อิทธิพลของอุณหภูมิและตัวทำละลายที่ใช้ในการขึ้นรูปต่อสมบัติ ของฟิล์มจากแป้งและสตาร์ชลูกเดือย Effect of Casting Temperatures and Solvents on Adlay Flour and Starch Film Properties

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บทคัดย่อ

สมบัติของฟิล์มจากแป้งลูกเดือยถูกกำหนดโดย 2 องก์ประกอบหลักคือ โปรตีนและสตาร์ช โดยวัตถุประสงก์ของงานวิจัย เพื่อศึกษาผลของอุณหภูมิและสารละลายที่แตกต่างกันในการขึ้นรูปฟิล์ม ต่อสมบัติของฟิล์มจากลูกเดือย ในงานวิจัยได้เตรียมสารละลายฟิล์มที่อุณหภูมิแตกต่างกัน (70 และ 80 องศาเซลเซียส) และความเข้มข้นของสารละลายเอทานอลที่ต่างกัน (0, 30 และ 40% v/v) พบว่าความ เข้มข้นของเอทานอลมีผลต่อ ความขุ่น ความหนา และความหนาแน่นของฟิล์มจากแป้งลูกเดือย (p<0.05) จากการทดลองพบว่า ที่อุณหภูมิ 70 องศาเซลเซียส ฟิล์มจากแป้งที่ถูกเตรียมค้วยน้ำกลั่น (เอทานอล 0%) มีแรงด้านทานการดึงยืดต่ำกว่าฟิล์มจากสตาร์ชในขณะที่การเตรียมฟิล์มที่อุณหภูมิ 80 องศาเซลเซียส มีผลทำให้แรงด้านการดึงยืดต่ำกว่าฟิล์มจากแป้งมีค่าสูงกว่าฟิล์มจากสตาร์ช (p≥0.05) อย่างไรก็ตามค่าการ ยึดตัวของฟิล์มจากสตาร์ชที่เตรียมค้วยอุณหภูมิที่แตกต่างกัน มีก่าสูงกว่าฟิล์มจากแป้ง เมื่อตรวจสอบ โครงสร้างของฟิล์มจากสตาร์ชที่เตรียมค้วยอุณหภูมิที่แตกต่างกัน มีก่าสูงกว่าฟิล์มจากแป้ง เมื่อตรวจสอบ โครงสร้างของฟิล์มจากสตาร์ชที่เตรียมค้วยอุณหภูมิที่แตกต่างกัน มีก่าสูงกว่าฟิล์มจากแป้ง เมื่อตรวจสอบ โครงสร้างของฟิล์มจ้ายกล้องจุลทรรศน์อิเล็กตรอนแบบส่องกราด พบว่าผิวหน้าฟิล์มที่เตรียมด้วย เอทานอล 30% ที่อุณหภูมิ 80 องศาเซลเซียส มีการกระจายตัวของสตาร์ชและโปรตีนทั่วทั่งโลรงสร้าง ฟิล์มมากกว่าฟิล์มที่เตรียมด้วยเอทานอล 0 และ 40% การศึกษานี้แสดงให้เห็นว่า ความแตกต่างของ อุณหภูมิและความเข้มข้นของเอทานอลในการขึ้นรูปฟิล์มมีผลต่อโครงสร้างฟิล์ม ซึ่งส่งผลต่อสมบัติ ด้านกายภาพและสมบัติเชิงกลของฟิล์มจากลูกเดือย

คำสำคัญ: แป้งลูกเดือย, สตาร์ชลูกเดือย, ฟิล์ม, สมบัติด้านกายภาพ, สมบัติเชิงกล, สมบัติเชิง โครงสร้าง

