

## Tensile Properties and Biodegradability of Rice Husk Powder-filled Recycled Polypropylene Composites: Effect of Crude Palm Oil and Trimethylolpropane Triacrylate

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**ABSTRACT:** Composites of rice husk powder (RHP)-filled recycled polypropylene (rPP) as a function of crude palm oil (CPO) and trimethylolpropane triacrylate (TMPTA) were prepared. Composites containing various amounts of RHP, acrylic acid (AA), CPO and TMPTA were prepared, and the effects on the properties of the resulting composites were investigated. As a function of RHP loading, the tensile strength was enhanced to an optimum value at an rPP to RHP ratio of 7:3. The addition of CPO increased the tensile strength (TS), elongation at break (Eb) and melt flow index (MFI) of the composites. The biodegradation of rPP/AA/RHP bio-composites was also increased with the addition of both CPO and TMPTA.

**Keywords:** Polypropylene, trimethylolpropane triacrylate rice husk powder, crude palm oil, acrylic acid

### 1. INTRODUCTION

Currently, chemical and material products from renewable resources are important for sustainable development.<sup>1-4</sup> The utilisation of renewable resources such as rice husk powder (RHP) as filler materials in polymers such as polypropylene (PP) is being actively pursued<sup>5-14</sup> due to the resulting improvement to the properties of plastic composite materials. The use of RHP as a filler is not only intended to minimise the environmental problems caused by the excessive plastic waste but also to reduce the production cost of the plastic composites.<sup>15-22</sup> Plastic products, especially those used for packaging applications, contribute to a large percentage of household waste. A majority of the waste comes from polypropylene-based mineral water cups, which cause environmental pollution upon disposal. Recycling of this waste product has been conducted extensively to reduce the environmental problems due to excessive

land fillings by non-degradable polymeric materials. As an alternative to recycling, PP waste can also be transformed into other useful products by blending it with other materials to produce composite materials that have a higher economic value and a longer service life.<sup>6-8,12-13,20-22</sup>

Studies on recycled polypropylene (rPP) using its highly reactive methine functional groups, which are capable of reacting with the cellulose of rice husk powder (RHP) in the presence of acrylic acid (AA) as a coupling agent, have been reported.<sup>23-25</sup> The cellulose of RHP contains three polar free hydroxyl groups and two glycosidic carbon groups, making it highly polar, whereas the methine group of rPP is non-polar in nature. The resulting polarity difference between RHP and rPP leads to lower binding and less interaction upon mixing. Modifiers and coupling agents have been widely used in such cases to introduce and improve the interfacial interaction between two phases of different polarity.<sup>17,19,21-28</sup> Coupling agents such as AA, which contains a non-polar vinyl group and a reactive polar hydroxyl group, can be used to improve the binding between RHP and rPP. The methine groups of rPP react with the vinyl groups of AA, while the hydroxyl groups of RHP react with hydroxyl groups of AA, forming an ester linkage that bridges rPP and RHP.<sup>24-26</sup> Further improvement of the interaction between rPP and RHP can be accomplished with the addition of trimethylolpropane triacrylate (TMPTA) as a compatibiliser.<sup>29,30</sup> In addition, crude palm oil (CPO) can be used as a plasticiser to facilitate the mixing and processability.<sup>31,32</sup>

In this study, RHP was used as filler in rPP/AA blends, and the effects of RHP loading on the properties of rPP/AA/RHP composites were investigated. In addition, the effects of TMPTA compatibilisation and CPO plasticisation on the properties of rPP/AA/RHP composites were also studied. The tensile properties, i.e., tensile strength, elongation at break, Young's modulus and the melt flow index (MFI) of all the prepared composites were measured. The degradation properties of the composites were determined via water absorption and soil burial tests. The functional groups present in the resulting composites were determined by Fourier transform infrared (FTIR) analysis to detect and demonstrate the chemical interactions between rPP and RHP in the presence of AA, TMPTA and CPO.

## **2. EXPERIMENTAL**

### **2.1 Materials**

rPP cups were obtained from a plastic recycling centre in Surakarta, Indonesia. Rice husk (RH) was collected from a rice milling centre in Kartasura, Indonesia. CPO was purchased from P.T. Ramayana trading company in Surakarta and was used without any further purification. All raw materials, such as AA, TMPTA, benzoyl peroxide (BPO) and acetone, were obtained from the Aldrich Chemical Company (St. Louis, U.S.A) in pro-analysis grade, except where mentioned, and used as received without any further purification. The burial soils (mixture media of garbage dump soil and cattle waste [1:1, w/w]) were collected from Putri Cempo landfill in Surakarta, Indonesia.

