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MECHANICAL TEST AND THERMAL STABILITY ON THERMOPLASTIC SAGO (*Metroxylon sagu* Rottb.) COMBINATION OF POLYETHYLENE AND POLYPROPYLENE

S. Munte[1], M. Banjarnahor[1], D.A. Tanjung[2,✉] and R.S. Budi[3]
[1]Department of Industrial Engineering, Faculty of Engineering, Universitas Medan Area, Medan-20223, Indonesia
[2]Department of Agrotechnology, Faculty of Agriculture, Universitas Medan Area, Medan-20223, Indonesia
[3] Department of Agrotechnology, Faculty of Agriculture, Universitas Islam Sumatera Utara, Medan-20217, Indonesia
✉Corresponding Author: dennyakbartanjung@gmail.com

ABSTRACT
Polyethylene is a polymer that has good elasticity properties, while polypropylene is a polymer with a high tensile strength value. Thus, the combination of these two polymers is expected to increase the value of tensile strength and elasticity. A biocomposite consisting of a mixture of Polyethylene-Polypropylene and thermoplastic sago starch (TPSS) was successfully prepared using the mixing method in an internal mixer. TPSS is made by reacting starch and glycerol with a composition of 65:35 wt.%. Compatibility was made by reacting Polyethylene-Polypropylene: Maleic anhydrate: Benzoyl peroxide with a composition of 88:9:3 wt. %. The match concentration used was 10wt.% based on the TPSS weight. The concentration of the Polyethylene-Polypropylene mixture starts from 0.10,15,20,25,30 and 100wt.%. The characterizations carried out are mechanical properties analysis are tensile strength, elongation at break and elasticity, FTIR analysis, and thermal properties are TGA and DTA. According to the test results, the addition of a mixture of polyethylene and polypropylene can increase the tensile strength from 1.7292MPa (without a mixture of polyethylene and polypropylene) to 4.8334MPa and the elongation at break also increases from 0.23% to 1.72% at a concentration of 30wt.%. Meanwhile, Young's Modulus decreased from 740 to 281MPa at a concentration of 30 wt. %.
Keywords: Biocomposite, Compatibility, LLDPE-g-MA, TPSS, PP-g-MA.
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INTRODUCTION

Native starch has several disadvantages, including low mechanical properties[1-3], high water sensitivity[4-5], and stiffness.[6–9] It is due to starch's nature, which tends to agglomerate and create concentrated stress areas, resulting in poor mechanical properties[10] and exhibiting high water absorption. The stiffness of starch can be plasticized by modifying the chemical structure of starch into starch thermoplastic using a plasticizer. Some plasticizers that can be used are glycerol, sorbitol, and others. The addition of synthetic polymers such as LLDPE and PP can improve the mechanical properties of bioplastics.[11–13] Synthetic polymers have advantages such as high mechanical properties, resistance to high temperatures, high flow properties, ease of shape, and hydrophobic properties. The natural and synthetic blend polymers are expected to improve mechanical and physical properties which are not much different from conventional plastics. When starch sago and polyethylene//Polypropylene are mixed, the two materials are incompatible. This is caused by differences in polarity, where sago starch is polar while polyethylene/polypropylene is non-polar By including the compatibilizer, the compatibility between the two mixtures can be increased. The addition of a compatibilizer is expected to increase the homogeneity of the mixed solution. In addition, another function of the compatibilizer in polymer blends is to increase the adhesion between phases.[13-14] Several studies that have been conducted related to the use of sago starch as a basic material for making bio-plastics state that the addition of LLDPE or PP to the TPPS

mixture with the aid of a compatibilizer can improve the tensile strength up to 4.31MPa at 30% LLDPE concentration.[15-16]

In another study adding polypropylene to the thermoplastic sago starch can aid increase the mechanical properties by 5.51MPa at 30% Polypropylene concentration.[15] Based on the two studies above, the researcher wants to continue research by combining two synthetic polymers (LLDPE and PP) in sago-based bio-plastics. Therefore, the purpose of this study was to the effect of polyethylene –polypropylene blend on mechanical properties.

EXPERIMENTAL

Material and Methods

Sago starch Parang brand is a production of Warna Jaya Indonesia. Obtained from the traditional market of Serpong-Tangerang, Indonesia. Moisture content was 14%. LLDPE UF 1810S1 pellets are obtained from PT. Chandra Asri Petrochemical Tbk (TPIA) Cilegon-Indonesia. The Density of the Polypropylene Pellet is gained from PT. Chandra Asri, Indonesia. Maleic anhydride for synthesis Merck KGaA, 64271 Darmstadt Germany, Benzoyl peroxide Merck for sinthesis Merck KGaA, 64271 Darmstadt Germany and Glycerol Analytical Reagent merck Univar Production of Ajax Finechem. They are obtained from the Rudang shop Medan-Indonesia.

