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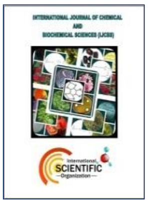

Improved mechanical properties by adding a compatibilizer to the thermoplastic of sago starch/polypropylene blends

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Abstract

The addition of maleic anhydride (MA) grafted onto polypropylene (PP-g-MA) as a compatibilizer into the thermoplastic of sago starch (TPSS)/Polypropylene (PP) blends is expected to improve the mechanical properties. TPSS is made by mixing 65% by weight of starch and 35% by weight of glycerol. The compatibilizer was made by mixing PP, MA, and Benzoyl Peroxide (BPO) in a ratio of 88:9:3% by weight. The composition of TPSS and PP in the manufacture of bioplastic composites is set at 80:20% by weight. Variations in the concentration of PP-g-MA started from 0,6,8,10,12, and 14 wt.%. The test results obtained, the tensile strength increased significantly after the addition of PP-g-MA from 0.6449 N/mm2 (without PP-g-MA) to 4.6393 N/mm2 at the concentration of 10% PP-g-MA. The water absorption test decreased to 8% at a concentration of 10% PP-g-MA and the degradation test stated that bioplastic composites could decompose up to 180 days in freshwater media with a weight loss of 67%, in seawater media 72% and in medium soil 69% in the concentration of 10% PP-g-MA each.

Keywords: Bioplastic, Compatibilizer, Coupling Agent, Thermoplastic

Full length article *Corresponding Author, e-mail: novesarjamarun@sci.unand.ac.id

1 Introduction

Plastics are synthetic or semi-synthetic organic polymers that have the characteristics of being light, durable, and inexpensive [1]. Over the past 50 years, conventional fossil-based plastics have become a part of our daily lives. Plastic production continues to increase as the population increases from 1.5 million metric tons in 1950 to 359 million metric tons in 2018[2] and 18% produced in Europe [3] . The development of the plastic industry is a must to meet the needs of the world's growing population. However, it should also be considered that conventional plastic production is based on the availability of fossil fuels [4] and the effects of waste on living things and the environment. Behind the advantages of its unique properties, plastic also has disadvantages, especially in handling waste. Handling plastic waste requires special care for recycling and when disposed of in landfills, it has a long decomposition time in the environment even more than a century[5]. Apart from the immediate landscape problem, plastic pollution (especially from microplastics) in soil, marine and freshwater ecosystems causes serious problems for living organisms and can harm human health [6-10]. The current management of plastic waste is far from perfect and around 5–13 million tonnes of plastic end up in the oceans every year [11-12].

To overcome the problem of plastic waste generated, researchers are starting to look at biomass as an alternative to synthetic polymers. The advantages of biomass are that it is inexpensive, lighter, impact-resistant and easy to implement. Besides, the most important thing is that it is biodegradable [13], so as to prevent it in landfills. Then, the structure of the material is simplified to finally be converted into H_2O, CO_2, CH_4, and/or into new biomass substances and finally into residues that are non-toxic to humans and the environment. Bio-based plastics are made from biomass example cellulose, starch, lignin, and many others). Some bio-based plastics are able to degrade naturally in the environment into harmless natural substances, while some of them require conditions. Global production of bioplastics has shown continuous growth over the last few years. However, bioplastics still represent a small market share [14]. Starch is one of the biomass which is a natural polymer that can be easily plasticized using glycerol to reduce the stiffness properties [15-18]. However, starch-based bioplastics are generally characterized by poor mechanical properties and high water sensitivity [19-20] .

Mechanical properties can be improved by using reinforcement such as vegetable fiber, cellulose, clay, and the addition of synthetic polymers such as PE, PP and others [21-24]. Based on the description above, it is necessary to research on the manufacture of bioplastic composites with a mixture of sago starch as a matrix and polypropylene as a reinforcement with variations in the concentration of compatibilizer (PP-g-MA).