### **2.2 Preparation of rPP and RHP**

The rPP cups were washed with water and dried, then cut into small pieces with dimensions of approximately 2 mm × 2 mm. The RHP was cleaned with water, then washed with ethanol and dried in an oven at 40°C before grounded into 150 mesh sized particles.

### **2.3 Preparation of RHP-filled rPP Composites**

rPP composites were prepared using a laboplastomill internal mixer from Toyo Seiki Japan at temperature 180°C and mixing time of 10 min. Five different weight ratios of rPP to RHP were used to prepare the composites: 10/0, 9/1, 8/2, 7/3 and 6/4 with respect to the rPP and RHP content. The compositions of the other materials were as follows: 0.03 phr of BPO initiator, 2 phr of CPO, AA (10% of RHP weight) and TMPTA (10% of AA weight). The resulting composites were prepared for testing by compression moulding using a hot press (Paul Weber Maschinen-u-Aparatebau D-7064 Rhemshaldel-Brunbach) at 180°C for 5 min.

### **2.4 Composites Testing**

The melt flow index of the composites was determined using an Atlas melt flow indexer, in accordance with the Standar Nasional Indonesia (SNI 06-0528-1989). Tensile strength (TS), elongation at break (Eb) and Young's modulus (YM) were measured in accordance to ASTM D638 using a tensiometer (MPG SC-2 DE). Water absorption tests were carried out according to ASTM D570-95. Soil burial tests were carried out in cellulolytic bacteria-enriched landfill soil for four months to determine the biodegradability of the composites.<sup>3,25,27</sup> FTIR analysis was conducted to characterise the functional

groups present in the composites using a Shimadzu FTIR-8201 PC FTIR analyser with potassium bromide (KBr) disks or films. Morphological observations of the exposed surfaces of the composites after soil burial tests were carried out using a scanning electron microscope (SEM) model JSM-35C.

### 3. RESULTS AND DISCUSSION

#### 3.1 FTIR Analysis of Raw Materials and rPP/TMPTA/AA/RHP/CPO Bio-composites

The FTIR spectra of the raw materials are shown in Figure 1. The characteristic peaks of rPP appeared at 2962 and 1373  $\text{cm}^{-1}$ , representing the  $-\text{CH}_3$  methyl groups. The absorption peak at 2723  $\text{cm}^{-1}$  indicates  $>\text{C}-\text{H}$  methine stretching (typical of PP), while the absorption at 2890  $\text{cm}^{-1}$  is due to  $-\text{CH}_2-$  stretching, and the peaks at 1454 and 1165  $\text{cm}^{-1}$  are attributed to  $-\text{CH}_2-$  bending. The FTIR spectrum of AA in the neat liquid shows a broad absorption band at 3448  $\text{cm}^{-1}$  due to  $-\text{OH}$  hydrogen bonding, and the absorption at 1728  $\text{cm}^{-1}$  is from the carbonyl group of AA. The absorption peak at 2890  $\text{cm}^{-1}$  is attributed to the  $>\text{CH}_2$  methylene vinyl group. The absorption peak at 1412  $\text{cm}^{-1}$  indicates the presence of the acrylic  $>\text{C}=\text{CH}_2$  group.

The FTIR spectrum of RHP is also shown in Figure 1, which demonstrates a broad absorption band at 3448  $\text{cm}^{-1}$  from the cellulose  $-\text{OH}$  hydrogen bonds, a band at 1651  $\text{cm}^{-1}$  from the  $>\text{C}=\text{C}<$  aromatic lignin of RHP, and a peak at 1049  $\text{cm}^{-1}$  representing the  $>\text{C}-\text{O}-\text{C}<$  stretching from the cellulose of RHP. The FTIR spectrum of CPO contains the following: an absorption band at 3458  $\text{cm}^{-1}$  due to  $-\text{OH}$  hydrogen bonding; a peak at 2942  $\text{cm}^{-1}$  from  $-\text{CH}_2-$  stretching; and an absorption band at 1742  $\text{cm}^{-1}$  indicating the presence of  $>\text{C}=\text{O}$  groups.<sup>25,26</sup> The intensity of the absorption at 3458  $\text{cm}^{-1}$  due to hydrogen bonding  $-\text{O}-\text{H}$  groups of CPO is lower compared to those from AA and RHP. Hydrogen bonding in CPO occurs only between free fatty acids (FFA), which are present at low concentrations in CPO. However, pure AA compounds are capable of forming hydrogen bonds with RHP, which has cellulose as its main component, and each of its constituent monomers have three hydroxyl groups and three alkoxy groups, which are capable of forming hydrogen bonds.