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ABSTRACT

The properties of adlay flour based films could be influenced by their two major components - protein and starch. This research aimed at studying the effect of different temperatures and casting solvents on adlay film properties. By using different casting temperatures (70 and 80 °C) and different ethanol concentrations (0, 30 and 40% v/v), it was found that the ethanol concentrations had significant effect on the adlay flour film opacity, thickness and density. The stress value (MPa) of flour films, prepared by using distilled water (0%EtOH) at 70°C, was lower than that of starch films; whereas, the stress value of flour films prepared at 80 °C was higher than that of starch films ($p \ge 0.05$). Nevertheless, the strain values of starch films, prepared with different casting temperatures were higher than those of flour films. Moreover, the scanning electron micrographs showed that the surface of flour films prepared from aqueous ethanol was rougher than that of flour film prepared from distilled water. Furthermore, the internal structure of the adlay flour film prepared by 30% EtOH at 80°C had better dispersion and miscibility of starch and protein components throughout the film structure than those of film prepared by 0 and 40% EtOH. This study suggested that the casting temperatures and ethanol concentrations had affected the film structure, leading to the change of the physical and mechanical properties of adlay flour film.

Key words: Adlay flour, Adlaystarch, film, physical properties, mechanical properties, structural properties

INTRODUCTION

Adlay or Job's tears (*Coix lachryma-jobi* L.) is a cereal crop, which grows in Asian countries. It is mainly composed of starch and protein (mainly prolamin, known as coixin). Coixin protein is an alcohol soluble protein, which has molecular weight pattern similar to zein and kafirin protein from corn and sorghum respectively (Shewry and Tatham, 1990). The molecular weight of coixin protein is composed of 22 to 23 kDa, 16 to 19 kDa and 27 to 28 kDa which were classified into alpha (α -coixin), beta (β -coixin) and gamma (γ -coixin) coixin respectively (Dechkunchon, 2008). In addition, adlay starch had amylose content of around 29% (Dechkunchon, 2008). In the recent year, starch and protein were mostly used as biomaterials due to their being inexpensive, widely available and renewable and ease of handling. Generally, the starch based film formation was focused on the amylose network, due to the fact that it gives the stronger film; while the amylopectin decreases the tensile stress (Alves *et al.*, 2007). Furthermore, from the previous study, the temperature and time during film preparation could also affect the leaching of amylose and the presence of remnants or ghost granules leading to the change of structural and mechanical properties of the film (Paes *et al.*, 2008; Koch *et al.*, 2010). However, the disadvantage of starch film is its sensitivity to humidity because of its hydrophilic character, resulting in the loss of physical, mechanical and barrier properties (Zhang and Han, 2006). For the protein film, especially prolamin film, it showed good water resistant properties as compared to the other protein films due to its hydrophobic character and good tensile strength (Parris et al., 1997; Belton et al., 2006; Wang et al., 2008; Shi et al., 2009). In general, the preparation of prolamin film requires dispersion or solubilization of protein in aqueous alcohol solutions. There is research studied the effect of casting solvents on zein film properties and found that the casting solvents had affected the rearrangements of the amino acid residues in zein film formation through the development of hydrophobic interaction, hydrogen bonds and limited disulfide bonds between zein chains (Guo et al., 2005; Chen et al., 2014).

As mentioned above, both starch and protein have advantages and disadvantages on film properties; therefore, adlay flour which is natural blends of starch and protein is an interesting alternative material for preparing the film. Because adlay flour consists of starch (hydrophilic) and protein (major hydrophobic), it has the advantage of forming film, and at the same time it had been used as food and medicine. The aims of this research were to prepare films based on adlay starch and flour and to study the effect of casting temperatures and solvents on physical and mechanical properties of these films.

MATERIALS AND METHODS

Materials

Adlay grains were obtained from Loie

province in Thailand in May, 2010. Ethyl alcohol (99.5% purity) was purchased from Macron chemicals (Malaysia). Glycerol (99.5% purity) was purchased from Carlo Erba Reagents (France).

Methods

Adlay flour and adlay starch preparation

Adlay flour was prepared by dry milling method prior to screening through a 100-mesh sieve. Adlay starch was isolated by being modified from Sira and Amaiz (2004) method. Adlay grains were soaked in 0.25% (w/v) NaOH at 25°C for 18 h. Then the grains were drained and washed by distilled water until they reached neutral pH. Consequently, the grains were ground and screened through a set of various sieves (80, 100, 200 and 270 mesh), respectively. After that the filtrate was centrifuged by a refrigerated centrifuge at 460 x g for 20 min. The brown layer was then removed by scraping and then the white layer was resuspended in distilled water before repeating the centrifugation and scraping steps several times until the starch slurry became white. Then the starch slurry was dried by circulated hot air oven at 45°C for 18 h, until the moisture content of starch was less than 12% (w/w).

