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Tensile Strength and Thermal Resistance Analysis of Polylactic Acid (PLA) and Cassava Starch with Cellulose Paper Sugarcane Bagasse as Filler

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ABSTRACT

At this time, the material made from plastic is widely used because it has strong and inexpensive properties. However, the amount of plastic waste is an environmental problem in every region. One way to overcome is to create environmentally friendly plastics that can decompose naturally. The purpose of this study was to prepare and investigate the tensile strength, thermal resistance and scanning electron microscopy (SEM) morphology observations of polylactic acid (PLA) and cassava starch (CS) with cellulose paper from sugarcane bagasse (CPSB) as filler. The fabrication process uses the solvent casting method with constant PLA and CS concentrations of 80% and 20% with variations in the CPSB mass fraction of 0.5%, 1%, 1.5%, and 2%. SEM observations showed that the mixture of PLA and CS had poor bonding due to the different hydrophobicity of the two materials. The content of CS causes agglomeration. The addition of CPSB reduces the tensile strength from 21.96 to 16.56 MPa and the modulus of elasticity from 96.75 to 74.90 MPa. The thermal resistance of the composite mixture increases with increasing CPSB load.

1. INTRODUCTION

Along with technological advances and the increase in global population requires the discovery of new materials that are environmentally friendly which are good for use in various applications. Currently, the most widely developed environmentally friendly material is bioplastic. However, most conventional plastics such as polyethylene (ethene), polypropylene (propene), polystyrene, poly vinyl chloride (PVC), and polyethylene terephthalate (PET) are not biodegradable, and the increasing number of these types of plastics in the environment has become a threat to the planet [1]. To overcome this problem, several steps have been taken, starting from reducing the use of plastics and replacing the basic materials that make up plastic with environmentally friendly materials such as biodegradable plastics or biocomposite [2].

Biocomposites have considerable advantages such as high strength, low density, renewability, biodegradability and cost reduction. It is composed of two main materials, namely matrix and fiber. The type of fiber that is commonly used is natural fiber because it is strong and competitive compared to synthetic fibers [3]. Natural fibers have three important parts such as cellulose, hemicellulose and lignin. From the three parts, cellulose is a component that affects the properties of biocomposites. Alkalization and bleaching treatments help to increase the cellulose content in plant fibers [4]. The combination of the matrix and natural fibers can improve the mechanical properties of the biocomposite. It was reported by previous studies that the increase in mechanical properties is due to the good bond between matrix and fiber [5].

Several biodegradable matrices have been studied in biocomposites such as polylactic acid [5], polyvinyl alcohol [6], chitosan [7], starch [8], and other biopolymers [9]. From the existing biopolymer matrix, PLA is a candidate matrix material that has the potential as an alternative to environmentally friendly packaging products. This is due to the competitive mechanical properties and thermal resistance compared to synthetic polymers [5]. However, PLA is a material that has an expensive price. Therefore, the addition of starch is recommended to reduce production costs as reported by previous researchers [10].

Mixing of PLA and starch causes a decrease in mechanical properties. This was due to poor interaction between PLA and starch in the form of starch agglomeration around the PLA matrix [11]. One way to improve the properties of the biocomposite is by means of a hybridization process by adding fillers in the form of natural fibers. Natural fiber is one of the important components to improve the mechanical and thermal properties of biocomposites [10]. One candidate for natural fiber as reinforcement in biocomposites is sugarcane bagasse fiber. Sugarcane bagasse fiber has a cellulose content of about 80% [8]. This content can produce a good interaction between the matrix and the fiber.

Several previous studies reported that the addition of natural fibers such as pineapple leaf fiber [12], water hyacinth [13], and oil palm empty fruit bunches [14] into the matrix increased the tensile strength of the biocomposite. Previous researchers also reported that the addition of 3% betel leaf fiber which was treated with alkalization could increase the tensile strength of the biocomposite film [15]. In this study, the researchers studied the tensile strength characteristics, fracture morphology, and thermal resistance of PLA/cassava starch reinforced with sugarcane bagasse cellulose fibers in paper form. According to the researcher's knowledge, there have been no studies reporting on this object.