Figure 2 shows the FTIR spectra of the rPP/AA/RHP, rPP/TMPTA/AA/RHP and rPP/TMPTA/AA/RHP/CPO composites. The spectra reveal a shift of the absorption peak of the carbonyl group of AA from 1728  $\text{cm}^{-1}$  to 1732  $\text{cm}^{-1}$ , which represents the absorption peak of the ester group resulting from the coupling of AA to both rPP and RHP. The absorption peak at 1747  $\text{cm}^{-1}$  represents the carbonyl group of CPO. In addition, it can be observed that the

absorption peak of the  $>C=CH_2$  acrylic double bond at  $1412\text{ cm}^{-1}$  was eliminated due to the reaction between the vinyl double bond and the methine group of rPP.<sup>25,26</sup> Hence, this result demonstrates that a reaction between rPP and RHP occurred in the presence of AA. The addition of 2 phr CPO as a plasticiser does not produce any significant changes in the hydrogen bonding intensity, as evidenced by the absorption of the hydroxyl group at  $3458\text{ cm}^{-1}$ , and in addition, the visible appearance of the absorption peak due to the triglyceride carbonyl groups of CPO at  $1747\text{ cm}^{-1}$  are quite significant. The addition of TMPTA, which contains three acrylate ester carbonyl groups, increases the intensity of the absorption peak of the acrylic carbonyl ester group of AA-RHP at  $1732\text{ cm}^{-1}$ .

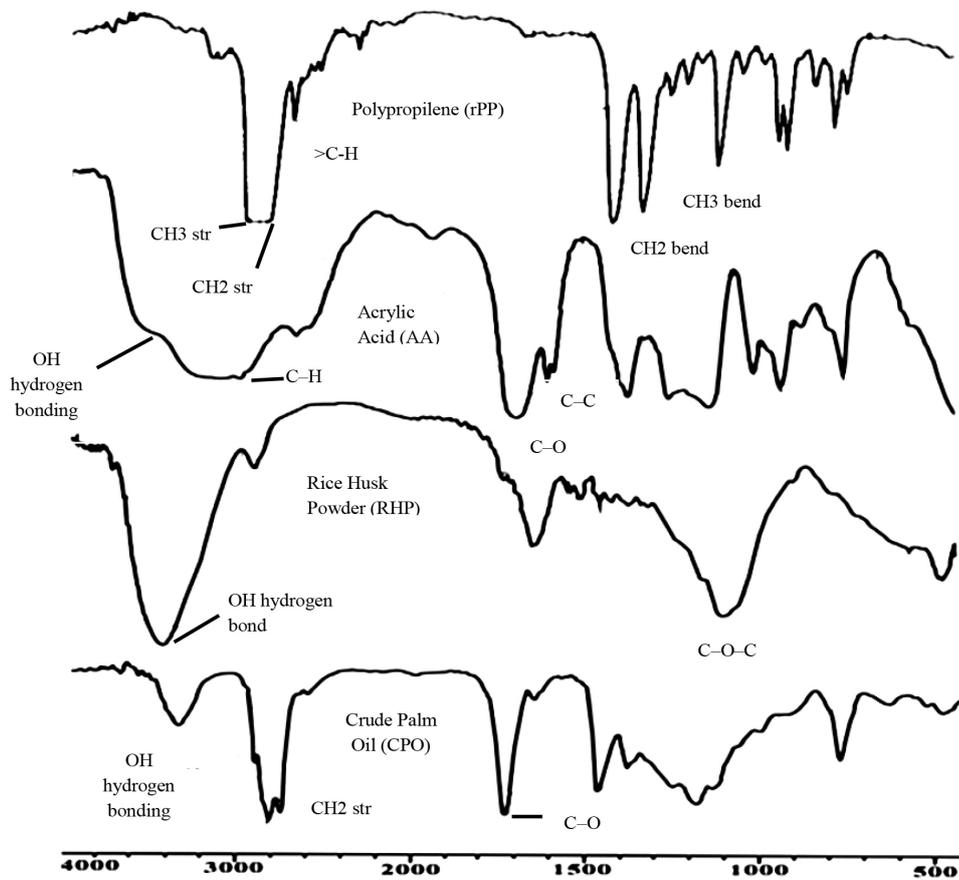


Figure 1: Raw material FTIR spectra of rPP, AA, RHP and CPO.

