

# Fabrication of Biodegraded Composite from Polypropylene, Polypropylene Grafted Maleic Anhydride, and Chempedak Seed (*Artocarpus integer*) Starch

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**Abstract.** Fabrication of biodegraded composite from polypropylene, polypropylene grafted maleic anhydride, and chempedak seed (*Artocarpus integer*) starch has been conducted. The stages were prepared starch of Cempedak seed, grafting process of maleic anhydride on polypropylene, and fabrication of composite with a composition ratio of PP: PP-g-MA: starch of chempedak seed (9.5:0.5:0.1)g, (9.5:0.5:0.2)g, (9.5:0.5:0.3)g, (9.5:0.5:0.4)g, (9.5:0.5:0.5) g, and PP: starch of chempedak seed (10:2)g. The composites obtained were characterized using tensile strength and elongation testing, surface morphology analysis by SEM, analysis in nature with the ability to decompose by biodegradable test, and functional groups analysis by FTIR spectroscopy. The results showed that the biodegradable composites have the best properties in the ratio of PP: PP-g-MA: starch of chempedak seed (9.5:0.5:0.1)g with the test results of tensile strength was 28.743 N/m<sup>2</sup>, and also the SEM results show that flat surface and improved homogeneity, and strong group interactions as well as having good biodegradable properties.

**Keywords:** Composite Biodegradable, Starch, Chempedak Seed, PP-g-MA

Received [5 May 2021] | Revised [21 June 2021] | Accepted [14 July 2021]

## 1 Introduction

Today, the use of composite materials has begun to be widely developed in the manufacturing industry. Composite materials that are environmentally friendly and can be recycled again, are the demands of today's technology. One of the composite materials that are expected in the industrial world is a composite material with filler material in the form of natural or artificial fibers. A composite material is a combination of two or more different materials into a microscopic unit form, which is made of various combinations of properties or a combination of fiber and matrix. Currently, fiber-reinforced composite materials are engineering materials that are widely used because of their strength and specific properties that are far above other

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engineering materials in general, so that their properties can be designed close to the needs (Jones, 1975).

Most polymers are extremely durable. In fact, this is a property that allows them to compete with other durable materials such as glass and metals. As previous research has focused on increasing the lifetime of certain polymers. However, durability can create new problems. Recently, conservationists have increased their attention to polymer waste which is an eyesore. Although synthetic polymers currently represent a relatively small percentage of all solid waste, they are still very eye-catching, especially because of their widespread use in the packaging (Stevens, 2007).

Polypropylene is a type of lightweight plastic raw material, its density is 0.90-0.92 g/cm<sup>3</sup>, has the highest hardness and brittleness, and is less stable to heat due to the presence of tertiary hydrogen. The use of fillers and reinforcement allows polypropylene to have good chemical quality as a polymer material and is resistant to cracking under pressure even at high temperatures (Gachter, 1990).

Polypropylene is a non-polar polymer that can be changed from non-polar to polar by grafting a polar functional group into the chain in the presence of an initiator. The grafting of maleic anhydride on polypropylene aims to improve the compatibility and reactivity of polypropylene. Polypropylene grafting reactions have been carried out a lot but the melt method is better than the mixing method in solution (Gracia, 1997).

Starch is a very abundant carbohydrate in nature and can be obtained from various grains such as rice, cassava, sago, corn, and so on. Starch is a high polymer carbohydrate arranged in Glucopyranose units, with a glucoside sequence. Carbohydrates are classified enzymatically as monosaccharides, disaccharides, and polysaccharides. It is a colorless, crystalline substance that usually has a taste and is insoluble. Indonesia is a potential area as a producer of starch such as cassava, sago, corn, sweet potato, and so on because these plants are spread throughout Indonesia (Wahidoen, 1991).

(Clemons, 2003) reported preparation of a composite material by combining polypropylene plastic material with mixed cellulose and then produced in two ways, namely press molding and by injection system of polypropylene (PP) matrix material to produce a biodegradable composite. Cellulose was chosen as filler because of its good heat stability and high purity.

Widyasari (2010) examined the study of the addition of thermoplastic Onggok (tapioca starch) to the characteristics of polyethylene composite plastics, in which research has been performed. It can be concluded that composite plastics that have the best mechanical physical properties are LDPE and thermoplastic Onggok was added 30% of glycerol with a ratio of 60:40 (LDPE:

thermoplastic pile) in general, composites can be enzymatically degraded and can grow mold, the application of this composite product is suitable for biofoam, namely substitution of styrofoam which is easily degraded.

