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Mechanical Characteristics of Stearic Acid Addition in Polylactic Acid (PLA) and Cassava Starch Bioplastic Blends

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Abstract. *This study aims to determine the effect of adding stearic acid (SA) to a bioplastic mixture of cassava starch (CS) and polylactic acid (PLA). The bioplastic was produced using a solvent casting method. The addition of SA can affect the mechanical properties of the film. The maximum tensile strength of the film increased from 5.12 MPa (without SA) to 7.61 MPa (5% SA). The same trend also applies to the Young's modulus and elongation at break, which increased from 25.45 MPa and 20.17% to 35.02 MPa and 21.64% after the addition of 5% SA. This improvement in mechanical properties is supported by the compatibility of PLA and CS due to the optimal presence of SA. These findings prove that SA is an effective compatibilizer in improving the mechanical properties of PLA and CS-based bioplastics. The resulting film products have the potential to be used as environmentally friendly packaging materials that are competitive with synthetic materials such as Low-Density Polyethylene (LDPE) and Ethylene Vinyl Acetate (EVA).*
Keywords: *Polylactic Acid; Cassava Starch; Stearic Acid; Bioplastic; Mechanical Properties.*

1. Introduction

The use of synthetic plastics as packaging causes various problems, especially environmental pollution [1]. This is because synthetic plastics have resistance to natural degradation making it difficult to decompose [2]. The use of similar but more environmentally friendly replacement materials is urgently needed. Bioplastics or commonly called biodegradable plastics are the right alternative material in this problem. Bioplastics are generally divided into several types such as polylactic acid (PLA), Polyhydroxybutyrate (PHB), poly-ε-caprolactone (PCL), and so on [3]. However, PLA is one of the most desirable types of bioplastics because it has mechanical properties that meet the quality standards of conventional plastics in general, such as PP and PET [4].

PLA as a bioplastic still has weaknesses, and among the most important is the high level of brittleness [5]. The use of blended materials is necessary to improve the weakness of PLA. Cassava starch (CS) is one of the suitable candidates because it can maintain flexibility, low cost,

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and biodegradability [6,7]. The use of cassava starch as a PLA blend material has been explored by several researchers. The addition of cassava starch to PLA at a concentration of 45% was reported to increase the elongation at break of the film by 7.63%, although it had a negative effect on its tensile strength [8]. A mixture of cassava starch and glycerol for thermoplastic starch (TPS) as a PLA filler was reported to have high water absorption due to the hydrophilic nature of the filler [9]. Tensile strength was also reported to increase with a maximum cassava starch content of 30% in PLA [10]. The combination of PLA and cassava flour accelerates the rate of biodegradation because of the amorphous and hydrophilic properties of the flour [11]. A decrease in mechanical properties can also occur for the same reason, namely poor compatibility between cassava starch and PLA when mixed [12].

The addition of compatibilizer agents is needed to support the compatibility between cassava starch and PLA. To date, stearic acid has received more attention from authors for its ability as an effective compatibilizing agent in its application to biopolymer composites. Several studies have reported the use of stearic acid as a compatibilizer agent such as the one conducted by Meng et al. [13]. He reported that corn starch treated with stearic acid can be evenly dispersed and compatible with PLA/PBSA matrix. The addition of stearic acid can increase the tensile strength and elastic modulus of propylene/silica nanocomposites [14]. Elongation at break was reported to increase by 37% in polypropylene/bentonite composites when stearic acid was added [15]. An increase in tensile strength also occurred in polypropylene (PP)/recycled acrylonitrile butadiene rubber (NBRr)/ empty fruit bunch (EFB) composites. The main contributing factor is the treatment of EFB fibers with stearic acid (SA). As a result, the carboxyl group of SA bonds with the hydroxyl group of cellulose (hydrophilic) which can interact with the polymer matrix (hydrophobic) [16].

From some of these literature studies, stearic acid has great potential as a good compatibilizer agent for starch and PLA-based bioplastics. Therefore, an experimental approach is very interesting to do regarding the use of stearic acid in bioplastic applications. The purpose of this study is to analyze the effect of stearic acid addition with different contents (0, 5, 10, 15, dan 20 wt%) on the mechanical and morphological properties of PLA/CS bioplastics. In this study, it is expected that stearic acid can be an effective compatibilizer agent for PLA/CS in order to support mechanical properties that are competitive with synthetic plastics.