2. MATERIALS AND METHODS

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2.1 Materials

The PLA used in this study was in the form of pellets (PL-2000) (density 1.21 g/cm^3) produced by Miyoshi Oil & Fat Co., Ltd. (Japan). Sugarcane bagasse fiber was obtained from ice seller in Jember. The sodium hydroxide (NaOH), chloroform and distilled water (pro analysis) were obtained from CV. True Prosperity. Cassava starch as a matrix was produced commercially under the brand name "Cap Pak Tani". Cassava starch contains 19% water, 15% amylose, and 85% amylopectin.

2.2 Preparation of Cellulose Paper from Sugarcane Bagasse (CPSB)

Sugarcane bagasse was supplied by ice seller in Jember and cleaned with water, then dried under the sun for 3-5 days. Alkalization process was carried out by soaking 10 g of dry palm fiber in 6% NaOH solution for 90 minutes at room temperature. After that, the fibers were washed using distilled water until the pH was neutral. Furthermore, it was dried in drying oven for 24 hours at 60 ^{o}C. Then, it was mashed using a blender and sieved with sizes of 80 and 100 mesh, in order to obtain an average size of 177 µm of sugarcane bagasse powder.

2.3 Manufacturing of Biocomposite PLA/CS/CPSB

The method of manufacturing biocomposite was solvent casting. Chloroform was used as a solvent for dissolving PLA. PLA was dissolved by using chloroform in a glass beaker for 120 minutes. It was covered by aluminum foil to cover on the beaker glass to prevent the evaporation of the chloroform. After the PLA was completely dissolved, CS and CPSB were added with variation of mass fraction. They were stirred using a magnetic stirrer for 20 minutes with a rotating speed 150 rpm at 50-60 ^{o}C. After 20 minutes, the aluminum foil was unscrewed and let the mixture sit for 5 minutes to release any air bubbles formed during the stirring process (air bubbles were lifted along with the evaporation of the chloroform). The biocomposite mixture was poured into the rectangular mold and dry for 24 hours in a zip lock plastic.

2.4 Tensile Test

Tensile test was carried out using the Universal Testing Machine HT-2328. The tests were carried out according to ASTM D882 standard at room temperature with a strain rate of 1 mm/min on all specimens.

2.5 Scanning Electron Microscope (SEM) Observations

Observation of the fracture surface of the bicomposite using Scanning Electron Microscopy (SEM) Hitachi TM3030 Plus model with a voltage of 5 kV. Observations were made under vacuum with a secondary electron signal for imaging processes with 1000x and 1500x magnification. The fracture surface was cut into 5 x 15 mm and observed.

2.6 Thermogravimetric Analysis (TGA)

The TGA test was carried out by using the TGA Q500 V20.6, USA. The sample weighed between 4 and 5 mg. The scans were run from room temperature to 600 ^{o}C at a heating rate of 10 ^{o}C/min under a nitrogen flow of 25 ml/min

3. RESULTS AND DISCUSSIONS

3.1 Tensile Properties

In terms of tensile strength, pure PLA has excellent strength comparable to synthetic polymers. However, due to the high price and complex production process, pure PLA is not suitable for food packaging applications. To reduce costs, cassava starch was chosen for the PLA mixture. The ratio of cassava starch mixture as much as 20% of the biocomposite mixture.

Figure 1 displays the tensile properties of PLA and its biocomposite film. From the tensile test data, it can be seen in Figure 1a the tensile strength value of the biocomposite decreased significantly with the addition of cassava starch as a biocomposite. The tensile strength of pure PLA is 20.99 MPa and in a mixture of PLA with starch 17.42 MPa. This phenomenon is probably due to the presence of a number of free OH bonds between PLA and starch. Free OH affects the low compatibility between PLA and cassava starch [16]. However, the value of the tensile strength does not decrease drastically with the increase in the mass of fiber. At the addition of 0.5% fiber, the tensile strength was 16.56 MPa. Meanwhile, the addition of 2% fiber, the tensile strength was 15.31 MPa. According to previous report, the polymer blends with high starch content produce in partial agglomeration resulting in poor interfacial adhesion between PLA and starch. These results decrease in tensile strength due to the brittleness of cassava starch [17].

