Preparation of biodegradable foam from sago residue

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Abstract: We prepared biodegradable foam by pressing and baking a mixture containing native sago residue, cellulose, polyvinyl alcohol, magnesium stearate, and water. The foam was opaque and brown; it had a rough surface and a porous internal structure. The foam was denser than commercial polystyrene foam. However, the modulus of elasticity and modulus of rupture in flexure of the foam prepared from native sago residue were higher than those of polystyrene foam, indicating that the foam from sago residue was stiff, had high strength, and would be suitable for holding and protecting heavy materials. We also prepared another foam using acid-treated sago residue instead of native sago residue. The foam from the acid-treated residue was thinner and more porous than the foam from the native residue. A higher modulus of elasticity in flexure of the foam from the acid-treated residue was also observed. Thus, we found that partial acid hydrolysis of starch in sago residue modifies the structural and physico-mechanical properties of the resulting foam.

Keywords: acid treatment, biodegradable foam, packaging, physico-mechanical properties, porous structure, sago residue

Introduction

Sago palm is an important crop for the starch industry in Southeast Asia and Oceania (Singhal et al., 2008; Awg-Adeni et al., 2013). A large amount of sago pith waste (sago residue) is produced after starch extraction and is usually discarded nearby, generating environmental problems (Awg-Adeni et al., 2010). The utilization of sago residue is expected not only to reduce the disposal of waste, but also to produce other beneficial materials. Many studies have attempted to utilize sago residue for producing beneficial materials, e.g., fermentable sugar, enzymes, adsorbents, and bioethanol (Singhal et al., 2008; Linggang et al., 2012; Awg-Adeni et al., 2013). However, foam material for packaging has not yet been produced from sago residue.

In the packaging industry, researchers attempt to design alternative biomaterials to replace synthetic polymers in view of environmental and economical considerations. Biodegradable foam is one of the inventions expected to be used for ordinary packaging that can be degraded by microorganisms. Some studies have shown that it is possible to prepare biodegradable foam from starches, natural fibers, and water (Benezet et al., 2012). Fibers adhere to the starch matrix and act as reinforcements (Shogren et al., 2002), while water acts as a foaming agent and a plasticizer (Bergeret and Benezet, 2011). In some cases, other polymers such as polyvinyl alcohol (PVA) and additives are also added to improve the properties of the foam (Shogren et al., 2002; Cinelli et al., 2006; Benezet et al., 2012). PVA is a biodegradable synthetic polymer that improves the strength, flexibility, and water resistance of foam (Shogren et al., 1998). Foam is generally prepared by either the extrusion process or the pressing and baking process (Glenn and Orts, 2001). In the extrusion process, materials are fed into a rotating screw

extruder with gradual heating. The pressing and baking process, on the other hand, involves the casting of materials in a closed heated mold. The preparation of biodegradable foam from cassava starch (Salgado et al., 2008), wheat starch (Zhou et al., 2007), and potato starch (Cinelli et al., 2006) has been extensively studied. Biodegradable foam was also prepared from sago starch (Pimpa et al., 2007). However, foam prepared from sago residue has not been reported.

In this study, we prepared biodegradable foam from sago residue and characterized it. The physico-mechanical properties were determined, and the microstructure of the foam was observed. We also prepared foam from acid-treated sago residue.

Materials and Methods

1. Sago residue

Sago residue, after the extraction of starch, was obtained from a sago producer in West Java, Indonesia, and was sun dried for 2 days. The content of cellulose, hemicellulose, and lignin in the residue was determined according to Linggang et al. (2012). Briefly, the content of cellulose was determined by subtracting the weight of lignin from the weight of the insoluble material after treating the sago residue with a dilute-acid detergent solution (0.5 M sulfuric acid containing 0.05 M cetrimonium bromide). The content of hemicellulose was the difference between the insoluble material after treatment of the sago residue with a dilute-acid detergent solution and that with a neutral detergent solution containing 0.1 M sodium dodecyl sulfate. The content of lignin was measured by weighing the insoluble material after hydrolysis of the insoluble material after treatment of the sago residue with a dilute-acid detergent solution using 13 M sulfuric acid. The starch content was determined by quantifying reducing sugars in acid hydrolysates of starch using the method ISO 5377:1981 (http://www.iso.org).