2. Materials and Methods

2.1. Materials

PLA PL-2000 (density 1.26 g/cm^3) was purchased from Miyoshi Oil & Fat Co., Ltd. Cassava starch (99% purity) was purchased from Intelligent Materials Pvt, Ltd. $CHCL_3$ Pro

analysis (99% purity) purchased from CV. Makmur Sejati. Stearic Acid (MERCK 800673) Pro analysis was purchased from Shagufta Lab Scientific.

2.2. Bioplastic Film Preparation

Bioplastics were made using the solvent casting method as schematized in Fig. 1. PLA was dissolved in a glass beaker using 25 ml chloroform for 120 minutes covered with aluminum foil. Cassava starch (CS) and stearic acid (SA) were added according to sample variation, where the ratio of CS and PLA was kept constant and SA was varied as shown in Table 1. The mixture suspension was then stirred using a magnetic stirrer for 20 minutes at a temperature of 50-60°C at a speed of 150 rpm. The PLA/CS/SA suspension was allowed to stand for 5 minutes by opening the aluminum foil to remove bubbles. The suspension was then poured into a glass mold. Drying is carried out in a ziplock plastic bag for 24 hours at room temperature to minimize voids in the sample.

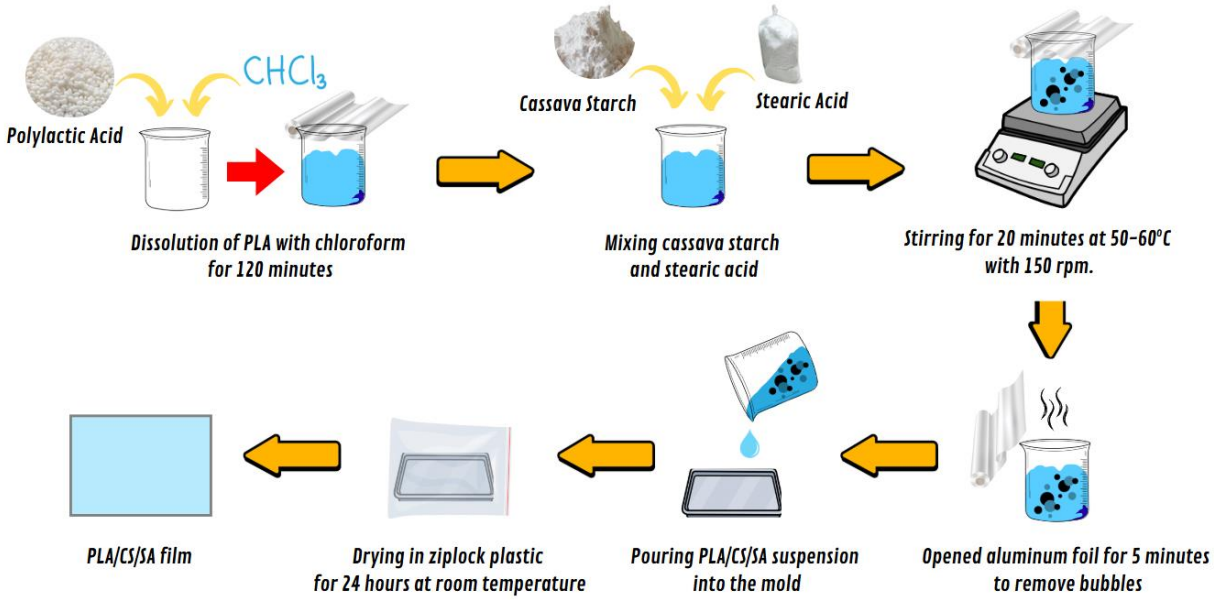

Fig. 1. Bioplastic Film Fabrication Scheme

Table 1. Bioplastic Film Composition

Sample Code	Mixture		
	PLA (wt%)	Cassava Starch (wt%)	Stearic Acid (wt%)
PLA/CS	60	40	0
PLA/CS/SA5	60	40	5
PLA/CS/SA10	60	40	10
PLA/CS/SA15	60	40	15
PLA/CS/SA20	60	40	20

2.3. Tensile Test

Tensile testing of all samples was carried out according to ASTM D882-18 standard with

length × width dimensions of 130 × 12 mm each and thickness ≤ 1mm. The tensile tester used was HT-2328 Universal Testing Machine with a speed of 10 mm/minute.

