sealing coat in sugar-coated tablets and in food and wood industries. The water permeability was found in our study to be about $4 \times 10^{-9} \text{ g h}^{-1} \text{ mm}^{-1} \text{ mmHg}^{-1}$, whereas for other enteric-coated polymers was about $3 \times 10^{-4} \text{ g h}^{-1} \text{ mm}^{-1} \text{ mmHg}^{-1}$ (6). However, the application of shellac was gradually reduced as a result of its instability, organic solvent required, and the substitution of semisynthesis and synthesis polymers. Luce (7) found that tablets coated with shellac did not disintegrate upon storage for only 1 year. The instability might be the result of the polymerization (8). Several approaches had been made to improve shellac stability and aqueous solubility. These approaches were the preparation in pseudo latex aqueous solution (9, 10), the modification into shellac derivatives (11, 12), and the preparation of shellac in ammonium salt form (4, 8, 13). Of all these attempts, the use

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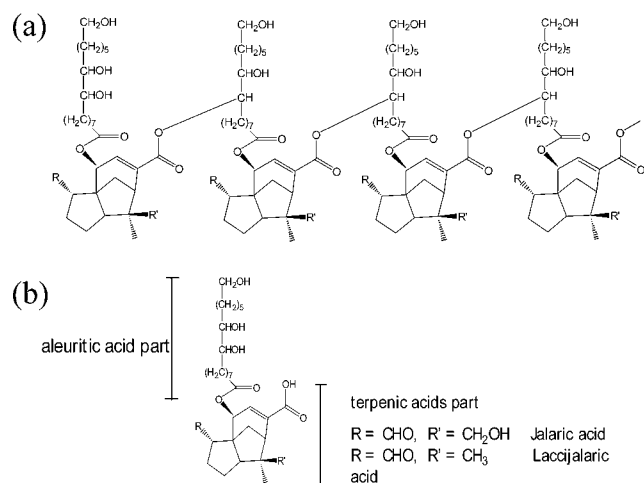


Figure 1. Chemical structures of polyesters (a) and single esters (b) of shellac.

of ammonium salt could improve the problem of stability to some extent, however, further investigation is needed to verify the application of salt form.

Plasticizers played an important role on film coating (14). Type and amount of plasticizers affected some properties of films. The increasing of plasticizer generally decreased tensile strength and increased percent elongation of films. Poly(methacrylic acid, methylmethacrylate 1:1, Eudragit L) films containing various plasticizers showed strong correlation between thermal properties and mechanical properties (15). Water vapor permeability of film was changed by the addition of plasticizer. Polyethylene glycol increased water vapor permeability of hydroxypropyl methylcellulose (HPMC) film whereas castor oil and dibutyl phthalate reduced water vapor permeability. In some cases, water vapor permeability did not change by the addition of plasticizers (16). This could be the result of the solubility and the affinity of plasticizer to the polymer. The mechanism of release of theophylline coated products was also affected by the addition of plasticizer (17).

Therefore, the objective of this study was to investigate the effect of salts and plasticizers on stability of shellac. Shellac films prepared in two salt forms, that is, ammonium and 2-methyl-2-amino 1-propanol (MAP), were compared with free acid form. Shellac prepared in free acid form with various plasticizers was comparatively evaluated. The evaluation parameters were acid value, percent insoluble solid, and other film properties such as water vapor permeability and mechanical properties.

MATERIALS AND METHODS

Materials. Shellac was kindly supplied from Union Shellac Part., Ltd. (Bangkok, Thailand). The solvents used were ethyl alcohol and ammonium hydroxide obtained from Merck, Germany and 2-methyl-2-amino 1-propanol (MAP) from Fluka, Switzerland. Plasticizers used were diethyl phthalate (DEP) from the British drug house (BDH) chemicals, United Kingdom; triacetin (TA) was from Eastman Chemical company, United States; and polyethylene glycol 400 was purchased from Dow chemical company, United States. All the reagents were of reagent grade. The chemical structure of these plasticizers is shown in Table 1.