Characterization of adlay flour and adlay starch Chemical properties

The protein, lipid, ash, crude fiber and moisture content of both starch and flour were determined by following AOAC (2000) methods.

Amylose content

The amylose content of adlay starch was

determined by following Juliano (1971) method. Adlay starch was weighed and then 1 ml of absolute ethanol and 9 ml of sodium hydroxide solution (0.1 N) were added. The sample was then heated in boiling water for 10 min before adjusting the volume to 100 ml with distilled water and adding 1 ml of acetic acid solution (1 N) and 2 ml of iodine solution (0.2 g iodine and 2.0 g potassium iodide in 100 ml of distilled water. Then the absorbance of sample was measured by spectrophotometer at 620 nm and compared with the amylose content from the standard curve of amylose type III from potato (Sigma-Aldrich, Germany).

Thermal property

The gelatinization temperature and the enthalpy change of adlay starch and flour were determined by differential scanning calorimeter (DSC; model Pyris, Perkin-Elmer, USA) by following Dechkunchon (2008) with slightmodification. A sample was weighed in the stainless steel pan and then distilled water was added to get 85% (w/w) moisture content prior to being sealed and equilibrated at 25°C for 18 h. After that, the sample was heated at a rate of 5°C/min from 25 to 130°C. The gelatinization temperatures were reported in terms of the onset (To), peak (Tp) and conclusion (Tc). The enthalpy of gelatinization (Δ H) was calculated as Joules per gram of dry sample (J/g).

Adlay flour and adlay starch film preparation

Adlay flour and starch films were prepared by casting method, both the adlay flour and starch film solutions containing 4% (w/w) of starch. The adlay flour and starch were dispersed in distilled water or aqueous ethanol solutions (30 and 40% (v/v)). Subsequently, the suspensions were heated to 70 or 80°C and maintained at that temperature for 15 min. Then glycerol (0.5 g/g dry starch) was mixed into the suspensions and the film solution was then degassed prior to being poured onto polystyrene Petri dish and dried at 35°C in a hot air oven. Before the measurements of film properties, the film samples were equilibrated in a desiccators containing saturated magnesium nitrate salt solution (RH around 54%) for 48 h.

Adlay flour and adlay starch film properties

Physical and chemical properties

Appearance of adlay flour and starch films

Adlay flour and starch films were visually observed for their appearances: clarity, color, and surface texture. The picture of films was taken by a digital camera.

Thickness of film

The thickness of films was measured by a manual micrometer (Mitutoyo, Kanagawa, Japan) at 10 random positions on the films.

Density of film

The samples were cut into 2×2 cm and then the film thickness was measured before drying at 105° C as described later. The density of film was calculated by the following equations (Dias *et al.*, 2010).

density(
$$\rho^{s}$$
) = $\frac{m}{A \times \delta}$ (1)

Where m is the mass (g) of dried film, A is the area (4 cm²) of film specimen and δ is the thickness (cm) of film.

Moisture content of film

The moisture content of film samples was determined by slight modification of Zhang and Han (2006) method. The film samples (2×2 cm) were placed into dried aluminum can and dried at $105\pm2^{\circ}$ C by circulating hot air oven for 24 h. Then the dried samples were cooled to room temperature in desiccators. The moisture content (%MC) was calculated by the following equations.

moisture content (%MC) =
$$\frac{\text{Mi} - \text{Md}}{\text{Mi} - \text{Mf}} \times 100$$

Where M_i , M_d and M_f are the weight of aluminum can with a film sample before drying, aluminum can with a dried film sample and empty aluminum can, respectively.

