A Biodegradable Film from Jackfruit (Artocarpus heterophyllus) and Durian (Durio zibethinus) Seed Flours

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2015, **16** (4), pp. 395 – 404

A BIODEGRADABLE FILM FROM JACKFRUIT (*ARTOCARPUS HETEROPHYLLUS*) AND DURIAN (*DURIO ZIBETHINUS*) SEED FLOURS

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Received: October, 03, 2015
Accepted: December, 07, 2015

Abstract:

Keywords: *jackfruit seeds, durian seeds, mechanical properties, morphology, biodegradable films*

INTRODUCTION

Lately, research on biodegradable plastic films (or bioplastic films) continues to grow all over the world due to the claims of environmental concern. Natural biodegradable polymers such as polysaccharides are potential raw material for biodegradable films. The most widely used polysaccharides in biodegradable films making is starch due to its low cost and abundant availability [1]. Starch is made of two natural polymers, i.e. amylose and amylopectin. Amylose is an unbranched polymer with molecular weight in the order of about 10^6, while amylopectin is a highly branched polymer having molecular weight in the order of 10^8 [2]. Amylose in starch contributes to good mechanical properties while amylopectin, due to its branched structure, generally leads to weaker films [3, 4].

Previous research on making films from pure starch revealed that the films were very brittle due to the high cohesive energy density of the polymers [5, 6]. Usually plasticizers are added to increase film flexibility by increasing the intermolecular spacing among polymers [7]. This will increase the molecular mobility, lower the glass transition temperature (Tg), and decrease the degree of the crystallinity [8]. The most commonly used plasticizers are water and low molecular weight polyols, such as glycerol [5].

Many groups of researchers have used starch from different sources to make films and coatings. It has indicated that starch is a promising material for biodegradable films. In addition, flours contain many components other than starch, like, cellulose, lipids and proteins which may contribute to specific characteristic of the film [9, 10]. However, only few studies on the utilization of flour for the production of films have been conducted. Rayas et al. [11] have prepared films from three types of wheat flours. Tapia-Blácido et al. [12] and Colla et al. [13] have produced films using amaranth flour as raw material. Dias et al. [14] and Sousa et al. [15] have used rice flour to produce biodegradable films. Andrade-Mahecha et al. [16] have employed the flour from achira, a native plant of the Andes in South America. Pitak & Rakshit [17] and Pelissari et al. [1] have made biodegradable film from banana flour. More recently Solano & de Gante [18] have produced biodegradable film from blue corn flour.

Interesting renewable sources, which are now being considered as waste, for the production of biodegradable films are fruit seeds, such as jackfruit (*Artocarpus heterophyllus*) and durian (*Durio zibethinus*) seeds. Jackfruit and durian are two of the most popular tropical fruits grown in Asia, including Indonesia. Jackfruit and durian are generally consumed fresh. The husk and the seed, which are 20-25% [19] and 8-15% [20] for durian and jackfruit, respectively, are usually disposed. However, in some parts of Indonesia, jackfruit seed are sometimes boiled or roasted and are eaten as a snack, while durian seeds are rarely eaten. Durian and jackfruit seeds have high amylose content, which are 22.76 % and 24.40 % respectively [21, 22]. Therefore, they are potential raw materials for biodegradable film [6, 23].

The objective of this work was to produce biodegradable films from jackfruit and durian seed flours with glycerol as the plasticizer. The mechanical properties of the films studied were Young's modulus, tensile strength, and elongation at brake. The surface and cross section of the films were observed using Scanning Electron Microscope.

MATERIALS AND METHODS

Raw materials

Jackfruit and durian seeds were obtained from fruit plantation located in Ambarawa, Central Java province, Indonesia. The seeds were cleaned and peeled manually and cut into thin slices. Then the slices were solar dried for 2 days, milled, and sieved to –100 +140 mesh. Glycerol as the plasticizer and other chemicals used in this work were of analytical grade and were purchased from Merck.

Composition analysis

The proximate analysis (moisture, ash, fat, and protein content (N x 6.25)) of durian and jackfruit seeds flours were determined according to AOAC method [24]. Carbohydrate content was calculated by differences. Amylose content was measured according to Juliano [25] by iodine method after lipid extraction. Each measurement was performed in duplicate.

Film preparation

The films were prepared by casting technique. Mixed flour (MF) with various compositions was dispersed in distilled water to form a 10 % (w/w) mixture. Glycerol with concentrations of 0.30 and 0.40 g/g MF was then added to the mixture. Table 1 shows the detailed variation and coding for the films. After adding 0.1 N HCl solution as much as 1.1 $mL\cdot g^{-1}$ MF to the mixture, it was then heated to 95 °C with constant stirring at 200 rpm for 10 minutes. The solution was then neutralized with 0.1 N NaOH solution, degassed under vacuum for 20 minutes, casted on a 20 × 20 cm acrylic plate, and dried at 50 °C for 20 hours. Before characterization, the resulted films were kept at room temperature for 7 days in an environment with relative humidity of 60 %.