2. Preparation of acid-treated sago residue

Sago residue (83 g, dry weight basis) was suspended in 1.5 L of 2.0 M sulfuric acid and incubated at 37°C for 10 days with daily shaking. The mixture was then neutralized using 2.0 M sodium hydroxide and filtered under vacuum using filter paper. The solid particulates were washed with distilled water repeatedly and dried at 37°C for 3 days.

3. Preparation of foam from sago residue

Sago residue (2 g; native residue or acid-treated residue), cellulose (0.5 g), PVA (2 g; degree of hydrolysis of 99.4-99.8 mol%; PT Tirta Marta, Cikupa Tangerang, Indonesia), water (9 g), and magnesium stearate (0.5 g) were homogenized using a high-speed mixer with a wire whisk attachment (Cucina HR 1530/8, Philips, Amsterdam, Netherlands) for 10 min. Cellulose was prepared from sago residue by the method described by Hisyam (2012). Magnesium stearate was used as a mold release agent (Salgado et al., 2008). The sago residue mixture was cast by pressing and baking at 170°C for 3 min using a baking machine. This equipment, consisting of a steel insert and a steel cavity mold, was made according to Glenn and Orts (2001) with minor modifications of the mold sizes as follows: the inside dimensions of the insert mold are 200 mm in length, 122 mm in width, and 13 mm in height; while the inside dimensions of the cavity mold are 203 mm in length, 119 mm in width, and 13 mm in height. Both molds were preheated before casting, and the insert (top) mold was moved downward to mate with the cavity (bottom) mold.

4. Physico-mechanical characterization of the foam

The foam was cut into small square pieces, and their length, width, and thickness were measured using a micrometer screw. Samples were then weighed, and their densities were calculated.

To evaluate the flexural properties of foam, the modulus of elasticity (MoE) in flexure and the

modulus of rupture (MoR) in flexure were determined using a universal testing machine (Model 4500, Instron, Canton, MA, USA) according to ASTM method D790-91 (1991). All measurements were performed in duplicate or triplicate, and the average values were reported.
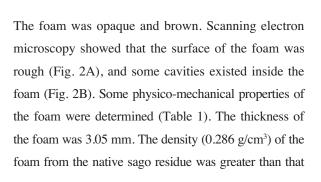
5. Scanning electron microscopy

The surface and a cross section of the foam were observed using an S-4000 scanning electron microscope (Hitachi Science Systems, Hitachinaka, Japan) with an accelerating voltage of 5 kV, as described previously (Srichuwong et al., 2005b).

Results

1. Characteristics of the foam prepared from native sago residue

Sago residue used in this study consisted of starch (63%), cellulose (11%), hemicellulose (7.6%), lignin (12%), and minor components (e.g., ash and minerals). The composition of this sago residue was similar to that reported by Abd-Aziz (2002).

To prepare the foam, native sago residue, cellulose, water, PVA, and magnesium stearate were mixed and baked in a pair of molds. Tray-like foam was obtained by pressing and baking the sago residue mixture (Fig. 1).
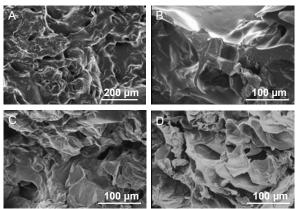
The foam was opaque and brown. Scanning electron microscopy showed that the surface of the foam was rough (Fig. 2A), and some cavities existed inside the foam (Fig. 2B). Some physico-mechanical properties of the foam were determined (Table 1). The thickness of the foam was 3.05 mm. The density (0.286 g/cm^3) of the foam from the native sago residue was greater than that

Fig. 2. Scanning electron micrographs of foam prepared from sago residue A, Surface of foam from native sago residue; B, cross section of foam from native sago residue; C, surface of foam from acid-treated sago residue; D, cross section of foam from acid-treated sago residue

Table 1. Physico-mechanical characteristics of foam prepared from sago residue

	Thickness (mm)	Density (g/cm^3)	MoE (MPa)	MoR (MPa)
Foam from native sago residue	3.05	0.286	387	16.3
Foam from acid-treated sago residue	2.15	0.362	705	17.6
Commercial polystyrene foam[a]	0.68	0.063	105	1.3

[a] Glenn et al. (2001)

of commercial polystyrene foam (0.063 g/cm^3; Glenn et al., 2001). The MoE (387 MPa) and MoR (16.3 MPa) of the foam from the native sago residue were high. The MoE represents the stiffness of a material, while the MoR reflects its strength.