Tensile properties

Tensile strength (Stress) and elongation (Strain) were measured with a texture analyzer (TA.XT plus, Stable Micro Systems, UK). Tensile tests were performed according to ASTM Standard Method D 882-91 (ASTM, 1994). Film samples were cut into rectangular strips (25×100 mm) and conditioned in saturated magnesium nitrate salt solution at 25 °C for 48 h. before testing. The tensile tests were measured until the break of sample by using tensile grips (A/TG). Tension force and deformation distance were recorded during extension at 5 mm min⁻¹ and distance between grips of 50 mm. The stress and strain values were calculated by the following equations (modified from Tang *et al.*, 1994).

Stress
$$(\sigma_{\text{truemax}}) = \frac{\text{Fmax}(L + \Delta \text{Lmax})}{\text{AL}}$$
 (2)

Strain(
$$\varepsilon_{\text{truemax}}$$
) = ln $\left[1 + \frac{\Delta L \max}{L}\right]$ (3)

Where Fmax is the tension force (N) at the maximum force, Δ Lmax is the film deformation distance (m), A is the area of the specimen (0.025 m × Thickness (m)) and L is the original length of the test film (0.05 m), respectively.

Microstructure of adlay flour and adlay starch films

Morphology of adlay flour and starch films at a cross section and surface was observed by scanning electron microscope (SEM) (JSM-6480LV, JEOL, Japan) with an accelerating voltage of 15 kV. The adlay films were dried by freeze dryer before attaching on the stub and coated with gold by sputter coater.

Statistical analysis

All measurements were analyzed by using a SPSS software version 12. Duncan new's multiple range test was used to compare mean difference (p < 0.05).

RESULTS AND DISCUSSION

Chemical compositions and thermal properties

The chemical compositions of adlay flour and starch are shown in Table 1. The results showed that the major chemical compositions of adlay flour were mainly composed of carbohydrate (80.06%) and protein (14.17%). From the previous study, the major protein of adlay flour is prolamin or coixin, alcohol soluble protein (Dechkunchon, 2008). In addition, the minor compositions of adlay flour are lipid, fiber and ash as shown in Table 1. On the other hand, the major chemical composition of adlay starch was carbohydrate (99.12%) and its amylose content was about 26.77% (Table 1).

The thermal properties of both adlay flour and starch determined by DSC are shown in Table 1. The result shows that the gelatinization temperature of adlay flour was about 64.75 - 69.78 - 73.67°C (To-Tp-Tc); while that of adlay starch was 65.00-68.94-73.00°C (To-Tp-Tc), respectively. The enthalpy of adlay starch was 15.24 J/g starch and significantly higher than that of adlay flour (4.02 J/g flour) because of the higher starch content in the starch sample, led to require more energy to disrupt the starch crystalline in gelatinization process (Uarrota *et al.*, 2013).

The chemical compositions and thermal properties of both adlay flour and starch mentioned above could be used to control the desirable film properties by manipulating the changing of starch gelatinization and protein solubility by casting temperature and solvent during film preparation. Therefore, in this study the effect of casting temperatures (70 and 80°C) and casting solvent types (water and ethanol) as well as concentrations (30 and 40%) were determined.

Duonoution -	Samples		
Properties	Flour	Starch	
Chemical composition (% db)			
Protein*	14.17 ± 0.00	0.24±0.02	
Lipid [*]	4.14±0.34	$0.00{\pm}0.00$	
Ash*	1.31 ± 0.01	0.41 ± 0.01	
Fiber [*]	0.32±0.04	0.22±0.01	
Carbohydrate*	80.06±0.31	99.12±0.04	
Amylose content **	-	26.77±0.72	
Thermal properties			
To (onset, °C) ^{ns}	64.75±0.58	65.00±1.17	
$Tp(peak, °C)^*$	69.78±0.30	68.94±0.14	
Tc (conclusion, °C) ^{ns}	73.67±0.12	73.00±0.43	
Δ H (Enthalpy, J/g (db)) [*]	4.02±0.60	15.24±1.57	

Table 1 Chemical and thermal properties both of adlay flour and adlay starch

*- Means in the same row were significantly different (p<0.05).

ns - Not significance

** Amylose content was only determined in adlay starch.

Adlay flour and adlay starch film properties

In figure 1, it showed the appearance of adlay flour and starch films. The results showed that the adlay starch films (Figure 1a, b) were more transparent and shinier than the adlay flour films (Figure 1c, d, e, f); moreover, the adlay flour films had slightly brown color. It may be due to the presence of other chemical compositions such as protein and lipid (Pelissari *et al.*, 2013) as shown in Table 1. Besides, the appearances of adlay flour and starch films prepared by using distilled water with different casting temperatures had similarity.