Preparation of Thermoplastic Sago Starch

Sago starch (65 weight percent) and distilled water (900 ml) were combined in a beaker to create sago starch thermoplastics (TPSS). After that, 80,77gm. (35 weight percent) of glycerol were added to the beaker. The hot-plate magnetic stirrer is used to position the beaker, with a temperature of 100^0C for 12 min, or the compound became gelatin. Then, it was dried in an oven for 24 hours to reduce the moisture content. [17]

Preparation of Compatibilizer (LLDPE-g-MA and PP-g-MA)

The compatibilizer was made by reacting 30.8 gm, LLDPE (88wt.%), 3.15gm, Maleate Anhydrate (9wt.%), and 1.05gm Benzoyl Peroxide (3wt.%). All materials were reacted in an internal mixer at a temperature of 160^0C for 12 minutes at a speed of 100 rpm. The same treatment was carried out for PP.[17]

Preparation of TPSS/PE-PP/Compatibilizer

TPSS/LLDPE-PP/Compatibilizer blends were made with concentration variation LLDPE-PP of 0,10,15,20,25,30, and 100wt.%. The compatibility content used is 10wt.% based on TPSS weight. Detailed data can see in Table-1.

Table-1: Amount Ingredients of Bioplastics Making

LLDPE-PP Content(%)	TPSS(g)	LLDPE(g)	Compatibility LLDPE(g)	PP(g)	Compatibility PP (g)
0	33	0	0	0	0
10	29.700	0.343	1.485	0.343	1.485
15	28.050	1.240	1.402	1.240	1.402
20	26.400	2.138	1.320	2,138	1.320
25	24.750	3.036	1.237	3.036	1.237
30	23.100	3.933	1.155	3.933	1.155
100	-	16.500	-	16.500	

Compression Molding

The TPSS/LLDPE-PP/Compatibility blends were printed with a hydraulic pressure compression manual at a temperature of 160^0C for 12 minutes. Every 4 minutes, the pressure was increased to maximum. All compression molded sheets in accordance with ASTM D638 Type 1.

Mechanical Properties Test

A Universal Band Tensilon machine was used for mechanical testing with ASTM D638 Type 1 specimens. 25^0C in the room, 60% relative humidity, and 10mm/min. Three specimens were used to calculate the average tensile strength, Elongation at breaks, and Young's modulus.

Spectroscopy Fourier Transform Infra-Red (FTIR) Analysis

The instrument used is the Fourier Transform Infrared Spectroscopy-Perkin Elmer System 2000). The samples tested were TPSS and TPSS/LLDPE-PP/Compatibilizer. Background scans: 32 with a resolution:16 cm^{-1} recorded. Width: 9:5 mm. the wave number interval is 4000-650 cm^{-1}, Sampling was measured in the form of a layer with a thickness of 1mm, which was made by hot press molding.

Thermal Analysis (TGA/DTA)

STA module's thermographic analysis used the TGDTA 7300 channel. The measurement was done at 9.286 mg with a heating rate of 10^0C / minute. The heating was begun at 28^0C to 700^0C. The rate of nitrogen gas was 50 ml/min.Temperature rise 50^0C/min.

RESULTS AND DISCUSSION

Mechanical Analysis

Parameters that need to be considered in the manufacture of biocomposites are mechanical properties. Figure-1, it can be analyzed that the value of tensile strength and elongation at break increases with increasing concentration of the polyethylene-polypropylene blends. The tensile strength value from 1.7292MPa (without polyethylene-polypropylene) increased to 4.8334MPa at 30% LLDPE-PP. Meanwhile, Young's modulus decreased with increasing concentration of the polyethylene-polypropylene mixture. (Table-2 and Fig.-2). The elongation value at break was 0.23% (without LLDPE-PP) after the addition of LLDPE-PP with a concentration of 30% to 1.72%. The addition of a polyethylene-polypropylene blends had a positive effect on mechanical properties. The amount of the polyethylene-polypropylene monomer mixture is still sufficient to react with starch at a concentration of 70% to react with the aid of compatibility as a linking agent between natural polymers (sago starch) and synthetic polymers (polyethylene-polypropylene mixtures).[18-19] The addition of a mixture of polyethylene and polypropylene can increase the tensile strength when compared to without using a mixture of polyethylene – polypropylene.

