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Effect of cellulose fiber from sorghum bagasse on the mechanical properties and biodegradability of polylactic acid

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Abstract

Synthesis of biocomposites using polylactic acid (PLA) as the matrix and cellulose fiber from sorghum bagasse as the reinforcement agent has been carried out in this study. The objective was to improve the mechanical properties and biodegradability of PLA. In this study, cellulose fiber has been extracted from sorghum bagasse. Micro and nano-sized cellulose fiber was then produced by hydrolysis using sulfuric acid. Biocomposite was then made by casting film method using acetone as the solvent. Based on mechanical test, nanocellulose fiber gives better results on mechanical properties. Moreover, the tensile modulus and biodegradability of PLA were increase with the increasing of nanocellulose fiber concentration. Based on results, the addition of 2.5% nanocellulose fiber, improved the maximum strain from 5.19% to 11.59%. Otherwise, the best biodegradability of biocomposite was achieved on the addition of 15% nanocellulose fiber.

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Keywords: Biocomposite; Nanocellulose; Polylactic acid; Sorghum bagasse

1. Introduction

Polylactic acid (PLA) is thermoplastic polyester produced from renewable resources and biodegradable in nature [1]. Compared to other biopolymers, PLA has transparent properties, good printability and easy to produce [2]. However, PLA has brittle properties and low resistance at high temperatures [3]. Fortunately, the brittle nature of PLA can be improved by adding plasticizers that will increase the elongation break value of PLA. The plasticizer usually used is glycerol. Nevertheless, the addition of plasticizer will reduce the mechanical properties of PLA. Therefore, the addition of fiber in order to strengthen the mechanical properties of PLA needs to be investigated.

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One of fiber type that can be used is cellulose fiber. This is due to cellulose fiber has an ability to improve the mechanical properties of PLA. Besides, it has another advantages including biodegradable, high rigid, and low density [2]. In fact, sorghum bagasse is one of potential resources of cellulose fiber since it contains cellulose up to 31.42%. Based on previous study, sorghum fiber has Young's modulus of 15 GPa. This value almost equals with the Young's modulus from corn fiber and more than cotton fiber [4]. Moreover, elongation and breaking tenacity of sorghum is equal with another fiber such as corn fiber and banana fiber [5]. Therefore, this study was conducted to synthesize PLA using cellulose fiber from sorghum bagasse as the reinforcement agent. Thus, the aims of this study were to improve the mechanical properties and biodegradability properties of PLA.

2. Materials

2.1. Materials

Poly(lactic Acid (PLA) 4060D was obtained from Mitsui Chemical, Inc, Japan. Sorghum bagasse was supplied from Indonesian Institute of Sciences (LIPI, Cibinong, Indonesia). All chemicals were purchased from Merck. Laccase and xylanase enzymes for pretreatment were purchased from Sigma Aldrich.

2.2. Preparation of cellulose fiber

Cellulose fiber was prepared using the method by previous researcher [6]. Initially, sorghum bagasse was grinded into a smaller size and then sieved until the sorghum size was 2–4 mm. Sorghum fiber was then treated by several methods including alkalization, alkalization and bleaching, and enzymatic.

The alkalization method was carried out by mixing the sorghum fiber into 5% (w/w) of KOH solution with a ratio of sorghum fiber and KOH solution was 1:10. The mixture was then autoclaved at 121 °C for 25 min. Afterwards, the mixture was then filtered and the solids obtained were washed until neutral.

For the alkalization and bleaching method, after the alkalization process, sorghum fiber was further treated by bleaching process using 2.3% (w/w) of sodium chlorite solution in acetate buffer (pH 4.9) for 2 h at 70 °C. The solids obtained were then washed with distilled water until neutral.

The enzymatic method was carried out by utilizing laccase enzyme from *Trametes versicolor* in order to degrade lignin and xylanase enzyme from *Thermomyces lanuginosus* in order to degrade hemicellulose. Initially, sorghum fiber was immersed in laccase enzyme solution (0.01 g of enzyme in 10 mL of distilled water) at pH 4.5, with ratio of fiber and solution was 1:10. Immersion was carried out for 2 h at temperature of 25 °C. Afterwards, sorghum fiber were washed and dried in order to remove residual enzymes. Subsequently, the treated sorghum fiber was further immersed in xylanase enzyme solution (0.02 g of enzyme in 10 mL of distilled water) at pH 7.3–8.1 with ratio of solution was 1:10. Immersion was carried out for 2 h at temperature of 75 °C. Afterwards, sorghum fiber were washed and dried to remove the rest of the enzyme.

2.3. Preparation of nanocellulose fiber

Nanocellulose fiber was prepared by hydrolyzing pretreated sorghum fiber using 64% of sulfuric acid with ratio of fiber and solution was 1:5 (w/w). Hydrolysis was carried out for 30 min at 45 °C. At the end of hydrolysis process, the mixture was added with water. The nanocellulose fiber solution was then separated by decantation until a neutral pH was obtained.