2. Materials and Methods

2.1 Material

The materials used in this study were commercial Sago starch from the Parang brand of PT Warna Jaya Indonesia, Maleic Anhydride, Polypropylene Pellet: 0.895gr/m3, Melt Flow rate 27gr / 10 minutes, Melt Point 130-157^{0}C gained from PT. Chandra Asri, Indonesia.[25]

2.2 Procedure

2.2.1. Preparation of Thermoplastic of Sago Starch (TPSS)

TPSS is made by reacting sago starch with glycerol plus water as a solvent. The ratio of sago starch and glycerol is 65: 35.[26] The amount of water solvent added is 250% of the total mixture. The mixture was cooked at 100^{0}C until it formed gelatin. To reduce the water content, gelatin is oven-baked for 24 hours at 80^{0}C, the water content is up to 5%.

2.2.2. Preparation of Compatibilizer (PP-g-MA)

The blends were mixed into Haake Polydrive Thermo. LLDPE was firstly added into the mixing chamber, followed by Maleic Anhydride after 5 minutes of a mixture, then BPO was added at last. The mixture was done at 150^{0}C and speed of 100 rpm, with the mixture total time was 13 minutes. The composition comparison of LLDPE: Maleic Anhydride: Benzoyl Peroxide was 88:9:3. The compound was cooled, and cut into pellets.

2.2.3. Preparation of TPSS/LLDPE/PE-g-MA blends

The preparation of TPSS / LLDPE/PE-g-MA blends was the final phase in this research, in which the whole phases that have been prepared above would be mixed here, such as TPSS, LLDPE, and compatibilizer. The ratio of TPSS and LLDPE is 80:20. Concentration of compatibilizer are 0,6,8,10, 12 14% based on weight of TPSS.

2.3. Instrumentation

The blend of TPSS / LLDPE was printed with hydraulic pressure compression at 150^{0}C for 15 minutes. Every 5 minutes, the pressure was increased to maximum. All compression molded sheets by ASTM D638 Type 1. Some of theInstrumentations used to support in this research are :Spectroscopy Fourier Transform Infra-Red (FTIR), Tensile Properties Test, Morphology test (SEM), Water Absorption Test, Biodegradability Test.

3. Results and Discussion

3.1. Mechanical Properties

TPSS/PP Blends with and without a compatibilizer was prepared to determine the best formation of the mixture. Tensile properties of the TPSS/PP composite mixture were investigated to reveal the effect of the concentration of the compatibilizer on the adhesion of the TPSS/PP interface. Figure 1-3 illustrates the variation in tensile strength, elongation at break, and Young's modulus of a TPSS/PP composite mixture with and without a compatibilizer. From Fig. 1 and 2, it can be concluded that the tensile strength and elongation at breaks increase as the concentration of the compatibilizer increases until the maximum load peaks at 10% of the compatibilizer with a value of 4.6393N/mm^2 for tensile strength and 0.75% for elongation at break and then there is a not so significant decrease. significantly as the concentration of the compatibilizer increases. This increase was due to the good interfacial adhesion of the two polymers of different polarity where hydrophilic TPSS and hydrophobic PP were added after the addition of a compatibilizer as a connecting agent. While the decrease in tensile strength and elongation at break occur due to the saturation point of the concentration of the compatibilizer being reached. Fig. 3 shows a very significant increase to 620N/mm^2 concentration of 10% PP-g-MA when compared without compatibility at 84 N/mm2. Furthermore, the trend of the graph decreases as the load of the compatibilizer increases. This observation has been reported by [27]. In general, it can be concluded that the addition of a compatibilizer shows better tensile strength when compared to composites without a compatibilizer according to reports in the literaturee [23, 28] .

3.2. FTIR Analysis

The FTIR test is needed to show the reaction that occurs after the two polymer materials are mixed as functional group identification. FTIR analysis of the reaction of Thermoplastic sago starch (TPPS)/PP-Compatibility (PP-g-MA) in Figure 4.47 detects the presence of a hydroxyl group at a wavenumber of 3287cm^{-1} strengthened by a C-O group at a wavenumber of 1013 cm^{-1} which is the characteristic peak of sago starch. Next is the wavenumber 2922cm^{-1} which is the C-H carbon chain. A wavenumber 1647cm^{-1} detects the presence of a carbonyl group (C=O) which is assumed to come from the anhydrous group of LLDPE-g-MA and finally, the presence of a CH2 group at wave number 1453cm-1 is a carbon chain from CH_2 .(Fig.4).