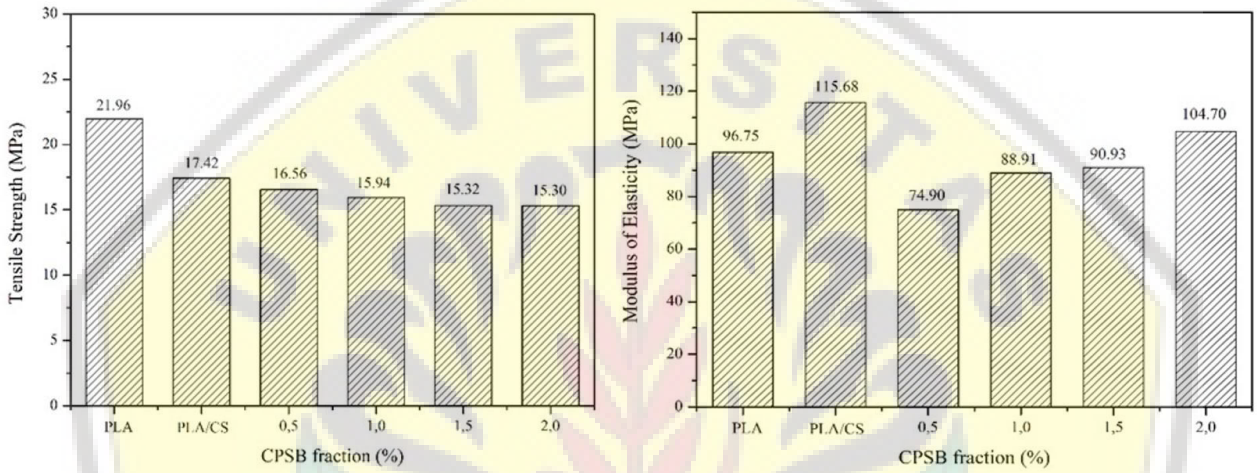

Figure 1. Tensile properties of all samples tested: (a) tensile strength and (b) modulus of elasticity

The mixture of PLA and starch is obtained more rigid which can be determined in modulus of elasticity (Figure 1b). Modulus of elasticity (ME) of each mixture is different in the range of 74 to 115 MPa. The ME increased significantly with the addition of starch content for 115.68 MPa. The results show that starch can provide stiffness to the biocomposite mixture [10]. However, the mixture containing 0.5% fiber reduces the stiffness value by 54%. The addition of 0.5% and 2% fiber in PLA/CS matrix has a modulus value of 74.90 MPa and 104.70 MPa, respectively. The results showed that the addition of fiber concentration increased modulus of elasticity. This phenomenon can be explained by the good hydrogen bonding, hardening effect and high crystallinity index of fillers in bicomposite matrix [18].

3.2 Fracture Morphology

Figure 2 shows the SEM observations on fracture surface all biocomposites after tensile test. Figure 2a shows the fracture morphology of bicocomposite PLA/CS with 0.5% CPSB. It can be seen that the morphology was rather rough and minim porosity. This proves that the matrix transfers tensile stresses to the fiber effectively due to a good bond between matrix and fiber. A similar case was also reported by previous researchers that the addition of fiber concentration below 5% resulted in a rough surface and good interaction [15].