2. Characteristics of the foam prepared from acid-treated sago residue

We prepared another foam using acid-treated sago residue instead of native sago residue. Shogren et al. (2002) described that the use of modified starch affects the structure and properties of the foam. Acid

Fig. 1. Foam prepared from native sago residue

hydrolysis has been used for a long time to modify starch properties (LeCorre, 2010). The thickness of the foam from the acid-treated residue (2.15 mm) was thinner than that from the native residue (Table 1). The density (0.362 g/cm^3) and MoR (17.6 MPa) of the foam from the acid-treated residue were almost the same as those from the native residue. However, a higher MoE (705 MPa) was observed in the foam from the acid-treated residue. As was the foam from the native residue, the surface of the foam from the acid-treated residue was rough (Fig. 2C). However, the foam was more porous than that from the native residue (Fig. 2D).

Discussion

Sago residue is considered suitable for making biodegradable foam for packaging because it contains a considerable amount of starch and fibers. In this study, we demonstrated that foam is prepared from sago residue by pressing and baking with a pair of molds. Preparing the foam by the pressing and baking method has an advantage, in that shapes of foam are easily modified by using different types of molds (Glenn and Orts, 2001). The MoE and MoR of the foam from the native sago residue were high, indicating that the foam had a mechanically strong structure (Table 1) and that it would be practically useful for packaging. During baking of the sago residue mixture, starch is gelatinized and water is evaporated, resulting in the formation of foam (Salgado et al., 2008). Fibers adhere to the starch matrix and act as reinforcements (Shogren et al., 2002). Starch and fibers have similar chemical structures, which might facilitate the formation of hydrogen bonds between them (Bergeret and Benezet, 2011). Cavities existing inside the sago residue foam (Fig. 2B) might be formed due to water evaporation during the pressing and baking (Salgado et al., 2008).

Partial hydrolysis of starch using dilute acid generates thin-boiling starches (e.g., Naegeli amylodextrin and Lintner starch). During acid hydrolysis, the amorphous parts of starches are preferentially hydrolyzed and dissolved; however, the main crystalline structures (acid-resistant residues) remain intact (Srichuwong, 2005a). Therefore, the use of acid-treated sago residue would modify the structural and physico-mechanical properties of the resulting foam. The thickness of the foam prepared from the acid-treated residue was low. Because the swelling power of the starch was reduced by partial hydrolysis, the foam could not expand sufficiently at the baking step. The water-retaining capacity of starch decreased with hydrolysis of the amorphous region of starch, which might result in the generation of more cavities inside the foam (Fig. 2D). Furthermore, the MoE (i.e., stiffness) of the foam from the acid-treated residue was higher than that from the native residue, which might be attributed to the fact that the acid-treated residue was less amorphous (Table 1). In contrast, the density and MoR of the foam from the acid-treated residue were similar to those of the foam from the native residue. This does not conflict with the observation by Glenn and Orts (2001) that MoR tends to be increased by the density of a starch-based foam.

The foam prepared from sago residue in this study has at least two disadvantages. First, the density of the foam is several times higher than that of commercial polystyrene foam, which would affect transportation costs in practical use. Second, the foam from sago residue readily absorbs a significant amount of water and becomes fragile (data not shown). Therefore, the foam can be applied only to low moisture materials (e.g., electronic equipment). Improving the density and water resistance of the foam is needed for low-cost and widespread use.

In conclusion, we succeeded in preparing biodegradable foam from sago residue. The foam was stiffer and had greater strength than commercial polystyrene foam. The use of acid-treated sago residue alters the microstructure and the physico-mechanical properties of foam. Biodegradable foam from sago residue would be suitable for holding and protecting heavy materials.

References

Abd-Aziz, S 2002 Sago starch and its utilisation. Journal of Bioscience and Bioengineering 94: 526-529.