2.4. Scanning Electron Microscopy (SEM)

Scanning Electron Microscopy was used to observe the fracture morphology of all PLA/CS/SA samples. This observation was carried out using a Hitachi SU3800 type SEM instrument with a magnification of 3000-10000x.

3. Results and Discussion

3.1 Tensile Properties

Fig. 2 shows a graph of the effect of SA on the mechanical properties of PLA/CS and PLA/CS/SA. The mechanical properties ($p < 0.05$) shown include tensile strength, Young's modulus, and elongation at break. In addition, the values of each tensile test are presented in Table 2.

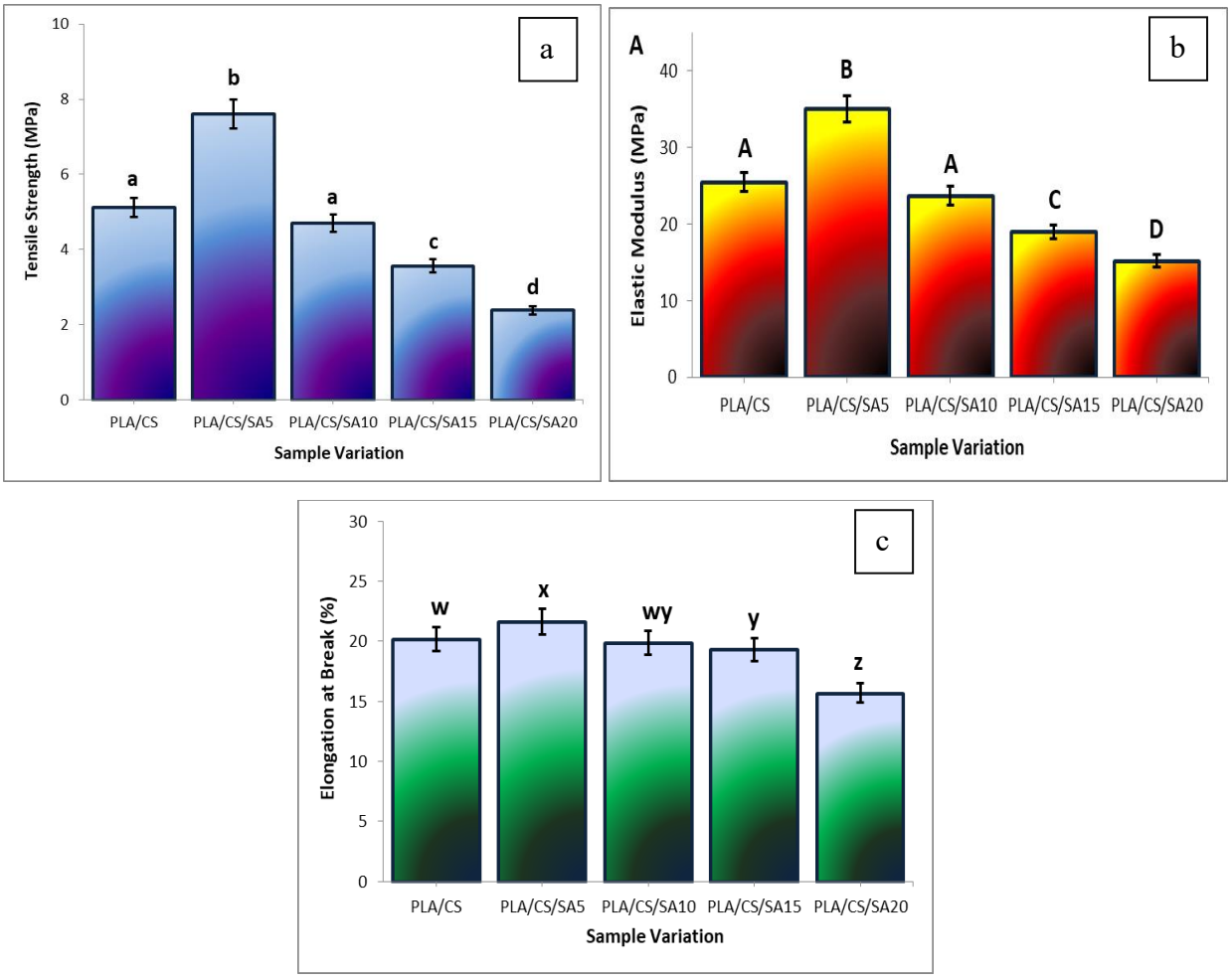

Fig. 2. Mechanical Properties: a) Tensile Strength, b) Young's Modulus, and c) Elongation Break