Meanwhile, Figures 2b displays the fracture surface of biocomposite PLA/CS with 1% CPSB. Some points of the surface indicate the presence of voids. This is due to the poor mixing of PLA, CS and CPSB in the fabrication process [19]. The poor affected in poor interaction between matrix and fiber. This is proven and reinforced by the tensile test results due to a decrease in the value of the tensile strength. Previous studies

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also reported that the presence of voids in the composite can reduce the tensile strength [19]. Furthermore, Figures 2c and 2d show a smooth fracture surface, agglomeration and porosity. As previous report, the agglomeration and porosity reduce the mechanical properties of biocomposites [20]. Starch granules and fibers are irregularly distributed in the matrix indicates the mismatch of starch composition in the PLA-based polymer mixture [16]. This causes the strength to be reduced due to weak interface adhesion. This is attributed to differences in the hydrophobicity of the matrix and filler [21].

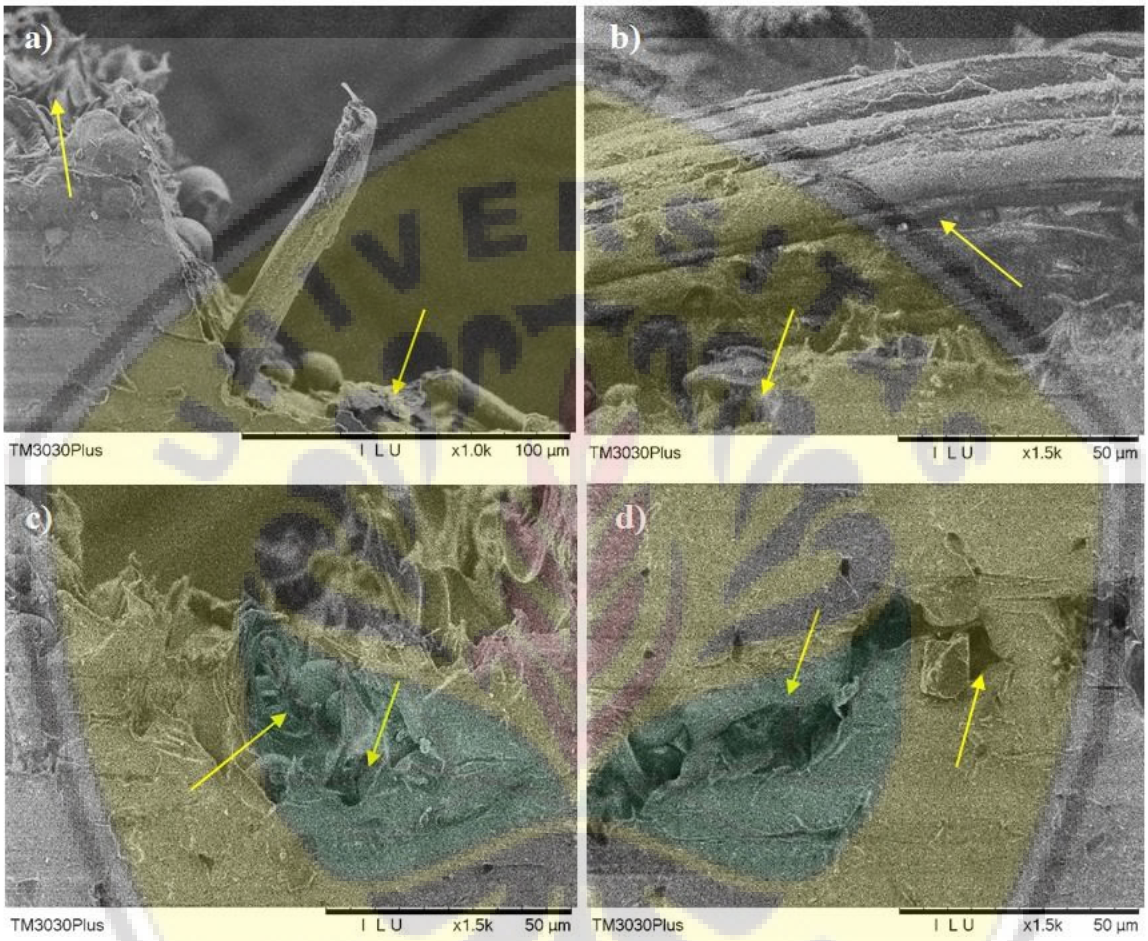

Figure 2. Fracture morphology of biocomposite based PLA/CS with: (a) 0.5%, (b) 1%, (c) 1.5%, and (d) 2% of CPSB

These differences in properties hinder the compatibility between the matrix and the fiber. Several studies have exposed the similar problem and suggested several solutions such as modifying the starch structure through silylation process to improve compatibility with PLA [22].