Bambang (2011) examined determination of the glucomannan biodegradable plastic composite formula from Porang tubers (*Amorphophallus muelleri* B) in terms of physical and mechanical characteristics. The composite formula that produces the best biodegradable glucomannan plastic characteristics is a composite with the formula 23 g of chitin + 25 g of glucomannan + plasticizers 29 with the modulus of elasticity was 58.70 kg/cm<sup>2</sup>, modulus of fracture was 59.91 kg/cm<sup>2</sup>, maximum tensile stress was 6.69 kg/cm<sup>2</sup>, thickness expansion was 33.30%, percentage of water absorption was 77.63%, and percentage change in length 1.81%.

Based on the description above, the authors would like to make the biodegradable composites using polypropylene grafted maleic anhydride and using chempedak seed flour as a filler to improve the physical and mechanical properties of the biodegradable composite material. It is hoped that this research will provide information for further researchers and produce composites as expected.

## **2 Materials and Methods**

### **2.1 Equipments**

In this study, the equipment used were Liebig cooler, hot plate stirrer, magnetic stirrer, 250 mL of bottom flask, thermometer, blender, 100 mesh of sieve, beaker glass, analytical balance, measuring cup, oven, spatula, internal mixer, vacuum pump, filter paper, a set of SEM, FT-IR spectroscopy, printer press, and universal testing machine.

### **2.2 Materials**

The materials used were polypropylene, dicumyl peroxide, chempedak seed, maleic, xylene, methanol, and acetone.

### **2.3 Preparation of Cempedak Seeds Starch**

Chempedak seed was washed thoroughly, dried in the sun to remove the water, then steamed to facilitate stripping the hard skin and epidermis of the chempedak seeds for 60 minutes. After cooling, then peel the skin and cut it into small pieces, then blend until smooth.

### **2.4 Fabrication of Polypropylene Grafted Maleic Anhydride (PP-g-MA) and Purification Process**

The grafting process was performed using an internal mixer with a ratio of polypropylene, dicumyl peroxide, and maleic anhydride, namely 95%: 3%: 2% (w/w) at 165°C and produced polypropylene grafted maleic anhydride (PP-g-MA). Furthermore, PP-g-MA was purified by

refluxing with xylene, precipitated with acetone, filtered and the precipitate was washed with methanol repeatedly. Then dried in the oven at 120°C for 6 hours.

## **2.5 Manufacturing of Composite Specimen**

Cempedak seed starch and PP-g-MA were weighed according to the following variations: 0.1 g; 0.2 g, 0.3 g; 0.4 g; and 0.5 g, and this process was also carried out on the weight of PP-g-MA with the weight of PP-g-MA was: 0.5 g, and the weight of PP was 9.5 g.

Sample 1 was mixed into a glass beaker, dried in a blender until smooth, and placed between 15 cm x 15 cm of aluminum plates that had been coated with aluminum foil to be shaped according to ASTM D638. Then pressed on a hydraulic press at 160°C for 30 minutes. The result was cooled at room temperature and removed from the mold.

## **2.6 Tensile Strength and Elongation Testing**

Tensile strength testing was carried out using an autograph tensile tester for each specimen based on ASTM D638, with a tensile speed of 50 mm/minute and a maximum load of 100 kgf. Torsion's Electronic System was turned on and left for 1 hour. The sample was clamped using a gripper. The stress, strain, and units were set and turned on by the recorder (ON). The recording ink was installed and set the x-axis (strain) and y-axis (stress) and set the unit. The sample was assembled. Pressed the start button. Zero the load and stroke values. Look at the numbers in the load (tension) and stroke (strain), when the sample has broken. The load and stroke values of the samples were recorded. The same treatment was carried out for each sample.

## **2.7 Surface analysis with SEM**

The process of microscopic observation using SEM was carried out by gluing the sample with a stab made of older specimen metal. Then after the sample was cleaned with a blower, the sample was filled with gold and palladium with a diospater machine with a pressure of  $1492 \times 10^{-2}$  atm. The sample was then put into a special room and then irradiated with a bouncing electron beam that can be detected with a scintillator detector which was then amplified with an electrical circuit that causes a CRT (Cathode Ray Tube) image to appear. The shooting was done after selecting a certain part of the object (sample) and the desired magnification so that a good and clear photo was obtained.