***Table 1**. Codes of film formulation of the films*

Glycerol (l), [$g\cdot g^{-1}$ MF]	Durian seed flour (d), [g]	Jackfruit seed flour (j), [g]	Code l-d-j
0.3	10	0	3-10-0
0.3	8	2	3-8-2
0.3	6	4	3-6-4
0.3	5	5	3-5-5
0.3	4	6	3-4-6
0.3	2	8	3-2-8
0.3	0	10	3-0-10
0.4	10	0	4-10-0
0.4	8	2	4-8-20
0.4	6	4	4-6-4
0.4	5	5	4-5-5
0.4	4	6	4-4-6
0.4	2	8	4-2-8
0.4	0	10	4-0-10

Mechanical properties

The Young's modulus, tensile strength, and elongation at break of the films were determined using texture analyzer Lloyd instrument type TA plus. The films were cut into 10 mm wide and 100 mm long slices for the analysis. All measurements were conducted in crosshead speed of 5 mm·sec^{-1}, gauge length of 20 mm, width of 2 mm, and thickness of 2 mm.

Scanning electron microscopy (SEM) analysis

The surface and cross section of the films were observed by SEM method using Joel JSM-T300 Scanning Electron Microscope device. Only films obtained from pure jackfruit and durian seed flours as well as from composite flour containing equal amount of both seed flours were subjected to SEM analysis.

RESULTS AND DISCUSSION

The proximate analysis of durian and jackfruit flours

The chemical compositions of durian and jackfruit seed flours used in this work are presented in Table 2. According to Table 2, amylose content of jackfruit seed flour (23.30 %) is slightly higher than that of durian seed flour (22.65 %). These results are close to Tongdang's work [22] who reported that amylose content of jackfruit and durian seed starch were 24.40 % and 22.76 %, respectively. On the other hand, the proximate and the amylose content are different from those obtained by Tulyathan *et al.* [26] and Mukprasirt and Sajjaanantakul [20]. This fact could be attributed to the cultivar, the growing environment, the harvesting season, and the fruit maturity differences [21].

Table 2*. Chemical composition of durian seed and jackfruit seed flour*

Composition (%, dry basis)	Durian	Jackfruit
Moisture	10.07±0.05	11.48±0.03
Fat	1.09±0.47	1.63±0.24
Protein	7.73±0.08	13.97±0.03
Ash	2.86±0.07	2.95±0.02
Total Carbohydrate[a]	78.25±0.52	69.97±0.73
Amylose	22.65±0.28	23.30±0.01

[a]determined by differences

Mechanical properties of the films

Table 3 shows the effect of mixed flour (MF) composition and glycerol concentration on Young's modulus, tensile strength, and elongation at brake of the films. Young's modulus, tensile strength, and elongation at brake rise with the increasing of jackfruit seed flour content for the same glycerol concentration.
The Young's modulus and tensile strength of the films made of merely jackfruit seed flour are much higher than that made of only durian seed flour for both glycerol

concentrations of 0.30 and 0.40 $g \cdot g^{-1}$ MF. The properties of films made of mixed flours vary, depending on the composition of the mixture.

Table 3. *Mechanical properties of the films*

Film formulation code (l-d-j)	Mechanical properties		
	Young's modulus [MPa]	**Tensile strength [MPa]**	**Elongation at break [%]**
30-100-0	6.76±1.14	2.14±0.20	-
30-80-20	8.85±2.03	2.81±0.36	-
30-60-40	9.98±1.81	3.12±0.31	-
30-50-50	14.20±3.31	4.39±0.38	29.26±1.73
30-40-60	24.31±4.03	4.55±0.57	32.29±1.47
30-20-80	43.52±2.5	4.78±0.25	34.46±1.96
30-0-100	51.31±3.65	5.31±0.39	37.83±0.20
40-100-0	2.71±0.53	1.07±0.10	-
40-80-20	3.83±0.08	1.38±0.15	-
40-60-40	4.90±1.92	2.39±0.47	-
40-50-50	11.37±1.07	3.15±0.33	37.17±2.07
40-40-60	13.01±1.83	3.30±0.21	38.09±0.86
40-20-80	20.89±0.99	3.55±0.65	40.82±3.73
40-0-100	45.76±2.46	5.01±0.21	44.11±2.10