3.3. Degradation Test

The degradation test is the most important in the manufacture of bioplastic composites. Its ability to degrade is needed to be safe for living things and the environment. In this bioplastic composite, the composition of starch is 80% while PP is 20%. Table 1-3 are the degradation test under 3rd environmental conditions: Fresh-water, Sea-water, and soil burial.

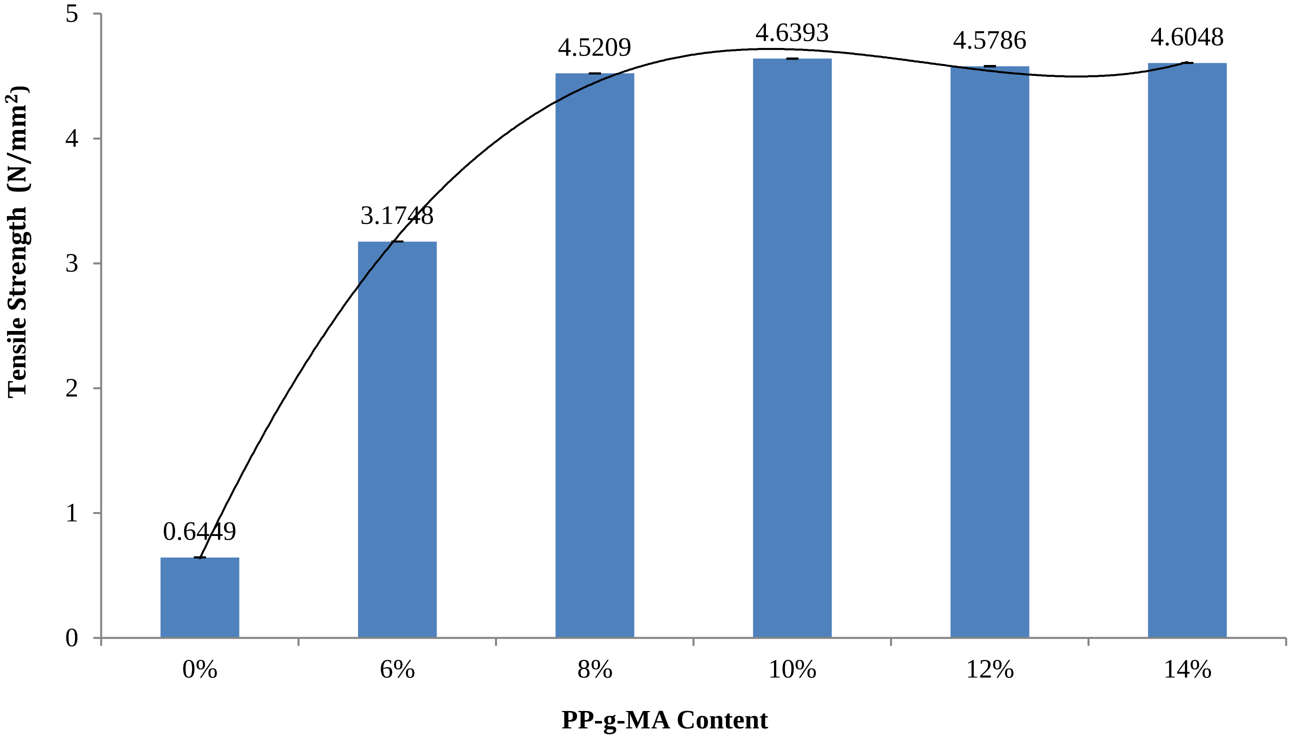

Fig. 1. Tensile Strength Test

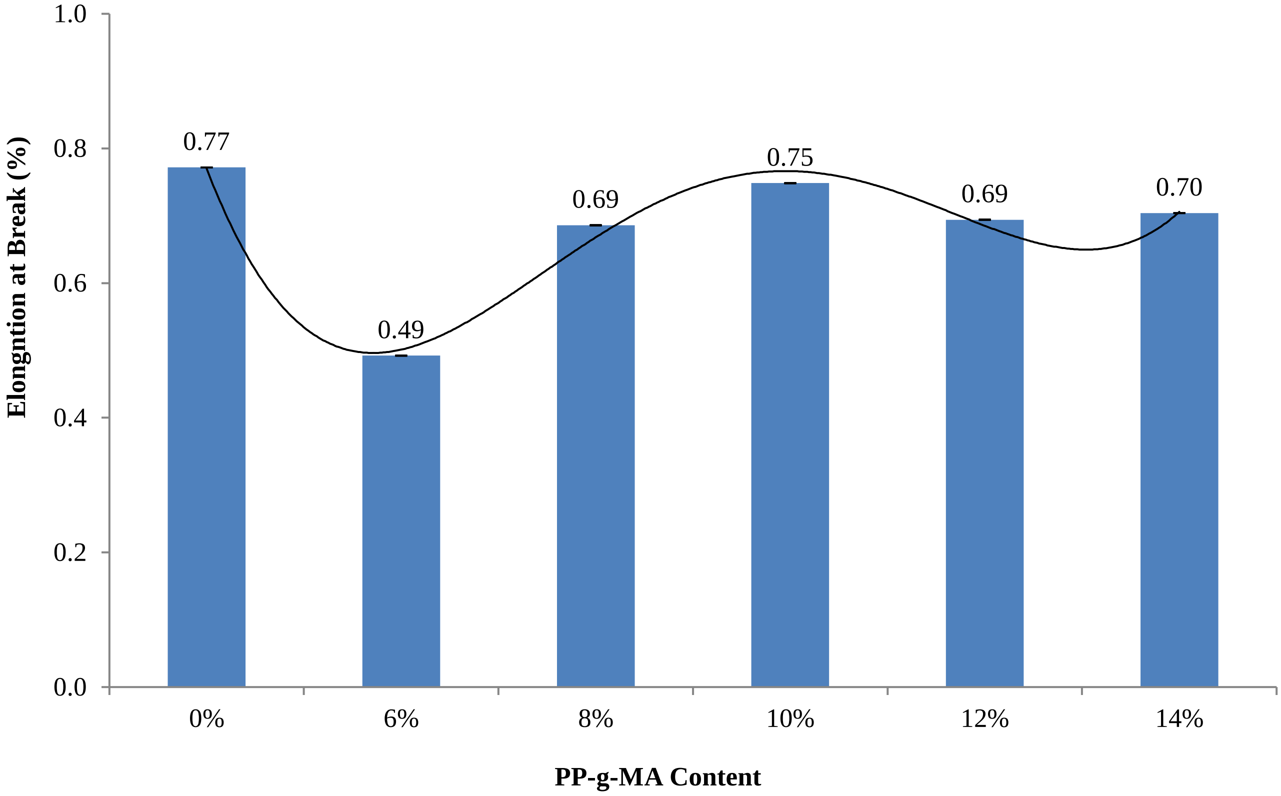

Fig. 2. Elongation at Break Test

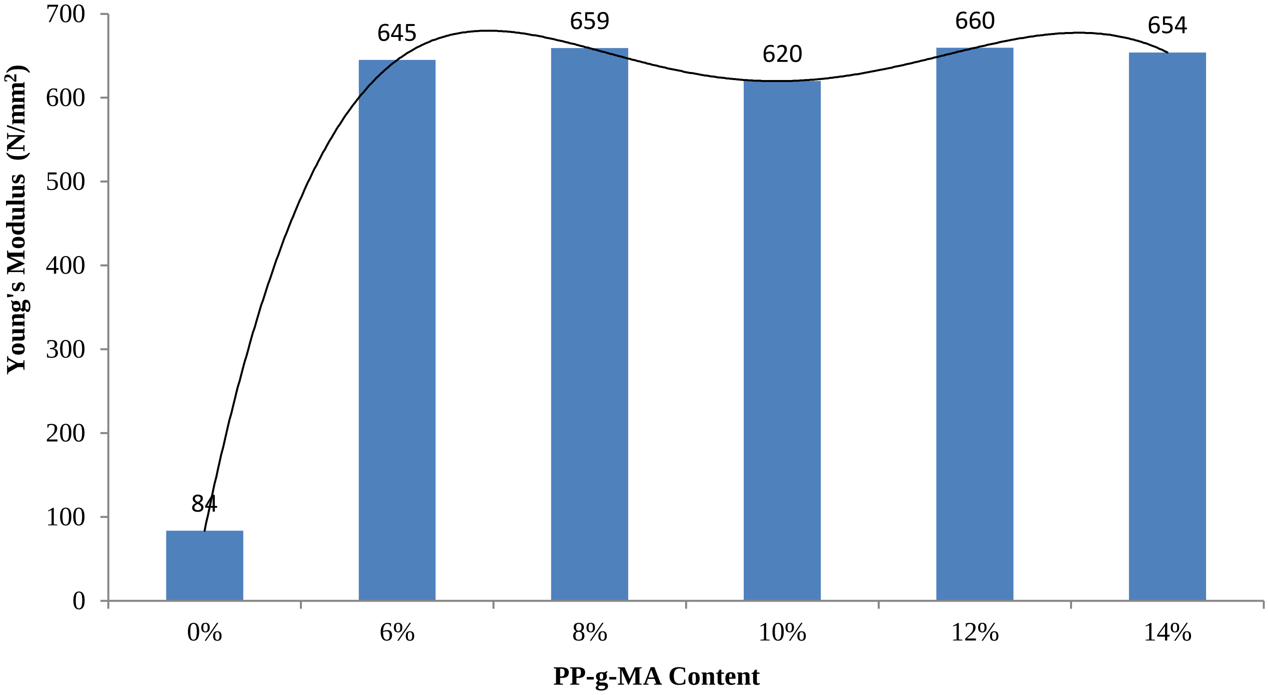

Fig. 3. Young's Modulus Test

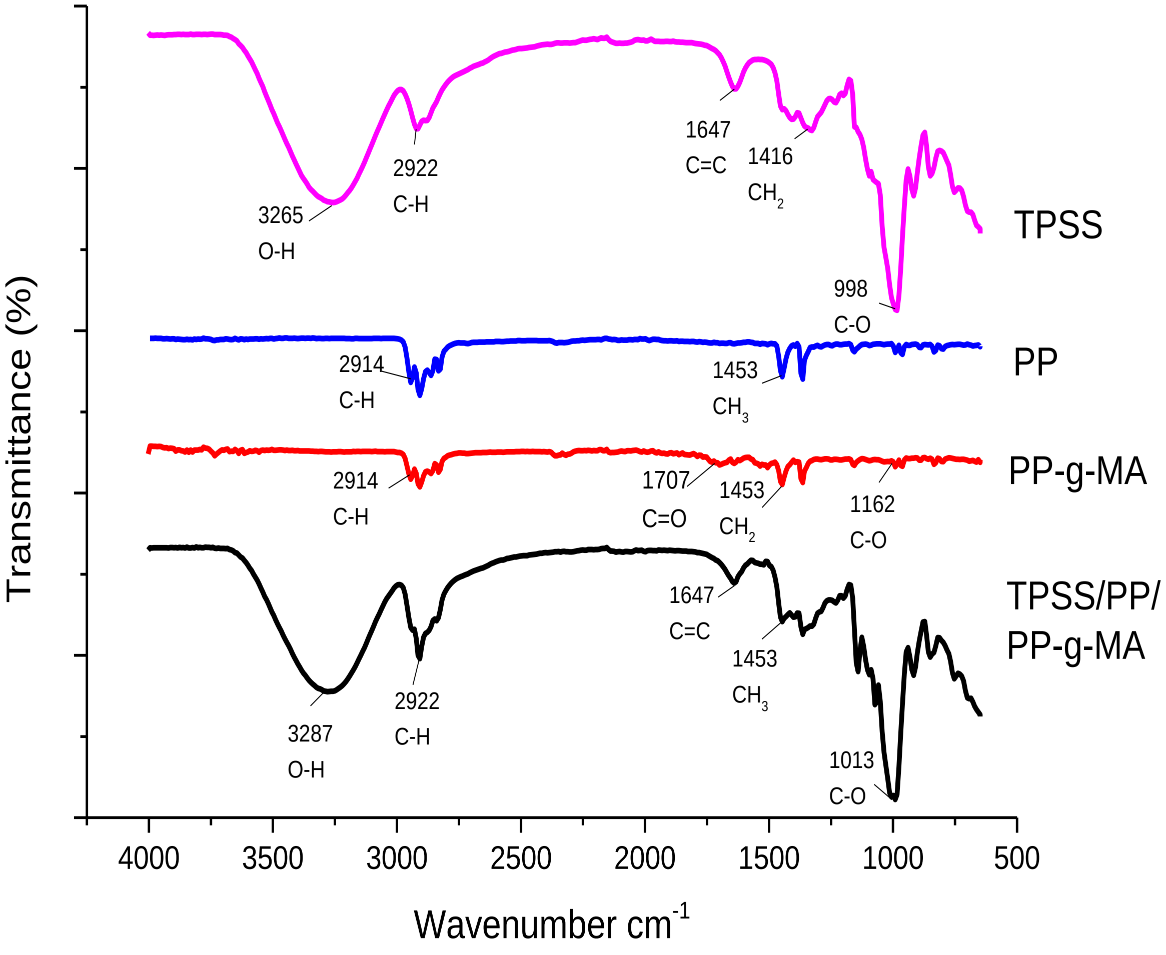

Fig. 4. FTIR Spectrum of TPSS

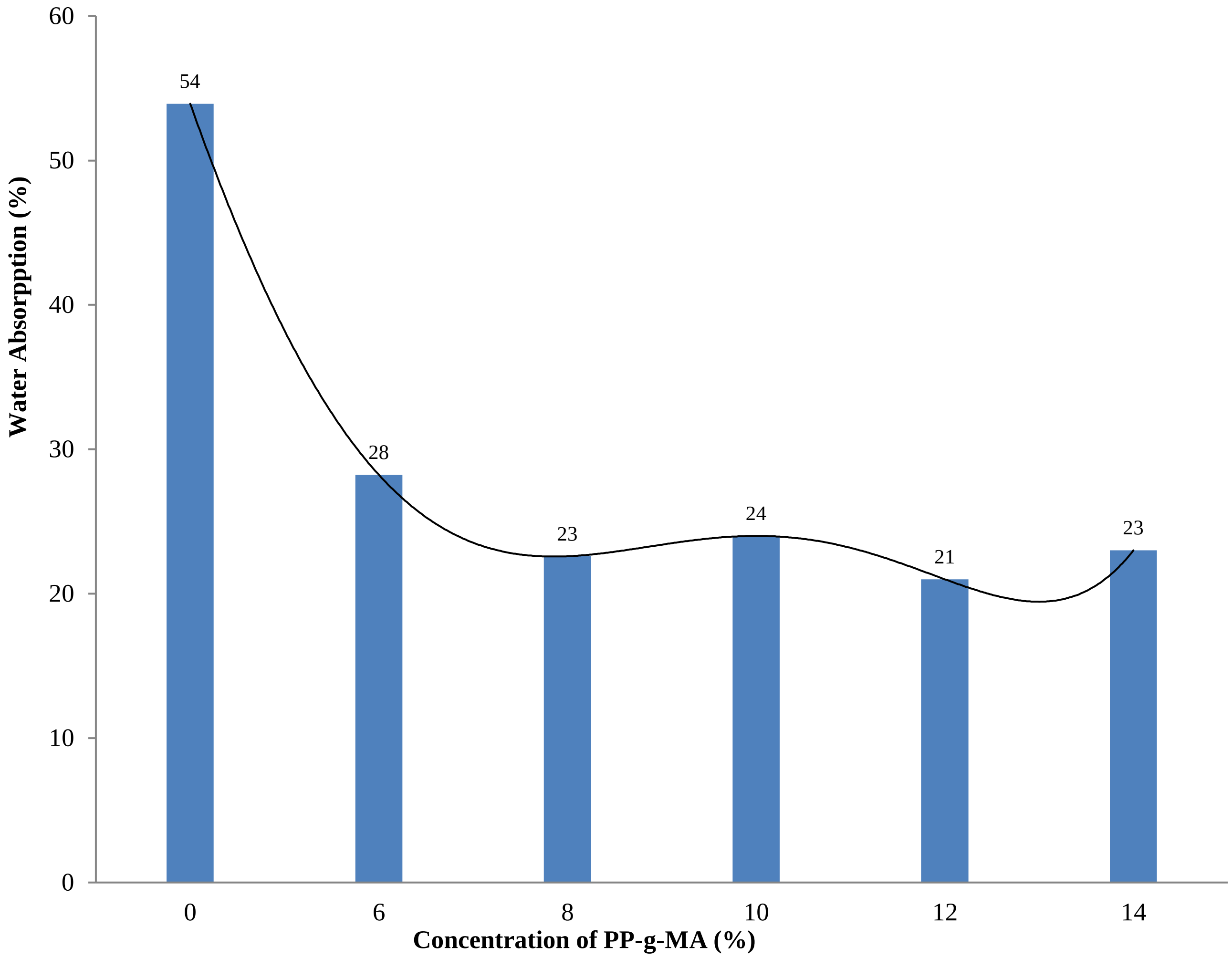

Fig. 5. Water Absorption Test

Table 1. Degradation test of Freshwater

Concentration of PP-g-MA %	Days-						
	10	**30**	**60**	**90**	**120**	**150**	**180**
0	21	31	40	47	55	61	63
6	34	54	51	53	54	57	59