Methods. Preparation of Shellac Films. For unplasticized shellac, films were prepared to investigate the effect of two salts, that is, ammonium hydroxide and 2-methyl-2-amino 1-propanol (MAP), in comparison with free acid. To prepare shellac in free acid form, 12% w/w shellac was dissolved in 95% ethyl alcohol while 12% w/w shellac was dissolved in ammonium solution and MAP solution to prepare

Table 1. Some Properties of Plasticizers and Shellac Films

Type	Formula	MW (g/mol)	Boiling point (°C)	% water of film ^(a)
TA		218.2	259	9.80
DEP		222.2	298	9.54
PEG		400.0	-	10.82
NO				8.58

^a Measured by Karl Fisher. NO, unplasticized shellac; TA, triacetin; DEP, diethyl phthalate; PEG, polyethylene glycol 400.

shellac in ammonium and MAP salt forms, respectively. The amount of ammonium hydroxide and MAP solutions were calculated on the basis of acid value of shellac, that is, the amount of salt which is used to interact with acid group of shellac. The effect of plasticizers on stability of shellac film in free acid form was also investigated using three plasticizers, diethyl phthalate (DEP), triacetin (TA), and polyethylene glycol 400 (PEG). For plasticized film, 12% w/w shellac was dissolved in 95% ethyl alcohol and 10% w/w plasticizer of dry shellac weight was added in shellac solution. All accurately weighed shellac solutions were then poured on polytetrafluoroethylene plate and were allowed to dry at 50 °C for 2–3 h. All the films were then peeled off the polytetrafluoroethylene plate. The stability study was then carried out by storing the shellac films in a stability chamber at 40 °C, 75% RH for 3 months to determine for acid value, percent insoluble solid, mechanical properties, and water vapor permeability coefficient (WVPC). Shellac was then evaluated at 0, 15, 30, 60, and 90 days. Prior to evaluation, the films were kept in a dried desiccator for 1 h to stabilize all samples.

Determination of Acid Value. The acid value of shellac was measured by titration according to the method described in the USP XXIII (18). The end point was determined by potentiometric titration instead of by using color indicator because of the dark color of shellac. The change of this value was recorded at 0, 15, 30, 60, and 90 days. The acid value was expressed as milligram of potassium hydroxide per 1 gram of shellac. The average of two measurements was performed.

Determination of Percent Insoluble Solid. Two grams of shellac sample was dissolved in 95% ethyl alcohol for 3 h. The solution was then filtered and weighed. The percent insoluble solid was dried until constant weight was obtained (about 50 °C, 12 h). The percent insoluble solid was measured every month for 3 months. The insoluble solid of two measurements was done.

Determination of Water Vapor Permeability Coefficient (WVPC). An adapted permeation cell for water vapor permeation study was used according to ASTM method (19). The method was using a glass bottle that was filled with dried granular calcium chloride and had a cap with an open circular hole of 3–4 cm in diameter. The shellac film with an exposed area of 800–900 mm² was placed inside the cap and was sealed tightly. The cell was then kept in a stability cabinet at 40 °C, 75% RH. The weight gained of water vapor was recorded every day and was

calculated for water vapor permeability coefficient (WVPC) value every month for 3 months. The WVPC of 10 cells for all films was, then, calculated as the following:

$$\text{WVPC} = (W \times t) / (A \times \Delta p)$$

where

WVPC = water vapor permeability coefficient ($\text{g h}^{-1} \text{mm}^{-1} \text{mmHg}^{-1}$),

W = amount of moisture transmitted per unit time in grams per hour

(from the slope of a plot of moisture permeation versus time, g/h),

t = thickness of the film (mm),

Δp = vapor pressure gradient across film (mmHg), and

A = exposed area of film (mm^2).

Determination of Stress and Strain at Break. Stress and strain at break were carried out by texture analyzer model TA.XT plus (Stable Micro Systems Ltd., United Kingdom). The prepared films were cut in a dumbbell shape with length of 25 mm and width of 6.045 mm. The thickness of film was measured by a micrometer. The test speed was 5 mm/min. The stress and strain at break of 10 samples were measured every month for a period of 3 months.

Scanning Electron Microscopy (SEM). Micrographs of the samples were obtained using scanning electron microscope (SEM, model MX 2000, Cam Scan, Cambridge, United Kingdom). The samples were coated with a fine gold layer before obtaining the micrographs. The accelerating voltage was used at 10 kV.

Differential Scanning Calorimetry (DSC). DSC thermograms of shellac films were measured by using a differential scanning calorimeter (DSC 7, Perkin-Elmer, United States). The samples of 2–4 mg were accurately weighed into solid aluminum pans and were sealed. The measurement was obtained at a heating rate of 10 °C/min under nitrogen purge.