The difference in film's properties can be related to chemical composition of the flour. The major component of flour is starch, which consists of amylose and amylopectin. The available hydroxyl groups on the starch chains participate in the formation of hydrogen bonds [6]. However, linear chains of amylose are able to interact by hydrogen bonds to a higher extent than the branched amylopectin chains [27]. So, amylose contributes more significantly to mechanical properties of films than amylopectin does [28 – 30]. Other reason is the easy entanglement of long linear amylose chains, which may act as self-reinforcement [2]. For these reasons, the jackfruit seed flour which has higher content of amylose, can make stronger film than durian seed flour. These results are consistent with those obtained by Li et al. [2], Rindlav-Westling et al. [30], and Cano et al. [31]. According to Rindlav-Westling et al. [30], tensile strength of film made of pure amylose was 20 MPa while that of pure amylopectin was 6 MPa. The tensile strength of the films in this work is lower than the values found by Rindlav-Weslting et al. [30]. It could be a result of the existence of irregularities at the microstructure level, and the presence of lipids in the flour, because lipids are not able to form a cohesive and continuous matrix. The presence of protein also affects the mechanical properties of the film in the same manner as lipid [14]. As can be seen in Table 3, the Young's modulus and tensile strength of the film made of jackfruit flour with glycerol concentration of 0.30 g/g flour in this study are 51.31 ± 3.65 MPa and 5.31 ± 0.39 MPa, respectively. It is better than the results reported by Dias et al. [14] who made film from rice flour with the same glycerol concentration. They found that the Young's modulus and tensile strength of the film were 22.2 ± 6.0 MPa and 1.3 ± 0.1 MPa, respectively. The films made of mixed flour with jackfruit seed flour higher than 60 % also shows higher values of Young's modulus and tensile strength than rice flour films. Moreover, jackfruit seed and durian seed are much considered as agricultural waste with no value. These facts indicate that durian seed and jackfruit seed flours are more favorable than rice flour for producing biodegradable film.

The difference between mechanical properties of films made of both flours can also be related to different cristallinity of amylose and amylopectin. Amylose and amylopectin exhibit different gelation characteristic due to their different crystallinity. According to Rindlav-Westling et al. [30], crystallization of amylopectin from solution is much slower than amylose. The film made of amylopectin becomes amorphous when it is dried, because it does not have enough time to crystallize. Meanwhile, amylose, which crystallizes faster, has sufficient time to form crystalline. This is why the amylose film has higher Young's modulus and lower elongation at break [30]. Glass transition temperature (Tg) of the material also affects the resulted films. Amylopectin, having Tg slightly lower than room temperature, will produce plastic film, while amylose with Tg slightly higher than ambient temperature, will form fragile film [29]. Hence, the film with higher portion of jackfruit seed flour results in higher amylose content, which in turn will produce a more fragile film.

As it is shown in Table 3, the elongation at break increases as the content of jackfruit seed flour increases, which means that the content of amylose increases as well. The linear and flexible structure of amylose molecules could also contribute to the higher elongation at break [2].

Molecular weight of the amylose also plays important role on mechanical properties. Films from lower molecular weight amylose have smaller tensile strength and elongation [29]. Although the content of amylose in both flours only slightly different, the molecular weight of amylose in both flour may be largely different. Therefore it may cause significant difference in mechanical properties.

The Young's modulus and tensile strength of all films decrease while the elongation at break increases as the concentration of glycerol increases from 0.3 to 0.4 $g \cdot g^{-1}$ MF. Similar trends were also observed by other researchers [32 – 36]. Having hydroxyl groups, glycerol forms hydrogen bonds with amylose and amylopectin molecules. This will reduce the intermolecular attraction between amylose/amylopectin chains, which further results in greater flexibility and thus decreases the tensile strength of the films. Another consequence of the more flexible film is the increase of elongation at break. Increasing glycerol concentration causes the increase of free volume in the film [34, 37]. Addition of plasticizers such as water and glycols will lower the Tg and make the starch more rubber-like. Plasticizers act by spacing out the molecules and reducing the interactions [38]. From the above results it can be seen that the Young's modulus, tensile strength, and elongation at break are strongly influenced by the concentration of glycerol.

Morphology of the films

SEM micrographs, as presented in Figure 1, provide information about the surface morphology and internal microstructure of the films. The micrographs show the surface and cross section of three films, i.e. 30-100-0 film (Figures 1.a and 1.bb), 30-50-50 (Figures 1.c and 1.d), and 30-0-100 (Figures 1.c and 1.f). The surface of film made of merely durian seed flour (Figure 1.a) shows big cracks, while that made of only jackfruit seed flour (Figure 1.e) looks smoother. These cracks explain the lower values of Young's modulus and tensile strength of durian seed flour film.

