IMPROVING MECHANICAL AND PHYSICAL PROPERTIES ON THERMOPLASTIC SAGO STARCH BLENDS WITH THE ADDITION OF POLYPROPYLENE

D.A. Tanjung¹,², N. Jamarun¹,*, S. Arief¹, H. Aziz¹, A.H. Ritonga¹,³ and B. Isfa¹

¹Postgraduate School, Department of Chemistry, Universitas Andalas, Padang-25163, Indonesia
²Department of Agrotechnology, Faculty of Agriculture, Universitas Medan Area, Medan- 20223, Indonesia.
³Department of Chemistry, University of Sari Mutiara Indonesia, Medan-20123, Indonesia.
*Corresponding Author: novesarjamarun@sci.unand.ac.id

ABSTRACT
This study aims to improve the mechanical and physical properties of the thermoplastic sago starch blends with the addition of polypropylene (PP). PP concentrations were given starting from 10, 15, 20, 25, and 30 percent (%), with the addition of a PP-g-MA compatibilizer as a coupling agent. From the mechanical properties, it is known that the tensile strength and elongation at break increase with increasing PP concentration, whereas Young's modulus decreases with increasing PP concentration. The Thermogravimetry Analysis (TGA) did not show a significant reduction in degradation on each temperature increase. From the morphological results, it could be seen that the PP is dispersed into the TPSS matrix with the help of this compatibilizer, evidenced by the absence of flocculants on the surface. The rate of degradation indicates that the addition of PP to the sago starch matrix could still be degraded by environmental elements such as freshwater, seawater, and soil burial.

Keywords: Bioplastic, Compatibilizer, Coupling Agent, Polypropylene

INTRODUCTION
Plastic is a raw material that we often encounter around us. It's strong, light, durable and cheap, making this material widely applied to products such as packaging, household appliances, electronics, and others.¹ In the last half-century, plastic production has continued to increase with the increase in population from the estimated plastic production in 2018 to reach 360 million metric tons and about 18.5% market share in Europe.² Increasing plastic production must pay attention to the availability of fossil raw materials both today and in the future because the material is sourced from the earth's crust which cannot be renewed. and must also consider the effect of the waste generated on living things and the environment. Plastic waste in soil, marine and freshwater ecosystems causes severe problems for living things and can endanger human health.³-⁸ The current management of plastic waste is not perfect and there are around 5–13 million tons of plastic ending up in the oceans every year.⁹

Handling plastic waste for recycling requires special treatment such as separation, transportation, processing and remanufacturing.⁵ Usually, plastic waste that is burned emits carbon dioxide (CO₂) and other irritating gases into the atmosphere. However, if thrown in a landfill, it has a long decomposition time in the environment, even more than a century.³

To reduce plastic waste resulting from the use of products from synthetic polymers, researchers began to look for alternative materials to replace synthetic polymers with natural polymers such as cellulose, starch, lignin, and many others.¹⁰-¹³ The advantages of natural polymers are that they are inexpensive, widely available in nature, lighter, impact-resistant, and easy to implement. Besides, the most important thing is that it is biodegradable.¹⁴ to reduce the buildup in landfills. With the help of microorganisms, plastic molecules are broken down into simpler ones and finally converted into H₂O, CO₂, CH₄, and/or into new biomass substances that are non-toxic to humans and the environment.

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The development of global bioplastic production has shown increasing growth over the last few years. However, bioplastics still represent a small market share. The development of starch-based bioplastics has drawbacks such as low tensile strength, stiffness, and high water absorption. Plasticization of starch is an effort to modify the structure of starch compounds to be more elastic by adding plasticizers such as glycerol, glycol, sorbitol, urea, formaldehyde, and sugar. Some researchers report that the addition of a strength agent from synthetic polymers (plastics) can increase the tensile strength and make it more elastic. However, to increase the interfacial adhesion between the two components of different polarities, it is necessary to add a compatibilizer to act as a liaison between hydrophilic starch and hydrophobic plastics.

In this study, researchers added a thermoplastic polymer (PP) into thermoplastic starch (sago starch). The difference in polarity between hydrophilic and hydrophobic by adding PP-g-MA compatibilizer as a coupling agent.

**EXPERIMENTAL**

**Material and Method**

Sago starch was gained from Warna Jaya company, Parang brand, and water content, 14%. The Density of Polypropylene Pellet: 0.895 gr/m3, Melt Flow rate 27 gr / 10 minutes, Melt Point 130-157°C gained from PT. Chandra Asri, Indonesia.