8	38	47	50	51	55	56	63
10	56	58	60	63	64	64	67
12	49	54	56	58	59	63	65
14	53	55	55	56	59	63	65

Table 2. Degradation test of Seawater

Concentration of PP-g-MA %	Days-						
	10	**30**	**60**	**90**	**120**	**150**	**180**
0	41	46	51	55	58	66	72
6	40	51	54	57	59	61	65
8	50	56	57	59	61	67	71
10	55	60	63	67	69	71	72
12	48	56	59	62	63	64	67
14	49	55	58	61	62	65	68

Table 3. Degradation test of Soilburial

Concentration of PP-g-MA %	Days-						
	10	**30**	**60**	**90**	**120**	**150**	**180**
0	29	33	47	55	63	67	68
6	31	46	49	54	61	65	70
8	38	41	49	56	62	64	69
10	41	53	56	59	61	64	69
12	38	55	56	58	60	61	63
14	45	53	55	58	60	63	66

The bioplastic samples tested for degradation were samples that had a composition of 80% TPPS, 20% PP with variations in PP-g-MA concentrations of 0,6,8,10,12 and 14%. From the graph below, we can analyze that until the 180th day the bioplastic samples without a compatibilizer have decomposed as much as 63%, this means that there are about 37% that have not been decomposed, 17% comes from sago starch. starch runs out. While the sample with PP-g-MA concentration of 14% was degraded up to 65%, or about 35% which had not been degraded, 15% came from sago starch. This sago starch will continue to decompose if given additional decomposition time (Table 1). Table 2, can be analyzed that the degradation rate in the sample without a compatibilizer on the 180th day has decomposed as much as 72%, or about 23% that has not been decomposed, 3% of which comes from sago starch which has not been degraded. Meanwhile, at the concentration of PP-g- MA 14% has been degraded as much as 68%, or about 32% which has not been degraded. 12% of it comes from sago starch, and if given additional degradation time, sago starch will continue to degrade until it is exhausted. Samples planted in soil were degraded up to 68% in