Statistical Analysis. Data was expressed as the mean \pm standard deviation (SD). The statistical analysis was carried out using analysis of variance (ANOVA) at the 0.01 significant level.

RESULTS AND DISCUSSION

Shellac, a mixture of polyesters and single esters, comprises several hydroxyl groups and carboxyl groups as shown in **Figure 1**. The aging of shellac was reported because of the polymerization among these groups, resulting in the instability of shellac (13). The instability caused the changes in various properties such as acid value, insoluble solid, WVPC, and mechanical properties. Therefore, these parameters were used to monitor the stability of shellac.

From our previous study, the stability of shellac could be improved when shellac was in ammonium salt form which was a result of the protection of ammonium at the carboxylic site of shellac group, resulting in less polymerization (13). To verify the effect of salt on the stability and to prepare into aqueous system, MAP salt was used. The comparison was then made among free acid, ammonium, and MAP salts as shown in **Figures 2–5**.

The percent insoluble solid of unplasticized shellac film in free acid form was increased statistically significantly ($p < 0.01$) after 3 months of storage whereas shellac in both salt forms changed slightly as shown in **Figure 2**. Insoluble solid of free acid at 3 months of storage was 8% while of ammonium and MAP salts were 1.2% and less than 1%, respectively, suggesting the higher polymerization upon storage for shellac in free acid form. **Figure 3** shows effect of storage time on acid value. It changed slightly and was not in accordance with the change of insoluble solid. The slight change of acid value was due to the large numbers of acid groups such that the change of insoluble solid could not cause a great change on acid value. In addition, shellac is composed of hard resin of polyester which has a lower acid value whereas the soft resin of single resin has a higher

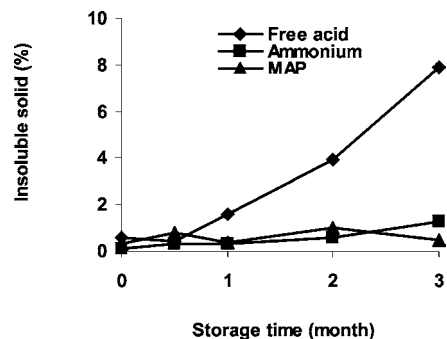


Figure 2. Effect of storage time on insoluble solid (%) of unplasticized shellac films in free acid, ammonium, and MAP forms (storage at 40 °C, 75% RH).

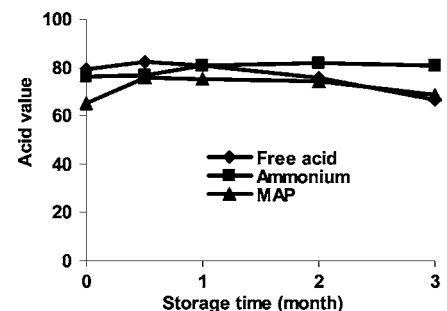


Figure 3. Effect of storage time on acid value (mg KOH/g shellac) of unplasticized shellac films in free acid, ammonium, and MAP forms (storage at 40 °C, 75% RH).

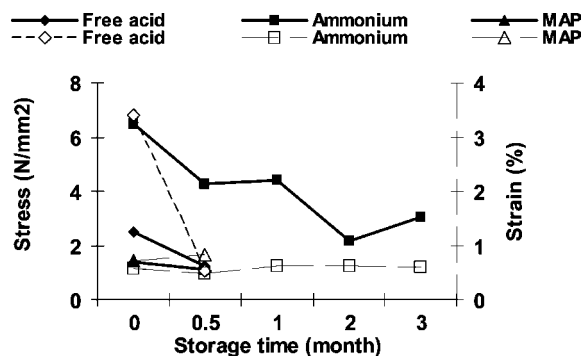


Figure 4. Effect of storage time on stress (dark symbol) and strain (blank symbol) of unplasticized shellac films in free acid, ammonium, and MAP forms (storage at 40 °C, 75% RH).