The surface and cross section of the films show irregularities. It may be due to many polymer molecules, such as starch, protein, lipid, and cellulose present in the matrix of the film. Mechanical properties of the film could be indicated by the uniformity of the surface. The uniformity of the film matrix will increase polymers integrity, hence raise the mechanical properties of the films [14]. Visual performances of film made of the mixed flour with higher durian seed flour content, are glossier and more transparent. It can be related to the higher content of gum in durian seed flour. The durian seed contains 18.0 % of crude gum [19].

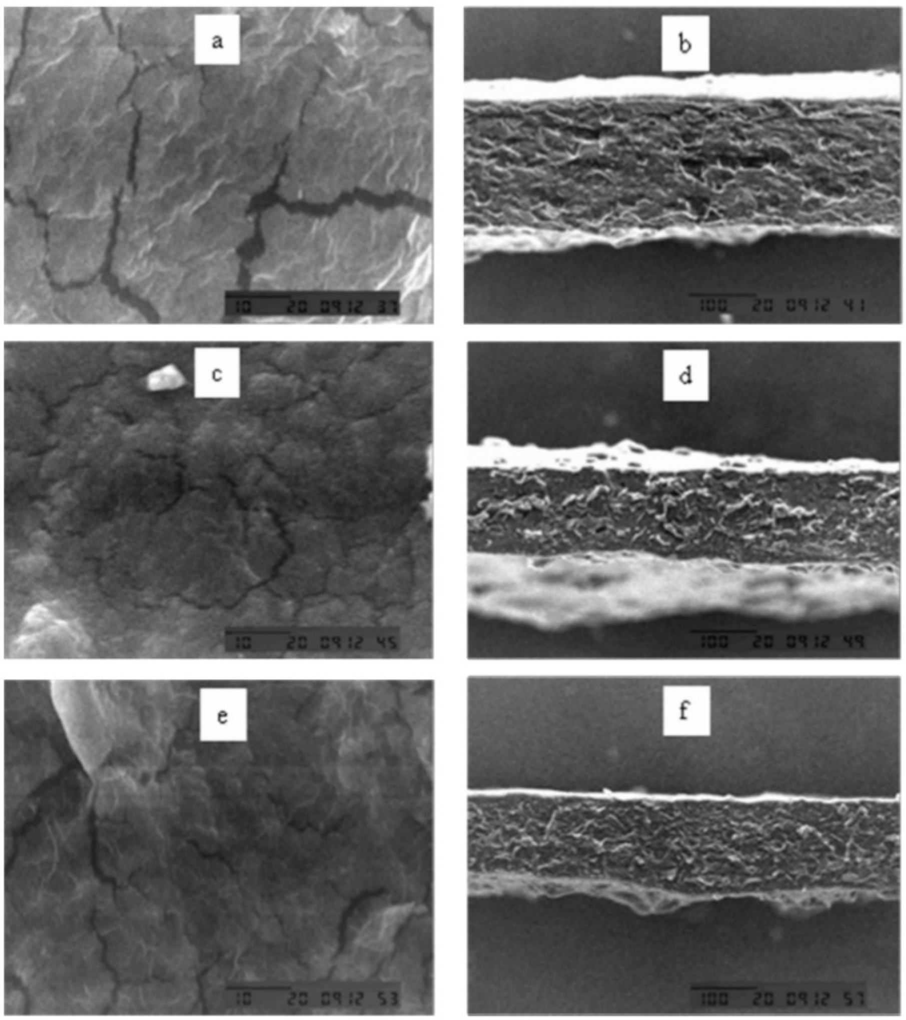

***Figure 1**. SEM images of the films surface and cross section at 2000 × magnification: (a) and (b) 30-100-0 film; (c) and (d) 30-50-50 film; (e) and (f) 30-0-100 film*

CONCLUSIONS

Biodegradable films made from jackfruit seed and durian seed flours have been developed. The properties of the films are influenced by the composition flour mixture and the glycerol concentration. Jackfruit seed flour contributes to the strength and elongation at break of the film as a result of its higher content in amylose. is also found that higher concentration of glycerol will result in lower values of Young's modulus and tensile strength and higher value of elongation at break.

The films made of mixed jackfruit and durian seed flour have higher values of Young's modulus and tensile strength than those of films made of rice flour [13]. This makes jackfruit and durian seed flour are potential to be developed as biodegradable films as they were prepared from merely natural sources, making them being decomposed more easily by microbes and therefore environmentally friendly.

ACKNOWLEDGMENT

The authors would like to acknowledge the Faculty of Engineering, Diponegoro University, Indonesia for the financial support for this research.

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