samples without a compatibilizer (table 3). This means that there are about 32% have not been decomposed, 12% of which comes from sago starch. This sago starch will continue to decompose if given an extension of the decomposition time. Meanwhile, at a concentration of 14% PP-g-MA has decomposed as much as 66%. This means that there are about 34% have not been decomposed, 14% of which comes from sago starch. The rate of degradation in the soil is influenced by moisture, the number of types of decomposing microorganisms, and the number of clones of microorganisms present in the soil.

3.4. Water Absorption Test

One of the main disadvantages of using starch in the manufacture of bioplastic composites is its tendency to absorb water. This test parameter is needed so that the bioplastic composite is not easy to absorb water and can cause a decrease in tensile strength. This tendency to absorb water is caused by the hydroxyl groups in starch that can easily bind to hydrogen atoms from water. In Fig. 5, it can be seen that the minimum ability to absorb water is 24% in a 10% compatibilizer blends. It is assumed that the maximum compatibilizer concentration as a link between starch and PP is at a concentration of 10% [30].

4. Conclusion

The addition of PP-g-MA as a compatibilizer as a coupling agent between natural polymers (starch) and synthetic polymers (PP) can improve mechanical properties. The maximum concentration of PP-g-MA in a mixture of 80% sago starch and 20% PP is 10% by weight of TPSS. In general, TPPS/PP/PP-g-MA samples can be degraded under all three environmental conditions. The addition of PP-g-MA into the TPPS/PP mixture did not affect the rate of starch degradation. and the water absorption ability is reduced by up to 24% at a concentration of 10% PP-g-MA.

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