acid value. The acid value of shellac is the average value of polyester and single ester (20). The polyester part tended to polymerize first and to form insoluble solid so that the average acid value changed slightly during the first 3 months. However, further study of free acid film has been carried out for 4 months. The insoluble solid of free acid was increased from 8% to 15% and acid value was also further decreased from 67 to 58 (initial was 80; data at 4 months of storage was not shown in **Figures 2 and 3**). The storage effect on mechanical properties was shown in **Figure 4**. Shellac in ammonium salt had a higher stress whereas shellac in MAP salt and free acid had a lower stress. All shellacs tended to decrease in mechanical properties upon storage. The change was statistically significant at 15 days of storage such that they could not be detected for shellac in free acid and MAP ($p < 0.01$). The loss in mechanical properties on storage was in agreement with other reports (21). **Figure 5** shows the change in WVPC. Initially, the higher WVPC was

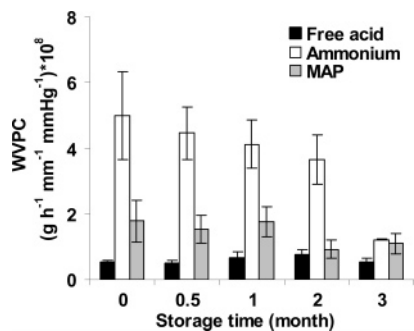


Figure 5. Effect of storage time on WVPC of unplastized shellac films in free acid, ammonium, and MAP forms (storage at 40 °C, 75% RH).

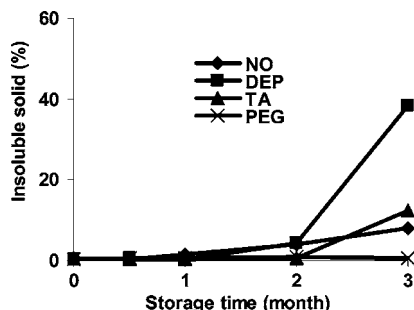


Figure 6. Effect of storage time on insoluble solid of unplastized (NO) and plasticized shellac films (DEP, TA, PEG) in free acid form (storage at 40 °C, 75% RH).

observed for shellac in salt forms because of their water solubility. WVPC decreased after 3 months of storage for shellac in salt forms whereas shellac in acid form changed slightly. The WVPC decreased because of the loss of water, resulting in the tightness of shellac molecules, enabling the difficulty in the permeation of moisture. Therefore, the modification of shellac by the application of salt form could prove to enhance the stability of shellac indicated by the slight change in % insoluble solid. Shellac in ammonium salt film gave a stronger film than MAP and free acid film. All shellacs still possessed the high water permeation protection during 3 months of storage.

Further study was on the application of plasticizer on the stability of shellac. The percent insoluble solid of unplastized film increased as prolongation of storage time, confirming the instability because of polymerization as shown in **Figure 6**. The addition of plasticizers had different effects on the stability of shellac. The pronounced change ($p < 0.01$) in insoluble solid was found for plasticized shellac with DEP and TA whereas the addition of PEG 400 gave the highest stability. The % insoluble solid of unplastized shellac (indicated by NO) and plasticized shellac with DEP, TA, and PEG at 3 months of storage was 8%, 40%, 12%, and 0.5%, respectively. The high stability of shellac plasticized with PEG 400 might be a result of the interference of a larger molecule of PEG 400 causing the difficulty in interaction among the active groups of shellac enabling the less polymerization. In addition, the amount of PEG 400 might still be retained in shellac as compared with other plasticizers because of the higher molecular weight and less volatilization. The pronounced loss of DEP and TA from poly-(butyl methacrylate, 2-dimethyl aminoethylmethacrylate, and methylmethacrylate, 1:2:1) (Eudragit E) film was also reported under the longer storage (22). The loss of plasticizer was by three mechanisms: volatilization (loss on surface bound by air), migration (loss on surface in intimate contact with a solid), and extraction (loss on surface bound by liquid) (23). In the present study, the loss might be mainly attributed to the volatilization