Table 2. Tensile Testing Value of Bioplastic

Sample Code	Tensile Strength (MPa)	Young's Modulus (MPa)	Elongation at Break (%)
PLA/CS	5.12±0.05^{a}	25.45±0.16^{A}	20.17±0.01^{w}
PLA/CS/SA5	7.61±0.29^{b}	35.02±0.06^{B}	21.64±0.03^{x}
PLA/CS/SA10	4.70±0.24^{a}	23.69±0.08^{A}	19.84±0.08^{wy}
PLA/CS/SA15	3.56±0.14^{c}	18.94±0.24^{C}	19.31±0.06^{y}
PLA/CS/SA20	2.38±0.06^{d}	15.17±0.02^{D}	15.69±0.04^{z}

Significant differences ($p < 0.05$) are indicated by the notation (a,b,c,d) for TS, (A,B,C,D) for YM, and (w,x,y,z) for EB.

In Fig. 2a, the tensile strength of PLA/CS film has a value of 5.12 MPa. The maximum increase in tensile strength occurred at the addition of 5% SA, which is strongly suspected to be caused by the even dispersion of CS because of the presence of SA [17]. Another study said, stearic acid helps in improving the compatibility between the polymer and the filler thus forming a strong adhesion between the two [18]. The addition of SA as a calcium carbonate coating has similar results, showing the highest tensile strength of LDPE-based composites [19]. This is better than lecithin as a compatibilizer agent in PLA and starch-based composites, which actually reduces tensile strength [20]. A decrease in tensile strength occurred in PLA/CS/SA10, PLA/CS/SA15, and PLA/CS/SA20, respectively. This is attributed to the high SA content which weakens the PLA/CS structure [21]. The elastic modulus of PLA/CS had a value of 25.45 MPa, and increased to 35.02 MPa when 5% SA was added. This result corresponds to the increase in tensile strength indicating the success of SA in forming a compatible PLA and CS structure. Unlike other compatibilizer agents such as maleic anhydride polyethylene (MAPE), which actually reduces the elastic modulus value when added at 2-6 wt% to PLA composites filled with rice husk fibers [22].

The decrease in elastic modulus is also similar to that of tensile strength, which is significantly lowest in PLA/CS/SA20 (Fig. 2a). This decrease is thought to be caused by the high SA content, which weakens the PLA/CS structure. This creates a mismatch in the distribution of the filler to the matrix, reducing the interface interaction between the two. Elongation at break as shown in Fig. 2c also shows a similar trend where the highest value is achieved by PLA/CS/SA5. The decrease in elongation is believed to be caused by the high SA content, which forms phase separation with the polymer matrix, resulting in an uneven microstructure and weak points in the film. The decrease in elongation at break is consistent with the results of other similar studies [23]. The addition of optimal compatibilizing agents generally also increases the elongation at break of bioplastic films, as is the case with citric acid and maleic anhydride, which play a positive role in increasing the elongation at break of PLA- and starch-based films [24].

3.2 Fracture Surface Morphology

Fig. 2a shows the fracture surface of the PLA/CS sample where agglomeration of CS particles was identified. This agglomeration occurs due to the absence of SA as a compatibilizer agent that keeps CS evenly distributed in the PLA matrix [25]. Another study also reported that the addition of cassava starch to other polymers in the form of polyvinyl alcohol (PVA) caused imperfect bonding between the two [26]. On the other hand, the addition of 5% SA showed good adhesion between CS and PLA on the fracture surface (Fig. 2b). This confirmed the improvement of mechanical properties including tensile strength, elastic modulus, and elongation at break in PLA/CS/SA. The distribution of ZnO particles in the HDPE matrix was also reported to be homogeneous with good interaction resulting from surface treatment with SA [27]. Fig. 2c shows the agglomerated surface of PLA/CS/SA20, which occurs from SA accumulation because of high content [28]. In addition, the phenomenon of porosity was also identified on the film surface, indicating poor adhesion between CS and PLA due to high SA content. A similar phenomenon was also reported in a previous study, where filler aggregation (sugarcane bagasse fibers) and porosity based on SEM observations resulted in a 41.9% decrease in tensile strength of PVA-based biocomposites compared to the control film [29].