## **2.8 Composite Biodegradability Test**

The biodegradability test was carried out by planting in the soil environment, starting with planting each specimen in a container containing 3 types of soil, namely sandy soil, garbage soil, and garden soil. This was done to look at the type of soil which has a better level of biodegradation. The rate of biodegradation of planting in the soil was observed for 1 month with observations every 10 days, by calculating the percentage change in specimen weight by

weighing the specimen and with the results of SEM and FTIR tests. Then calculated the initial weight before planting and weight after planting, and the difference was recorded as a % change in weight.

### 3 RESULT AND DISCUSSION

#### 3.1 Analysis of Tensile Strength ( $\delta$ ) and Elongation ( $\epsilon$ ) of Biodegradable Composites with Various Variations

The tensile strength and elongation of the biodegradable composite with several variations of the weight of the chempedak seed starch were shown in figure 1 and 2.

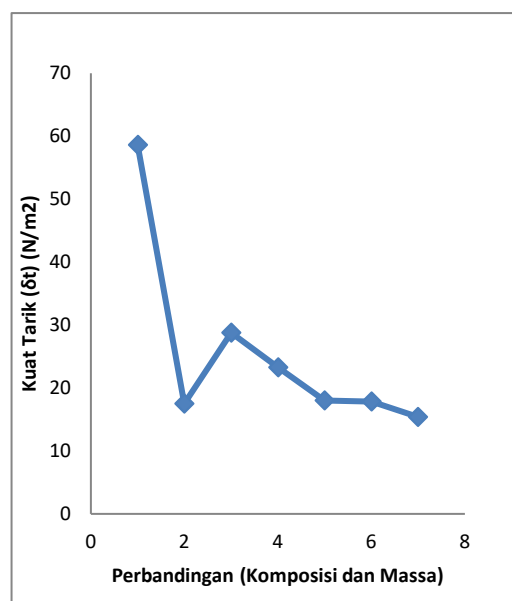


Figure 1. The biodegradable composite tensile strength graph

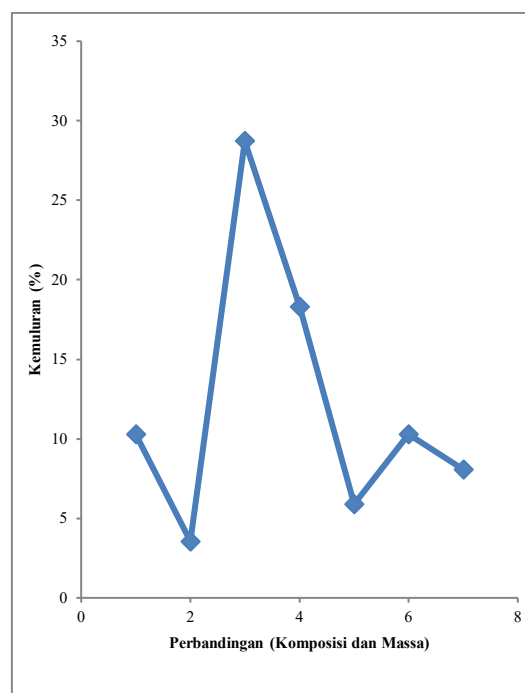


Figure 2. The biodegradable composite elongation graph

The results obtained showed that the optimum value was found in the mixture of PP:PP-g-MA: Chempedak seed starch (9.5:0.5:0.1) g with a tensile strength value of 28.743 N/m<sup>2</sup> and elongation 28.720%. The minimum tensile strength was found in the variation of the addition of 0.5 g of chempedak seed starch filler with a tensile strength value of 15.401N/m<sup>2</sup> and an elongation of 8,060%. The tensile strength decreases with the increase in the filler material. Chempedak seed starch was added due to the inability of the filler to support the transferred stresses by the composite matrix. Good filler dispersion and matrix-filler interaction may be the two main factors responsible for the decrease in tensile strength.