Table-2: The Mechanical Properties for Composite Bioplastic and TPSS

LLDPE-PP Content(%)	Tensile Strength (MPa)	Elongation at breaks(%)	Young's Modulus(MPa)
0	1.7292	0.23	740
10	3.6839	0.74	500
15	3.7380	0.95	395
20	4.0374	1.16	347
25	4.5092	1.57	201
30	4.8334	1.72	281
100	15.7159	4.90	320

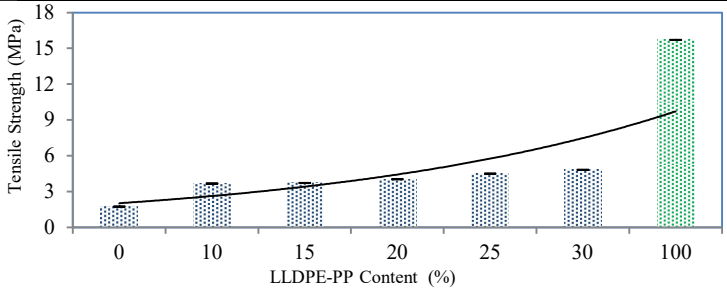

Fig.-1: Effect of LLDPE-PP Content on Tensile Strength

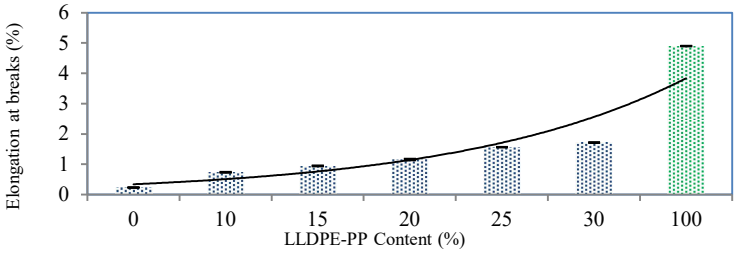

Fig.-2: Effect of LLDPE-PP Content on Elongation at Break

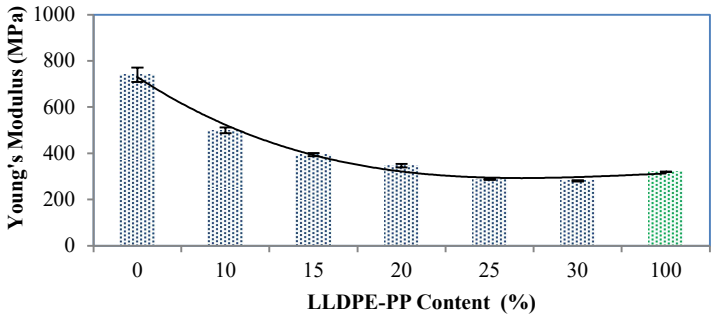

Fig.-3: Effect of LLDPE Content on Young's Modulus

FTIR Analysis

The addition of polyethylene-polypropylene blends into starch thermoplastic with the aid of a coupling agent produces a curve that is not much different from TPSS. wavenumber shift only occurs at a wavenumber of 1021cm^{-1} and is detected as a C-O group, while the other peaks detected the same wavenumber as TPSS such as the presence of a hydroxyl group (OH) with a broad peak at a wave number of 3265 cm^{-1} (Table-3 and Fig.-4), Hydrocarbon bonds (C-H) with a wavenumber of 2922cm^{-1}. Alkene group (C=C) at wave number 1647 cm^{-1} and wavenumber 1416 cm^{-1} is a CH_2 group. This wavenumber shift is proof that there has been a reaction between Starch tthermoplastic and polyethylene-polypropylene assisted by a coupling agent on oxygen bonds.[17-18,20]

Table-3: The Principal FTIR Absorption Peaks for Composite Bioplastic and TPSS

Functional Groups	Wavenumber cm^{-1}	
	TPSS	Composite Bioplastics
O-H	3265	3265
C-H	2922	2922
C=C	1647	1647
C-H_2	1416	1416
C-O	1021	998