2.4. Synthesis of biocomposite

The cellulose obtained was then carried out to a solvent exchange process using 300 mL of ethanol, repeated for 3 times, and continued with 300 mL of acetone, repeated for 2 times. PLA 4060D was then dissolved using 300 mL of acetone and 3.3% glycerol [7]. Cellulose was then added to the PLA solution with variations in concentrations of 2.5, 5, 10 and 15%. The solution was then stirred using an overhead stirrer for 3 h at 250 rpm. The solution was then molded on flexi glass and placed in acid room for 12 h.

2.5. Characterization

Composition of sorghum fiber after pretreatment was analyzed. 1 g of fiber (a) was refluxed for 2 h with 150 mL of water at 100 °C. Afterwards, fiber was dried in the oven and then weighed (b). Fiber was further refluxed with 150 mL of 0.5 M sulfuric acid for 2 h at 100 °C. Fiber was then dried in the oven and then weighed (c). Subsequently, the dried fiber was soaked in 10 mL of 72% sulfuric acid for 4 h and refluxed with 0.5 M sulfuric acid for 2 h at 100 °C. Fiber was then dried in the oven and then weighed (d). Fiber was then burned using a furnace at 575 ± 25 °C in order to produce ash (e).

The calculation of hot water soluble (HWS), hemicellulose, cellulose, lignin, and ash content on sorghum fiber using the following equations:

$$\text{Hot water soluble (HWS)} = \frac{a - b}{a} \times 100\% \quad (1)$$

$$\text{Hemi cellulose} = \frac{b - c}{a} \times 100\% \quad (2)$$

$$\text{Cellulose} = \frac{c - d}{a} \times 100\% \quad (3)$$

$$\text{Lignin} = \frac{d - e}{a} \times 100\% \quad (4)$$

$$\text{Ash} = \frac{e}{a} \times 100\% \quad (5)$$

Cellulose fiber's morphology was characterized by using Scanning Electron Microscopy (SEM). Mechanical properties of biocomposite was characterized according to ASTM D822-02 using Universal Testing Machine. Biodegradability from biocomposite was tested using soil burial test [8]. Sample was weighed before buried and every week after buried for 5 weeks.

3. Results and discussion

3.1. Effect of treatment on sorghum bagasse fiber composition

The fiber composition will determine the strength of the biocomposite produced. This is due to the cellulose content is directly proportional to the strength of the biocomposite. Therefore, the content of lignin and hemicellulose in bagasse sorghum must be removed through suitable treatment. Removal of lignin content aims to improve the water resistance of biocomposite. This is due to lignin has a function to transport water in plants so that it will be easy to absorb water [9]. Meanwhile, hemicellulose is removed because it will form a branched amorphous structure [10]. Based on experimental results, the composition of sorghum bagasse fiber after treatment is depicted on Table 1.

Table 1. Composition of sorghum bagasse fiber after treatment.

Composition	Without treatment	Alkalization	Alkalization and bleaching	Enzymatic
HWS ^a	0.0634	0.0563	0.0134	0.0423
Hemicellulose	0.1486	0.0398	0.0548	0.1218
Cellulose	0.3445	0.8014	0.911	0.7109
Lignin	0.3307	0.0987	0.0197	0.0964
Ash	0.1128	0.0038	0.0011	0.0286

^aHWS = Hot Water Soluble.

Based on results, alkalization and bleaching method provides best result on the composition of sorghum bagasse fiber obtained. Cellulose content from this method reaches 91.1% and this result is better than enzymatic method

that only produces 71.09%. On the other hand, reducing of lignin content on the alkalization and bleaching method was much more achieved than the alkalization method. Lignin content was reduced from 33.07% to 1.97%. This decrease is due to the sodium chlorite used in the bleaching method will reduce the content of lignin which is not eliminated in the alkalizing process.

3.2. Effect of fiber size on mechanical properties of PLA

The size of the cellulose fiber obtained is shown in Fig. 1. The smallest size is about 400 nm and the largest size is about 1500 nm.

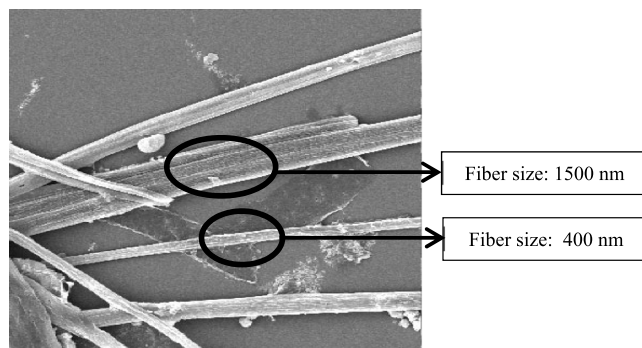


Fig. 1. Morphology of cellulose fibers (350x magnification).