In addition, when the effect of ethanol concentration was considered, the results showed that adlay flour films prepared by using distilled water and 30% EtOH could form continuous film; while the structure of adlay flour film prepared by using 40% EtOH had some pores that caused the discontinuity of film surface (Figure 1f). Moreover, the film was more opaque and its surface was rougher with the increasing of ethanol concentrations (Figure 1d, e, f). However, the adlay films prepared by using distilled water at 80°C, especially starch films, were more difficult to be taken off from casting surface than 70°C.

The discontinuous network of adlay flour film might be due to the less or free of biopolymer zones in the continuous phase of film matrix, and these zones led to the porous structure after film drying process (solvent evaporation process). Besides, the increase of opacity of adlay flour films when increasing ethanol concentrations might be due to the aggregation of starches and proteins in film solution, occurred during film forming process (data not shown) (Kurakake *et al.*, 1997; Kim and Xu, 2008).



Figure 1 Film appearance of adlay starch (a, b) and adlay flour film (c, d, e, f), prepared with distilled water (a, b, c, d), 30% EtOH (e) and 40% EtOH (f) at 70 °C (a, c) and 80 °C (b, d, e, f).

The thickness and density of flour and starch films were presented in Table 2. The results showed that the adlay flour films were thicker than the starch films significantly (Table 2) because the solid content of flour films was higher than that of starch films. When using different casting temperatures, the results showed that the thickness of films prepared at 80°C was not significantly different from that of films prepared at 70°C. However, when increasing the casting temperature, the density of films was decreased.

When comparing film sample prepared with different ethanol concentrations (0, 30 and 40%), the results showed that the adlay flour film prepared with 40% EtOH was the thickest but had the lowest density among all of films prepared with 30% EtOH and distilled water (0% EtOH) (Table 2). The difference of film thickness and density may be due to the difference of film solution viscosities, which tended to increase when the casting temperatures and ethanol concentrations increased (data not shown). The increasing of film solution viscosity may result from the different starch and protein dispersion, starch granule remaining including the polymer interactions, that responded to the different film formation.

Furthermore, the moisture content of adlay starch films (Table 2) was significantly higher than that of adlay flour films. This might be due to the flour composition, which contained alcohol soluble protein and lipid, leading to the reduction of water absorption of film (Belton et al., 2006; Colla et al., 2006). In addition, the moisture content of adlay flour and starch films tended to increase with the increase of the casting temperatures. On the contrary, the moisture content of adlay flour films decreased with the increase of the ethanol concentrations. Such a decrease may be due to the distribution of prolamin protein in the adlay flour film structure, since the increase of ethanol concentrations could increase prolamin solubility in adlay flour film solution (Kim and Xu, 2008).

Treatment		Thisland		Maint	
Sample	Casting solvent	Temp (°C)	(mm)	Density (g/cm ³)	Moisture content (%)
Starch	H ₂ O	70 °C	$0.16{\pm}0.01^{d}$	$1.11{\pm}0.04^{a}$	25.11±1.90 ^b
Starch	H ₂ O	80 °C	$0.15{\pm}0.02^d$	$0.95{\pm}0.05^{b}$	28.75±1.49 ^a
Flour	H ₂ O	70 °C	0.21±0.01 ^c	$1.03{\pm}0.06^{ab}$	$21.24{\pm}1.20^{d}$
Flour	H ₂ O	80 °C	$0.22{\pm}0.03^{\circ}$	$0.73{\pm}0.09^d$	24.97±1.02 ^b
Flour	30% EtOH	80 °C	$0.26{\pm}0.01^{b}$	$0.85{\pm}0.06^{\circ}$	$23.66{\pm}0.74^{bc}$
Flour	40% EtOH	80 °C	$0.32{\pm}0.04^{a}$	$0.66{\pm}0.15^{d}$	22.32±1.44 ^{cd}