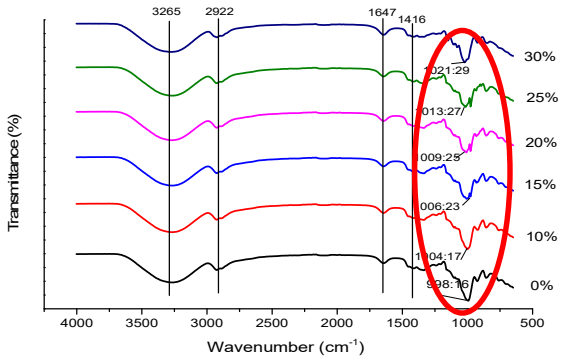

Fig.-4: FTIR Spectra of LLDPE-PP Concentration Variation

Thermal Analysis

The sample used in the TGA/DTA test below consists of 70% TPPS, 30% LLDPE-PP, and 10% Compatibility based on TPSS weight. Evaporations and Degradations occurs in three zone (Table-4 and Fig.-5). The first stage of evaporation and degradation occurs at a temperature of 0-316.5^{0}C with a mass loss of 29%. At this zone, the water and glycerol molecules have been completely evaporated.[21-22] Second zone, occurs at temperature 362.3^{0}C with a mass loss of 61.5%. In this phase, the starch compounds degradation begins at 300^{0}C.[22] In the final zone, degradation occurs at a temperature of 507.6^{0}C with a

mass loss of 91.9%. The decomposition of olefin compounds from Polyethylene/Polypropylene starting at 400^{0}C produces gas and oil.[23]

Table-4: The Decomposition Temperature of Starch Bioplastic and Composite Bioplastic

	1st Step	2nd Steps	3th Steps
TPSS	316.5	371.5	-
Composite Bioplastic	316.5	362.3	507.6

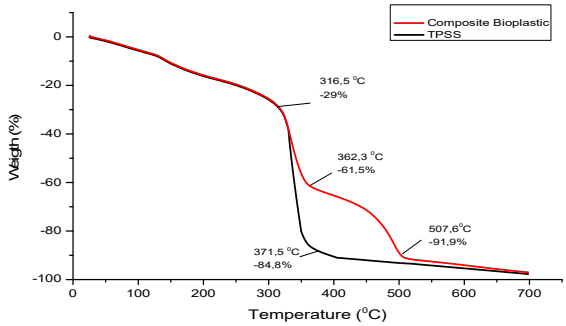

Fig.-5: TGA Curve of TPSS and Composite (LLDPE-PP 30wt.%)

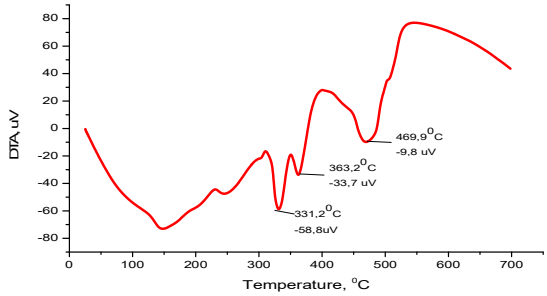

Fig.-6: DTA Thermogram of TPSS/PE-PP/Compatibilizer

The thermogram curve Fig.-6, it can be seen that there are several downward-facing peaks identified as endothermic processes. The first endothermic peak occurred at a temperature of 331.2^{0}C. The peak was widened facing downwards (endothermic process). This occurred in the second stage when viewed at the TGA baseline. At this stage, a thermal event occurs, namely dehydration or evaporation of water molecules and some of the glycerol. The second peak occurs at a temperature of 363.2^{0}C and is in the second stage of the TGA diagram. At this stage, the thermal degradation of starch occurs. The last endothermic peak occurred at a temperature of 469.9 ^{0}C in the third stage of the TGA curve. At this temperature, thermal degradation of a mixture of LLDPE and PP occurs.[18,24]

CONCLUSION

The addition of Polyethylene-Polypropylene can increase the tensile strength up to 4.8334 MPa at a concentration of 30%. The wavenumber shift in the TPSS/LLDPE-PP/Compatibilizer blends was detected at a wavenumber of 1021cm^{-1} as C-O. The results of the TGA thermal analysis stated that the degradation occurred in three zones. A significant decrease in mass occurred in the first stage at a temperature of 337^{0}C. Meanwhile, thermal DTA stated that an endothermic process occurred at a temperature of 331.2^{0}C, 363.2^{0}C, and 469.9^{0}C.

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CONFLICT OF INTERESTS

The author (s) declares that there is no conflict of interest in this research and manuscript.

AUTHOR CONTRIBUTIONS

All the authors contributed significantly to this manuscript, participated in reviewing/editing, and approved the final draft for publication. The research profile of the authors can be verified from their ORCID ids, given below:

S. Munthe https://orcid.org/0000-0001-9027-6817
D.A. Tanjung https://orcid.org/0000-0003-3556-4351
M. Banjarnahor https://orcid.org/0000-0002-2939-5697
R.S. Budi https://orcid.org/0000-0002-8275-3190

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