### 3.2 Composite Surface Morphological Analysis with SEM (Scanning Electron Microscopy)

The SEM results indicated in figure 3 show the fracture surface structure of the biodegradable composite (figure 3. a), In addition, the SEM result was a more even and homogeneous mixture (figure 3. B). This was because the mixture of matrix and filler can interact well and the mixture of PP:PP-g-MA: chempedak seed starch (9.5:0,5:0,1) g after being planted in waste soil for 30 days, shows a flat surface and no lumps. It indicated that the composite can interact well to produce a flat surface

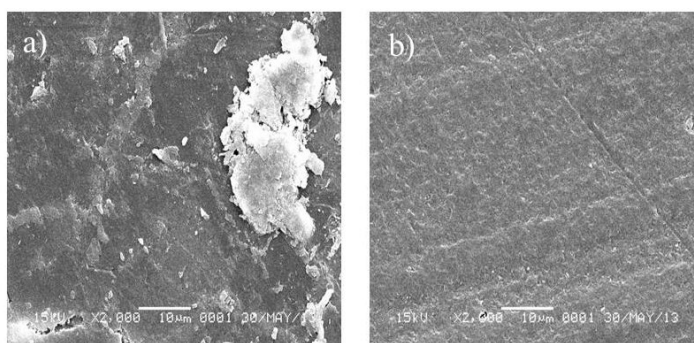


Figure 3. The SEM morphology of biodegradable composite (a) PP: chempedak seed starch (10:2) g, and (b) PP:PP-g-MA: chempedak seed starch (9.5:0.5:0.1) g

### 3.3 Biodegradability Analysis

The burial of the specimens was carried out on several types of soil for 30 days with observations every 10 days. The magnitude of the decrease in the mass of the polymer composite matrix specimen was in line with the length of burial time. The biggest decrease in the mass of the biodegradable composite was buried in waste soil, garden soil, and then sand soil. This may be because the number of nutrients in the waste soil was more than in other soils, so the number and types of microbes were also higher. Therefore, there was a synergistic performance between the activities of several microbes (Basuki Wirjosentono, 1998).

The main step of degradation was the breaking of the main chain to form low molecular weight fragments (oligomers) that can be assimilated by microbes. Based on this description, it can be seen that the degradation by planting was faster because all the factors that trigger the degradation were found in the external environment.

### 3.4 FT-IR (Fourier Transform-Infrared) Spectroscopy

The FT-IR analysis was carried out to determine changes in functional groups that indicated chemical interactions between one component and others. Analysis with infrared spectrum was performed by observing the typical frequencies of the functional group of FTIR spectra of each sample. The FTIR spectra were presented in Table 1 and 2.

**Table 1.** The wavenumber of PP: chempedak seed starch (10:2) g

Wavenumber (cm <sup>-1</sup> )	Functional Group
3500-3200	O-H
1840-1810	C=O
1377.0	CH <sub>3</sub>
1166.0	C-O
973.0	C-O
840.1	C-H

**Table 2.** The wavenumber of PP: PP-g-MA: chempedak seed starch (9.5:0.5:0.1) g

Wavenumber (cm <sup>-1</sup> )	Functional Group
2965.0	CH
1755-1735	C=O
1458.1	CH <sub>2</sub>
1377.1	CH <sub>3</sub>
1167.1	C-O
997.2	C-O
973.2	C-O
841.2	Aromatic C-H

Table 1 shows the wavenumber of each functional group of a mixture of PP: chempedak seed starch (10:2) g. The band at 3500 to 3200 cm<sup>-1</sup>, which indicated the presence of the O-H group. The band at 1840 to 1810 cm<sup>-1</sup>, which assigned the C=O bond.

From the results of functional group analysis using FTIR, it was known that the resulting spectra do not have a significant change in functional groups. This can be compared with Table 2. Table 2 shows that with the addition of PP-g-MA, there was only a wave shift and it does not appear that a new functional group formed and only the band at 1755 to 1735 cm<sup>-1</sup>, which

indicated the C=O band which was thought to come from the ester produced even though it was only small (an esterification reaction has occurred).

Thus, it can be concluded that the analysis with FTIR can also provide information that there was a chemical interaction between PP, PP-g-MA, and chempedak seed starch based on the change in the wavenumber of each material in the mixture of composites.

#### **4 Conclusion**

Based on the data obtained in this study, it can be concluded that: The high tensile strength and elongation stress values obtained were 28.743 N/m<sup>2</sup> and 28.720%. In addition, the biodegradability analysis shows that the percentage rate of biodegradation of this biodegradable composite was the highest, namely a 5% of reduction in mass in waste soil. The SEM results show that the mixture was flat and homogeneous and the FTIR results show that the band at 1755 to 1735 cm<sup>-1</sup>, which assigned the C=O bond that was derived from the ester produced even though the intensity was small (the esterification reaction has occurred). There has been an esterification reaction between maleic anhydride and the OH group of the raw material (starch), namely chempedak seed starch.

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