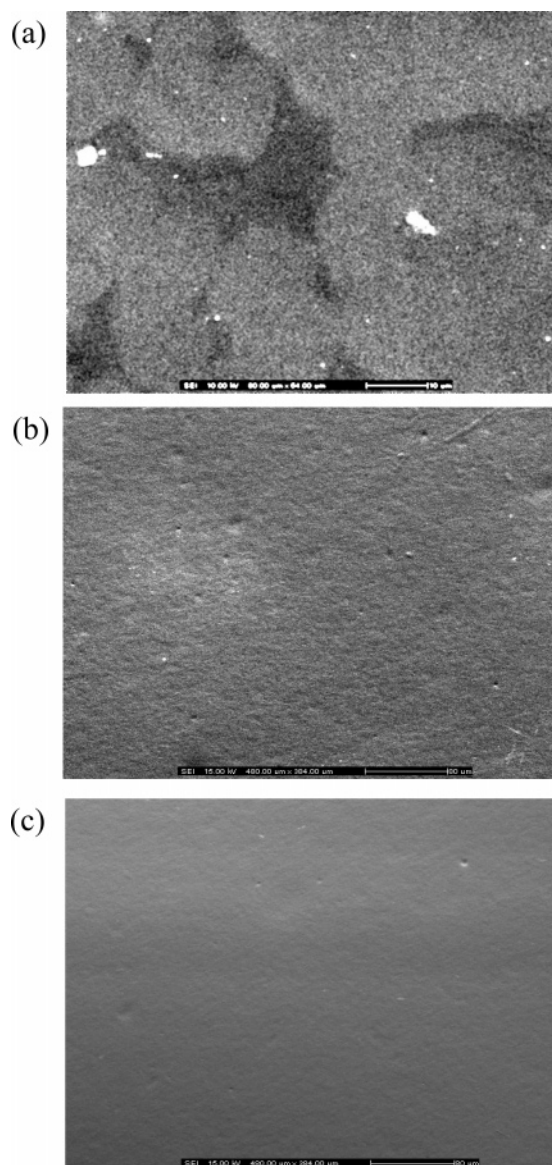


Figure 7. Scanning electronic micrographs of plasticized shellac film with DEP (a), TA (b), and PEG (c).

because of the lower molecular weight and lower boiling point (6, 22, 24, 25). The molecular weight and boiling point of plasticizers are shown in **Table 1**. The highest increase of insoluble solid at 3 months ($p < 0.01$) of DEP (about 40%) might be a result of the inhomogeneity of DEP within the shellac network because of the low affinity of DEP with shellac as shown in **Figure 7**, indicating the poor protection of DEP at the active sites of shellac and, hence, a high polymerization. The result was in agreement with the work of Lecomte et al. (26) which found that the affinity of polymer film with plasticizer played an important role on film property. Upon longer storage, the loss of DEP might occur in a greater extent because of both mechanisms, that is, volatilization and extraction since DEP is soluble in alcohol. The decrease in acid value was also in accordance with the increase in % insoluble solid as shown in **Figure 8**. The change of acid value and insoluble solid could be due to the loss of plasticizers indicating the polymerization of shellac film. The result confirmed that PEG 400 could stabilize shellac.

Initially, the addition of plasticizers attributed to the slight decrease in stress and to the significant increase in flexibility of film ($p < 0.01$), and thus mechanical properties could be

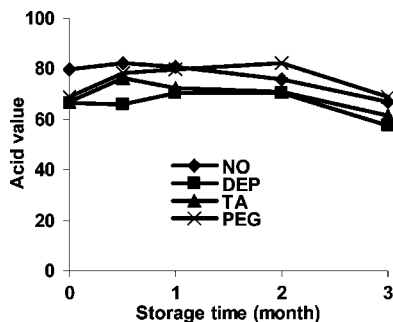


Figure 8. Effect of storage time on acid value (mg KOH/g shellac) of unplastized (NO) and plasticized shellac films (DEP, TA, PEG) in free acid form (storage at 40 °C, 75% RH).

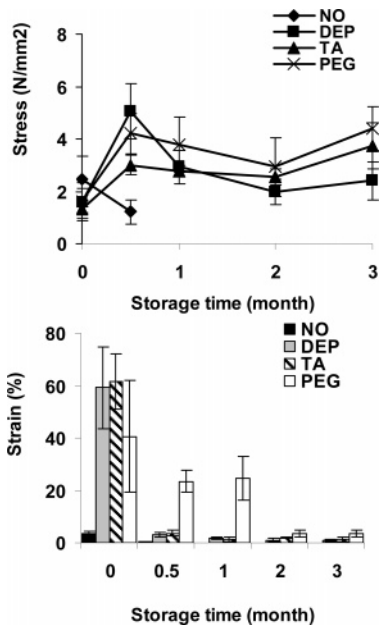


Figure 9. Effect of storage time on stress and strain of unplastized (NO) and plasticized shellac films (DEP, TA, PEG) in free acid form (storage at 40 °C, 75% RH).