The size of cellulose fiber will affect to the mechanical strength of PLA. Effect of fiber size on mechanical properties of biocomposites are shown on Table 2.

Table 2. Mechanical properties of biocomposites in different fiber size.

Sample	Young's modulus (GPa)	Tensile strength (MPa)	Strain at break (%)
PLA	0.95 ± 0.005	26.21 ± 0.91	5.19 ± 0.51
PLA + glycerol	0.73 ± 0.012	17.72 ± 0.31	11.64 ± 0.40
PLA + 10% microcellulose + glycerol	0.97 ± 0.10	6.98 ± 0.31	7.68 ± 0.18
PLA + 10% nanocellulose + glycerol	1.61 ± 0.062	15.68 ± 1.3	8.2 ± 0.14

Based on experimental results, the addition of glycerol and 10% micro-sized cellulose into PLA increased the Young's modulus of 0.97 GPa. Moreover, the best increase of Young's modulus of 1.61 GPa was achieved when glycerol and 10% of nano-sized cellulose was added. In addition, higher tensile strength was also achieved on nanocellulose fiber compared to the microcellulose fiber. This result is in accordance with previous researcher, which produce higher tensile strength using fiber size of 250 nm compared with fiber size 500 nm [11].

Based on results, it is also known that the usage of smaller fibers size will give a greater mechanical properties of PLA. This is due to on larger fiber size, the internal interaction is weak and an ability of a larger fiber size to withstand the load transferred from the matrix is low [11]. Besides, the smaller fiber size of cellulose will give a better interfacial adhesion between fiber and matrix. Therefore, it will give a greater surface area for the binds of fiber and matrix.

3.3. Effect of fiber concentration on mechanical properties of PLA

The mechanical properties of PLA can be increased by the addition of nanocellulose fibers. In this experiment, different concentrations of nanocellulose fiber has been added to PLA. The effect of fiber concentration on the mechanical strength of PLA is shown in Table 3.

The addition of nanocellulose fibers will strengthen the Young's modulus of biocomposite. Based on result, Young's modulus was increased to 2.03 GPa from 0.73 GPa as the increasing of nanocellulose fiber. Increased of Young's modulus is caused by an increase in molecular interactions between hydrogen, hardening effects, and also biocomposite crystallinity [1]. Besides, the tensile strength of biocomposite showed an increase from 17.72 MPa to 22.24 MPa at 2.5% of nanocellulose fiber concentration. Meanwhile, when the concentration of nanocellulose

Table 3. Mechanical properties of biocomposites in different fiber concentration.

Sample	Young's modulus (GPa)	Tensile strength (MPa)	Maximum strain (%)
PLA	0.95 ± 0.005	26.21 ± 0.91	5.19 ± 0.51
PLA + glycerol	0.73 ± 0.012	17.72 ± 0.31	11.64 ± 0.40
PLA + 2.5% nanocellulose + glycerol	1.29 ± 0.18	22.24 ± 3.27	11.59 ± 0.16
PLA + 5% nanocellulose + glycerol	1.39 ± 0.056	17.66 ± 0.62	10.51 ± 0.40
PLA + 10% nanocellulose + glycerol	1.61 ± 0.062	15.68 ± 1.3	8.2 ± 0.14
PLA + 15% nanocellulose + glycerol	2.029 ± 0.16	11.31 ± 1.2	6.66 ± 0.06

was increased by 15%, the tensile strength of the biocomposite was decreased. This is due to the agglomeration of cellulose caused by the Van der Waals force [12].

3.4. Effect of fiber concentration on biodegradability of biocomposites

Biodegradability of biocomposites represented by mass reduction are shown in Table 4. Based on results, biodegradability of biocomposites were better when the cellulose concentration was increases. This is due to the crystallinity of the biocomposite was also increases. Based on results, the best biodegradability of biocomposites shown when the cellulose concentration was 15% with mass reduction reach 46.4%.

Table 4. Mass reduction of biocomposites.

Sample	Biodegradability within days-								
	0	7	14	21	28	35	42	49	56
PLA	0	0,77	1,93	2,32	2,71	3,70	6,28	9,21	12,37
PLA + 2.5% nanocellulose	0	0,42	0,87	2,31	4,06	5,34	8,30	12,30	17,30
PLA + 5% nanocellulose	0	0,35	1,78	3,12	4,79	7,12	12,92	18,98	24,90
PLA + 10% nanocellulose	0	0,28	1,26	2,77	4,35	8,56	15,27	22,87	31,28
PLA + 15% nanocellulose	0	0,43	1,64	4,19	8,04	13,77	22,18	33,29	46,40

4. Conclusion

Mechanical properties and biodegradability of biocomposites made from PLA and cellulose fiber from sorghum bagasse has been improved. Smaller size of cellulose fiber was shown better mechanical properties. Addition of 2.5% nanocellulose fiber, improved the maximum strain from 5.19% to 11.59%. The best biodegradability of biocomposite was achieved on the addition of 15% nanocellulose fiber.

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