**TPSS Preparation**

TPSS was processed by blending sago starch with a plasticizer (glycerol), added with water as solvent, as reported by R. Abdul Majid. Water content was less than 5%.

**Compatibilizer Preparation**

Compatibility was examined by reacting PP with Maleate Anhydride and Benzoyl Peroxide (BPO) as an initiator with the comparison of 88:9:3 (W / W) used internal mixer instrument at the temperature of 150% and speed 100 rpm. The entire mixing process lasted 13 minutes.

**TPSS / PP Blends Preparation**

The mixing of TPSS with PP was the aim of this research. The process was conducted in the internal mixer as the following compositions:

| Sample | TPSS % | PP % | Compatibilizer based on TPSS % |
|--------|--------|------|-------------------------------|
| 1      | 100    | 0    | 0                             |
| 2      | 90     | 10   | 9                             |
| 3      | 85     | 15   | 8.5                           |
| 4      | 80     | 20   | 8                             |
| 5      | 75     | 25   | 7.5                           |
| 6      | 70     | 30   | 7                             |

**Compression Molding**

The sample was printed using manual hydraulic pressure compression at 150°C lasted for 15 minutes. All printed specimens were by ASTM D638 Type 1.

**Tensile Strength**

Tensile Strength was examined using Universal Brand Tensilon machine. The Chamber temperature was 25°C with a speed of 10mm/minute.

**Spectroscopy of Fourier Transform Infrared**

Infrared Spectroscopy of Fourier transformation at Micro Lab University of Lampung. Background scanning= 32, resolution =16, interval= 4000-650 cm⁻¹
Thermogravimetric Analysis (TGA)
The used Thermogravimetric Analysis was STA module, TGDTA channel 7300. Measurement committed at 7.73mg with heat rate 10°C / minute. The heating started at 30°C to 650°C. The Nitrogen rate was 50 ml/minute.

Morphology Test
Morphology test was examined to observe PP dispersion into TPSS matrix as interfacial adhesion tension, using Eletion Microscopy Scanning device (SEM) at UPTLSIT Universitas Lampung with Mag=150x, WD=9.0mm, signal A= SE1, EHT = 15.0 kV.

Water absorption Test
The sample sheet with a dimension of 20mm x 20mm, thickness 3mm. The sample was then in distillation and dried up at 80% temperature for 12 hours, and then it was cooled down in a desiccator and weighed as initial weight. The sample was soaked in distilled water for 10 (ten) days at room temperature and reweighed to obtain post-soaking weight.

Biodegradability Test
The sample was tested for its biodegradability performance with the difference of condition control. Each sample was soaked in fresh water, seawater, and soil-burial, every 10, 20, and 30 days to be in observation.

RESULTS AND DISCUSSION
Mechanical and Physical Properties
Figure-1 Shows the continuous increase with the increasing PP concentration at tensile strength and Elongation at Break. This showed that the addition of PP has increased the mechanical nature of blended sago starch thermoplastic with the help of a compatibilizer. It is proven that the tensile strength and elongation at break of TPSS / PP blends, each increased from 1,73 N / mm2 to 5,51 N /mm2 and 0.32% to be 1,05%. The observed increase was due to the good interfacial adhesion between two polymers and different polarity resulted in good tension transfer between matrix (TPSS) and dispersed phase (PP) evenly. In contrast, (young’s modulus) decreased with the increasing PP concentration from 1321 to be 524 N/mm². Detailed data can be viewed in Table-2.

| PP Loading (%) | Density (gr/cm³) | Tensile Strength (N/mm²) | Elongation at Break (%) | Young's Modulus (N/mm²) |
|----------------|------------------|--------------------------|-------------------------|-------------------------|
| 0              | 1,17             | 1,73                     | 0,00                    | 0                       |

Fig.-1: Tensile Strength (TS), Elongation at Break (EB) and Young’s Modulus (YM) for loading PP on Thermoplastic of Sago Starch

Table-2: Density, Tensile Strength (TS), Elongation at Break (EB) and Young’s Modulus (YM) for loading PP on Thermoplastic of Sago Starch.
Fourier Transform Infrared (FTIR)

In the PP-g-MA spectrum, the characteristic peak was seen from Alkane (C-H) and Carbonyl (C=O) at wavenumbers of 2914 cm\(^{-1}\) and 1707 cm\(^{-1}\). The addition of PP and PP-g-MA into the TPSS blend resulted in value change of wavenumbers from 3265 to 3287 cm\(^{-1}\), this is the hydroxyl groups (OH) in the present together with peak 998 cm\(^{-1}\) as peak characteristic (C-O). In the spectrum of TPSS/PP compatibilized blends, The peak was formed at wavenumbers of 1695 cm\(^{-1}\), identified as Esther groups formation resulted from the reaction of hydroxyl groups of TPSS and Anhydride groups of PP-g-MA. It can be observed in Fig.-2.