 Table 2
 Thickness, density and moisture content of adlay flour and adlay starch films

Means with different letters within the same column were significantly different (p<0.05)

Tensile properties

The mechanical properties of adlay films were shown in Table 3. The stress values of adlay starch film was significantly higher than that of adlay flour film (Table 3) when prepared by distilled water at 70°C; however, the stress values of starch and flour films prepared at 80°C were not significantly different. In addition, the stress values of adlay starch and flour films, prepared at 70°C, were higher than those of adlay films, prepared at 80°C, significantly (p<0.05). It was due to the fact that at higher temperature (80°C), it caused the increase of the degree of gelatinization and led to the higher amylose leaching and granule disruption. In addition, the presence of amylopectin in the film solution might interrupt amylose in forming tightly bound network and lead to the decrease of the stress values (Paes et al., 2008).

When compared with the adlay flour films prepared with different ethanol concentrations at 80°C (Table 3), the results showed that the stress value of the adlay flour film, prepared with 30% EtOH was higher than that of films prepared with distilled water and 40% EtOH, respectively.

In addition, strain value was related to the film extensibility and it was found that the strain values of adlay starch films were higher than those of adlay flour films significantly (Table 3). It might be due to the discontinuity of starch film structure when composited with other compositions, corresponding to the decreasing of elongation at break (Cano *et al.*, 2014). Interestingly, the stress and strain value of adlay flour film, prepared with 30% EtOH at 80°C was higher when compared with other film samples (Table 3). It may be due to the effect of ethanol on both starch and protein.

From the results mentioned earlier, they indicated that the adlay flour film prepared with 30% EtOH at 80°C had a potential to produce film. Because it had higher stress than that of starch film; moreover, it was resistant to the moisture absorption than the adlay starch film. In addition, the utilization of adlay flour as a raw material for bio-based film production could also reduce the cost and chemical waste that obtained by starch isolation process.

Treatment		Tensile values		
Sample	Casting solvent	Temp (°C)	Stress (MPa)	Strain
Starch	H ₂ O	70 °C	$0.90{\pm}0.38^{b}$	$0.29{\pm}0.04^{c}$
Starch	H ₂ O	80 °C	$0.37{\pm}0.05^{d}$	$0.74{\pm}0.07^{a}$
Flour	H ₂ O	70 °C	$0.74{\pm}0.06^{c}$	$0.23{\pm}0.02^{d}$
Flour	H ₂ O	80 °C	$0.47{\pm}0.08^{d}$	$0.28{\pm}0.04^{c}$
Flour	30% EtOH	80 °C	$1.13{\pm}0.18^{a}$	$0.36{\pm}0.09^{b}$
Flour	40% EtOH	80 °C	$0.43{\pm}0.10^{d}$	0.10±0.03 ^e

 Table 3
 Stress and strain values of adlay flour and adlay starch films, prepared with different casting solvents and different temperatures

Means (n=10) with different letters within the same column were significantly different (p<0.05)

Microstructure of adlay flour and adlay starch films

The morphology of adlay flour and starch films at a surface and internal structure was observed by scanning electron microscopy (SEM) shown in Figure 2. The surface of adlay flour and starch films prepared using distilled water with different casting temperatures was continuous; moreover, the surface of adlay starch film prepared at 70°C (Figure 2a) could be observed for granular particles more than that of adlay starch film prepared at 80°C (Figure 2b). It may be due to the remaining of partially gelatinized starch granules since the adlay starch was completely gelatinized at about 73°C (Table 1). However, the larger granular particles (>20 μ m) were also observed in adlay starch film prepared at 80°C. It might be due to the ghost granules and remnants (Paes *et al.*, 2008; Koch *et al.*, 2010). In addition, the internal structure of the adlay starch films was more homogeneous and denser (Figure 2g, h) than that of theadlay flour films. It may be due to the presence of other chemical compositions, as shown in Table 1, could interrupt the interaction between starch polymers to form film network resulting in space between the adjacent polymer chains being increased (Pelissari *et al.*, 2013). This evidence was in agreement with the decreasing of density as well as extensibility (strain) of adlay flour film as mentioned earlier (Table 2 and 3).