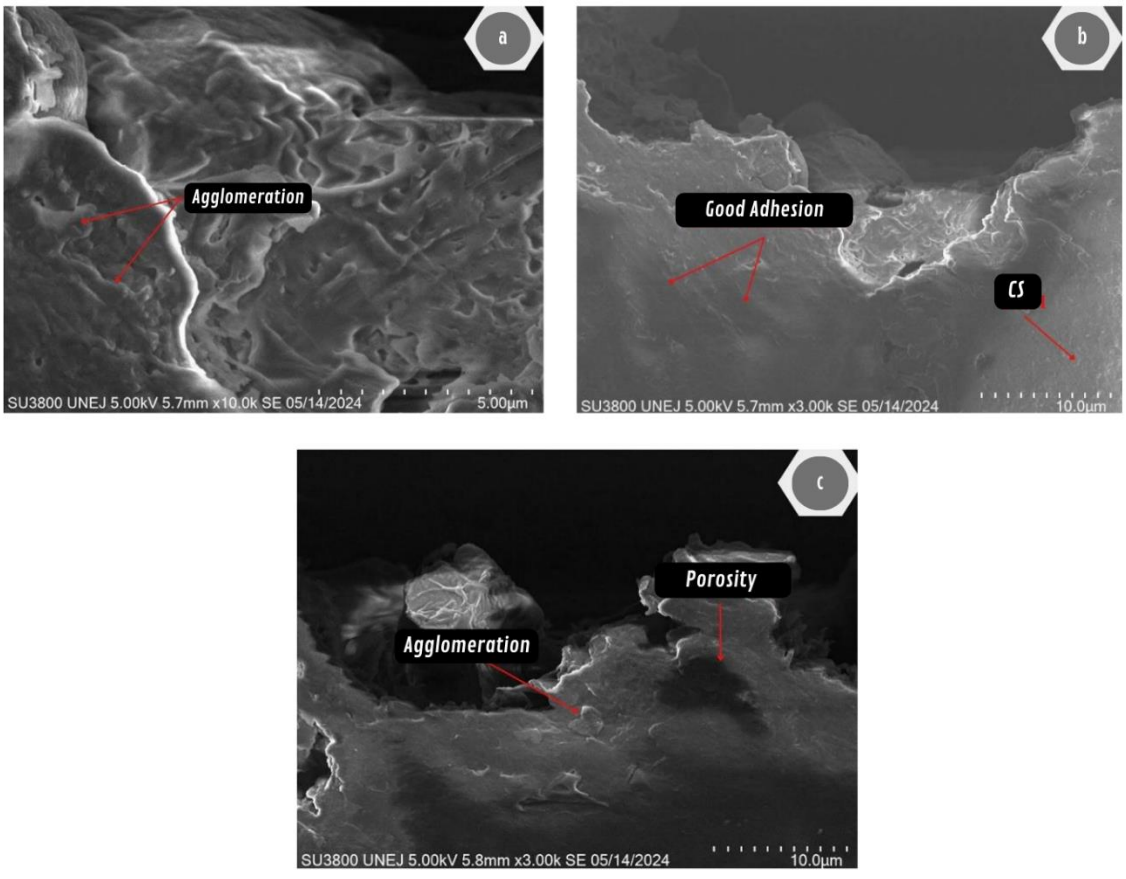

Fig. 3. Surface Morphology Observation of Bioplastic by SEM with variations of: a) PLA/CS, b) PLA/CS/SA5, c) PLA/CS/SA20

4. Conclusions

Bioplastic in the form of film from a mixture of PLA, CS, and SA can be made using the solvent casting method. The addition of SA affected the mechanical properties of PLA/CS/SA films. The tensile strength, young's modulus, and elongation at break increased maximum at the addition of 5% SA by 7.61 MPa, 35.02 MPa, and 21.64%, respectively. These results were supported by SEM which showed good adhesion between PLA and CS. Higher SA content in the film caused a decrease in mechanical properties due to agglomeration and porosity of the film. Bioplastic films with the right mixture in this study have the potential to compete with synthetic packaging and are certainly more environmentally friendly. The application of this film packaging product can be focused on dry food packaging, and further testing on food itself needs to be conducted in future research.

Data availability statement

Your data availability statement should describe how the data supporting the results reported in your paper can be accessed. If your data are in a repository, include hyperlinks and persistent identifiers (e.g. DOI or accession number) for the data where available. Example: Data will be made available on request.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgement

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