![Fig.-2: The Spectrum of TPSS, PP-g-MA and TPSS/PP Compatibilized Blends](image)

Thermogravimetric Analysis (TGA)

The first phase of degradation occurred at 200,3\(^{0}\)C with a mass loss of 11,8\%. This was shown by water molecules evaporation together with a plasticizer (glycerol). In the second phase, degradation occurred at 350,8\(^{0}\)C with a mass loss of 60,7\%. In the final phase, degradation occurred at 501,4\(^{0}\)C with a mass loss of 87,4\%. From The TGA curve, clearly seen that degradation of TPSS / PP compatibilized blends does not show a significant reduction in each phase. This can be observed in Fig.-3.

![Fig.-3: The Spectrum of Thermogravimetry Analysis TPSS/PE Compatibilized Blends](image)
Morphology Test
In this study, an SEM test was committed to observing PP dispersion into the TPSS blend. In fig.4, TPSS/PP with the absence of compatibilizer showed the occurrence of flocculent (black circle) on the surface. In the concentration of 20 wt. %, PP was clearly seen, attempted to coagulate somewhere resulted from bad interfacial adhesion between TPSS and PP. This was due to the different polarities of the blends. Meanwhile, fig.5 is TPSS / PP compatibilized blends of SEM micrograph, showing us an even PP dispersion on the surface or a good occurrence of interfacial adhesion between TPSS and PP, with the help of PP-g-MA as compatibilizer. This observation supports the resulting test of the previous elongation at the break, where the presence of PP-g-MA could help the interaction of the two materials with different polarity increase.

Water Absorption Test
The water absorption index of various PP concentrations is presented in Fig.-6. The figure showed the water absorption toward TPSS and TPSS/PP compatibilized blends. In the TPSS blend, water absorption was bigger than TPSS /PP compatibilized blends of 121,3%, resulting from the hydrophilic property of starch which was responsible for water absorption in the blend. Meanwhile, the ability of water absorption in TPSS / PP compatibilized blends decreased to the increasing content of PP. There has already been a reaction between hydroxyl groups from sago starch and anhydride groups from PP-g-MA, as it decreased the water molecule interaction with hydroxyl starch groups.

Biodegradation Test
Characteristic of bioplastic is degradable. The decomposer, namely microorganism, environment, and time affected degradation rate ability. In this study, speed and decomposition were observed based on the environment (freshwater soaking, seawater, soil burial), and length of degradation. From fig. 7,8, and 9, it can generally be concluded that the percentage of sample weight reduction increased to the increasing time of degradation. The degraded material reported was starch, while PP was not degradable within a
short time. If sorted based on the degradability speed of the three environments, namely; first freshwater, seawater, and soil burial.

If the remaining freshwater soaked sample, be in observation until the 30th day which was not degraded yet in each PP concentration (10, 15, 20, 25, and 30%) were 62, 56, 52, 45 and 41%. Soaked in the seawater were 55, 52, 47, 42, and 38% in the soil burial were 53, 45, 38, 32, and 25%. Detailed data can be viewed in Table-3.

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**Fig.-6: Water Absorption Test**

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CONCLUSION

From the explanations above, it can be concluded that the addition of Polypropylene (PP) to Thermoplastic Sago Starch (TPSS) could increase the mechanical property, like tensile strength and elongation at break. Compatibilizer (PP-g-MA) as the coupling agent could help to react TPSS with PP. The spectrum of FTIR showed that hydroxyl groups from TPSS have been in reaction with anhydride groups, forming Esther groups. TPSS / PP compatibilized blends can be degradable in the three environments, namely freshwater, seawater, and soil burial. The figure of SEM TPSS/PP/Compatibilized blends, showing that PP dispersed evenly in the whole surface with the help of compatibility.

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