Figure 2 Scanning electron micrographs at surface (a, b, c, d, e, f) and cross section (g, h, i, j, k, l) of adlay starch films (a, b, g, h) and adlay flour films (c, d, e, f, i, j, k, l) prepared with (distilled water (a, b, c, d, g, h, i, j), 30%EtOH (e, k) and 40%EtOH (f, l) at 70°C (a, g, c, i) and 80°C (b, h, d, j, e, f, k, l). Magnification is 350x.

The effect of casting solvents on morphology of adlay flour films is shown in Figure 2. The result showed that the surface of adlay flour films prepared with distilled water and 30% EtOH formed continuous film (Figure 2d, e); however, the adlay flour film prepared with 40% EtOH showed the discontinuous film (Figure 2f). In addition, the surface of adlay flour films was rougher when increasing ethanol concentrations (Figure 2d, e, f). It may be due to the aggregation of starch and protein portions (Kurakake et al., 1997; Kim and Xu, 2008). In addition, when considering the internal structure of the adlay flour films prepared with distilled water and 30% EtOH at 80°C (Figure 2j, k), it showed a good dispersion and film continuity than the film prepared with 40% EtOH (Figure 21). Moreover, the internal morphology of the adlay flour film prepared with 30% EtOH (Figure 2k) seems to be denser than that of the film prepared with distilled water (Figure 2j). This could be indicated that the good miscibility of chemical compositions was correlated with the density (Table 2) and tensile values (Table 3). Conversely, the adlay flour film prepared with 40% EtOH had shown the discontinuity and pores (Figure 21) which could relate to the decrease of tensile properties (Table 3).

In order to proof the assumption that the good dispersion and miscibility of starch and protein components as well as their interactions may be improved the mechanical properties of the adlay flour films, especially film prepared with 30% EtOH at 80 °C. As a result, the confocal laser scanning microscopy (CLSM) should be used to observe the distribution and miscibility between starch and protein portions in film matrix since the specific interaction between dyes and biopolymer molecules. In addition, the miscibility and interactions among chemical compositions in flour film matrix should be analyzed by dynamic mechanical analyzer (DMA) and fourier transform infrared spectrum (FTIR). Therefore, these techniques will be used to analyze the adlay flour films prepared with different casting solvents in order to understand the role of starch and protein on flour film properties.

CONCLUSION

The adlay starch films had more transparency than the adlay flour films; moreover, the adlay flour films had brown color. The different casting temperature did not seem to effect on film appearance. The thicknesses of the adlay flour or starch films prepared with distilled water were not different when prepared at different temperatures, whereas the densities of the films were decreased. In addition, the moisture contents of both adlay flour and starch films increased with the increasing of the casting temperatures. Furthermore, the stress values of both adlay starch and flour prepared with distilled water at 70°C were higher than those of films prepared at 80°C. In addition, the strain values of adlay starch and flour films prepared with distilled water at 80°C were higher than those of films prepared at 70°C; moreover, the strain values of adlay starch films were higher than those of adlay flour films.

The increasing of ethanol concentrations affected the increasing of opacity. In addition, the thicknesses of the adlay flour films were increased with increasing ethanol concentrations. However, the densities of adlay flour film prepared with 30% EtOH were higher than those of flour films prepared with distilled water and 40% EtOH. Furthermore, the increasing of ethanol concentrations resulted in the decrease of moisture contents in adlay flour films. Interestingly, the stress and strain values of adlay flour films prepared with 30% EtOH at 80°C were higher than those of adlay flour films prepared with other solvents, which could be related to the continuous network of flour film including the dispersion and miscibility of biopolymers, mainly protein and starch components within the film network.

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