detected after 15 days of storage in comparison to the unplastized shellac as shown in **Figure 9**. The decrease in stress could be the effect of the reduction of intermolecular attraction within the shellac network whereas the increase in flexibility was a result of the increase in free volume because of plasticizer (27). However, the addition of plasticizer did not lead to a change in softening temperature as shown in **Figure 10**. All plasticized shellac and unplastized shellac showed the softening temperature at 54 °C. This could suggest that plasticizer did not have the interaction with polymer chain. The increase in strain could be the result of the plasticized effect of water content as indicated in **Table 1** causing the increase in free volume. The effect of water as plasticizer was reported in another study (28). The increase in stress at 15 days of storage was not the effect of the change in crystallinity of shellac as all plasticized and unplastized shellacs were in amorphous state (data of powder X-ray diffractometry was not shown). The increase in stress might attribute to the closer intermolecular bonds of shellac network because of the loss in water content. Upon longer storage, the mechanical properties of all types of shellac films decreased, and the change was higher for plasticized shellac with DEP and TA. The change of mechanical properties was in agreement with the change in acid value and insoluble solid confirming the loss in plasticizer during storage resulting in the lower free volume and less flexibility of film.

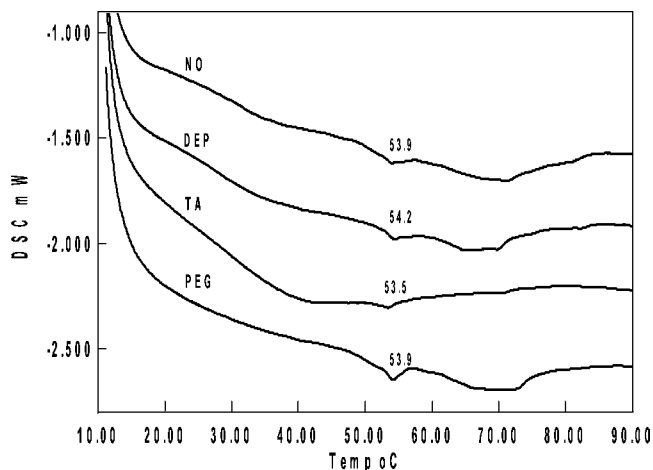


Figure 10. Effect of plasticizer on softening temperature of shellac films in free acid form with plasticizer (DEP, TA, PEG) and without plasticizer (NO).

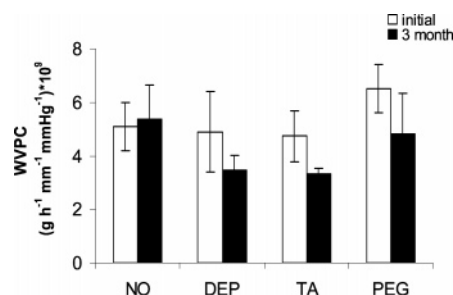


Figure 11. Effect of storage time on WVPC of unplastized (NO) and plasticized shellac films (DEP, TA, PEG) in acid form (storage at 40 °C, 75% RH).

The highest increase of insoluble solid for DEP did not result in a stronger polymer film as the strength of the film was from the interaction of the polymer chain network. Heng et al. (6) found that the change in mechanical properties of ethylcellulose films on storage was mainly attributed to the loss of plasticizer during storage. **Figure 11** shows the effect of aging on WVPC. Initially, the WVPC of unplastized and plasticized shellac with DEP and TA films was relatively the same value in the range of $4-5 \times 10^{-9} \text{ g h}^{-1} \text{ mm}^{-1} \text{ mmHg}^{-1}$ while that of PEG 400 plasticized shellac films was higher. The result suggested that the plasticizer had a different effect on WVPC of shellac films. In case of PEG 400, the high WVPC of film should be due to higher water solubility or to hydrophilic nature of plasticizer and was in accordance with the water content of shellac film indicated in **Table 1**. During storage, the WVPC decreased for all shellacs as a result of the loss in free volume causing the tightness of shellac film and enabling the difficulty in the permeation of moisture. All the films exhibited the high-moisture protection on storage which is the main advantage of shellac film. The addition of plasticizer, hence, had different effects on the stability of shellac. PEG 400 demonstrated the lowest change of all parameters as compared with other plasticizers, suggesting the most suitable plasticizer for stabilization of shellac films in this study. Therefore, the means to reduce the polymerization process could be applied by means of modification shellac in salt form or by the addition of proper plasticizer.

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