3.3 Thermal Resistance

Thermogravimetric (TGA) analysis was carried out to investigate the thermal resistance of the CPSB filled PLA/CS composites. Figure 3 shows the TG curve for weight loss with increasing temperature. The initial step of thermal degradation occurs below 100 ^{o}C and is associated with the evaporation of water molecules from the sample [17]. This phenomenon occurs for every sample including pure PLA. With further heating showed the lowest degradation peak at 270 ^{o}C caused by evaporation of chemically adsorbed water molecules. In addition, the highest peak at 300 ^{o}C. This is due to the oxidation of partially decomposed starch and fiber degradation [20].

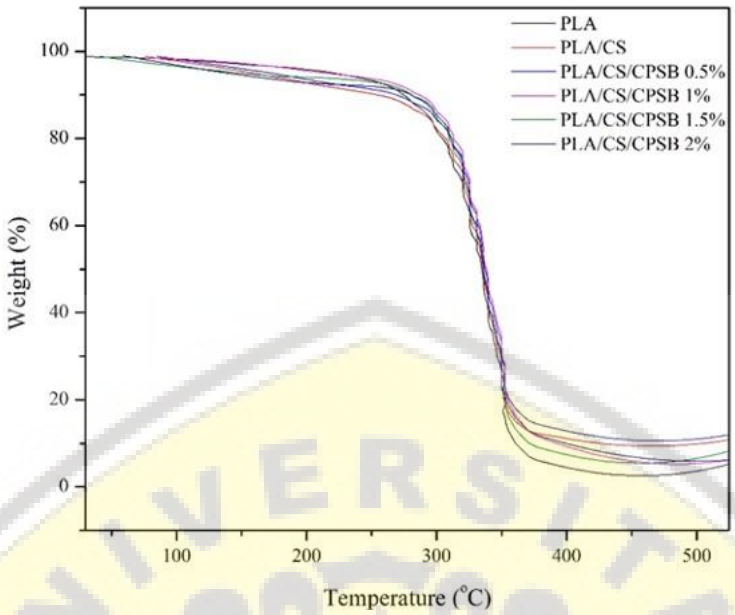

Figure 3. TGA curve of all biocomposite tested

The TGA curves show the peak of degradation ending at 420 °C. The weight loss for samples containing CPSB was lower than for pure PLA. This phenomenon is in line with research conducted by previous report [17], [20]. Another factor that can improve the thermal stability of biocomposites is the relatively good interfacial bond between CPSB and PLA/CS which causes strong hydrogen bonds, thereby reducing weight loss in the sample. This bonding contributes to the restriction of matrix chain mobility, which indirectly increases the thermal stability of the palm fiber reinforced PLA/Thermoplastic Starch polymer [17].

4. CONCLUSIONS

Biocomposite based PLA/Cassava starch filled by CPSB was produced by solvent casting method at room temperature. The addition of CPSB in PLA/starch matrix did not increase the tensile strength of the biocomposite. The highest tensile strength was in pure PLA film for 21.96 MPa. The addition of 0.5% fiber has the highest tensile strength value among other biocomposites. Fracture morphology also showed the presence of a rough surface indicating a good bond between the matrix and fiber. The thermal resistance value of biocomposite based PLA/CS filled by CPSB before degradation at 270 °C and decomposed at 320 °C. This biocomposite may be suitable for application as an environmentally friendly food packaging film.

5. ACKNOWLEDGMENTS

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