American Society for Testing and Materials (ASTM) 1991 Method D790-91: Standard test methods for flexural properties of unreinforced and reinforced plastics and electrical insulating materials. *In*: Annual Book of ASTM Standards. ASTM (Philadelphia).

Awg-Adeni, D.S., S. Abd-Aziz, K. Bujang and M.A. Hassan 2010 Bioconversion of sago residue into value added products. African Journal of Biotechnology 9: 2016-2021.

Awg-Adeni, D.S., K.B. Bujang, M.A. Hassan and S. Abd-Aziz 2013 Recovery of glucose from residual starch of sago hampas for bioethanol production. BioMed Research International 2013: 935852.

Benezet, J.C., A. Stanojlovic-Davidovic, A. Bergeret, L. Ferry and A. Crespy 2012 Mechanical and physical properties of expanded starch, reinforced by natural fibres. Industrial Crops and Products 37: 435-440.

Bergeret, A. and J.C. Benezet 2011 Natural fibre-reinforced biofoams. International Journal of Polymer Science 2011: 569871.

Cinelli, P., E. Chiellini, J.W. Lawton and S.H. Imam 2006 Foamed articles based on potato starch, corn fibers and poly(vinyl alcohol). Polymer Degradation and Stability 91: 1147-1155.

Glenn, G.M. and W.J. Orts 2001 Properties of starch-based foam formed by compression/explosion processing. Industrial Crops and Products 13: 135-143.

Glenn, G.M., W.J. Orts and G.A.R. Nobes 2001 Starch, fiber and CaCO$_3$ effects on the physical properties of foams made by a baking process. Industrial Crops and Products 14: 201-212.

Hisyam 2012 Isolation of cellulose from sago waste by using hydrogen peroxide delignification. Undergraduate thesis, Bogor Agricultural University (Bogor).

Le Corre, D., J. Bras and A. Dufresne 2010 Starch nanoparticles: a review. Biomacromolecules 11: 1139-1153.

Linggang, S., L.Y. Phang, M.H. Wasoh and S. Abd-Aziz 2012 Sago pith residue as an alternative cheap substrate for fermentable sugars production. Applied Biochemistry and Biotechnology 167: 122-131.

Pimpa, B., K. Muhammad, Z. Ghazali, K. Hashim, M.A. Hassan and D.M. Hashim 2007 Optimization of conditions for production of sago starch-based foam. Carbohydrate Polymers 68: 751-760.

Salgado, P.R., V.C. Schmidt, S.E. Molina Ortiz, A.N. Mauri and J.B. Laurindo 2008 Biodegradable foams based on cassava starch, sunflower proteins and cellulose fibers obtained by a baking process. Journal of Food Engineering 85: 435-443.

Shogren, R.L., J.W. Lawton and K.F. Tiefenbacher 2002 Baked starch foams: starch modifications and additives improve process parameters, structure and properties. Industrial Crops and Products 16: 69-79.

Shogren, R.L., J.W. Lawton, K.F. Tiefenbacher and L. Chen 1998 Starch-poly(vinyl alcohol) foamed articles prepared by a baking process. Journal of Applied Polymer Science 68: 2129-2140.

Singhal, R.S., J.F. Kennedy, S.M. Gopalakrishnan, A. Kaczmarek, C.J. Knill and P.F. Akmar 2008 Industrial production, processing, and utilization of sago palm-derived products. Carbohydrate Polymers 72: 1-20.

Srichuwong, S., N. Isono, T. Mishima, and M. Hisamatsu 2005a Structure of lintnerized starch is related to X-ray diffraction pattern and susceptibility to acid and enzyme hydrolysis of starch granules. International Journal of Biological Macromolecules 37: 115-121.

Srichuwong, S., T. Sunarti, T. Mishima, N. Isono and M. Hisamatsu 2005b Starches from different botanical sources I: Contribution of amylopectin fine structure to thermal properties and enzyme digestibility. Carbohydrate Polymers 60: 529-538.

Zhou, J., J. Song and R. Parker 2007 Microwave-assisted moulding using expandable extruded pellets from wheat flours and starch. Carbohydrate Polymers 69: 445-454.