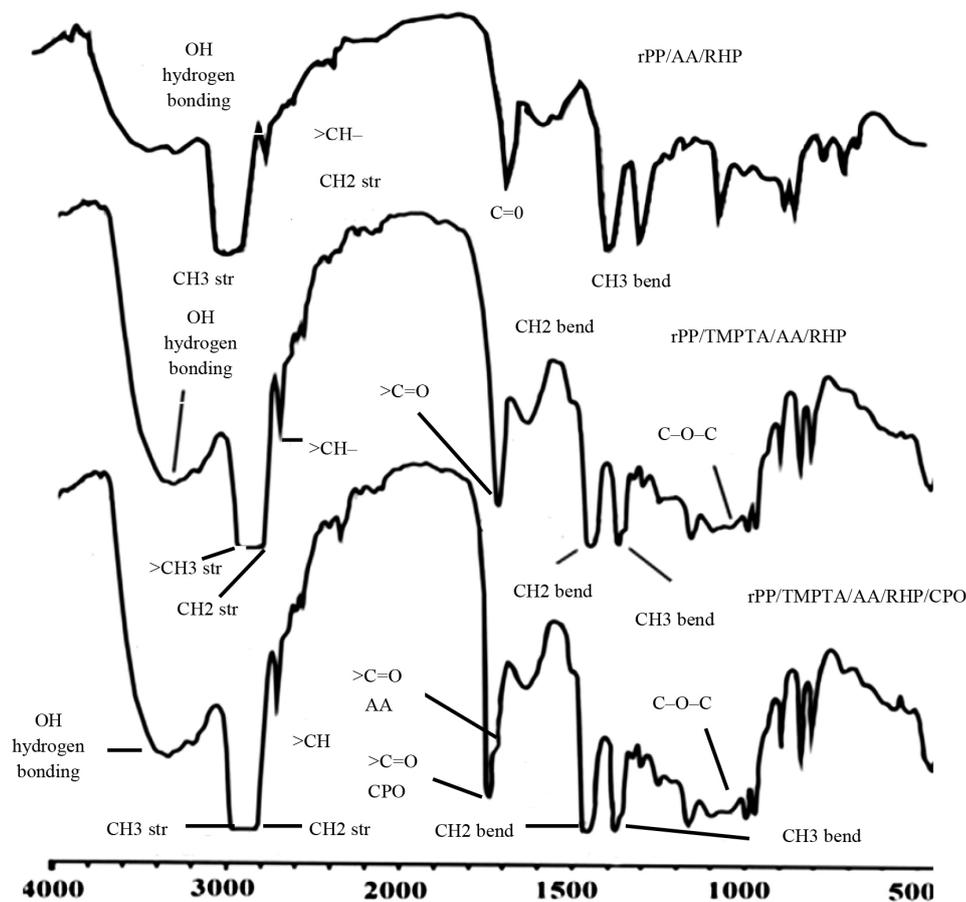


Figure 2: FTIR spectra of rPP/AA/RHP, rPP/TMPTA/AA/RHP and rPP/TMPTA/AA/RHP/CPO bio-composites at rPP/RHP weight ratio of 7/3.

### 3.2 Tensile Strength, Elongation at Break, Young's Modulus and Melt Flow Index

The optimum composition of rPP to RHP was determined by evaluating properties of the composites, i.e., tensile strength (TS) and melt flow index (MFI), and identifying which composites had the highest values. Due to the polarity difference between rPP and RHP, multifunctional compatibilising materials such as AA and TMPTA were used to enhance the compatibility between rPP and RHP. The composites were prepared by varying the rPP to RHP weight ratio, with and without the addition of 2 phr CPO plasticiser. It was found that the optimum composition of rPP to RHP required to produce composites with optimum properties was a rPP/RHP weight ratio of 7/3. Composites with a rPP/RHP ratio of 7/3 exhibited MFI values in the thermoplastic region (Figure 3),

and the highest TS value as shown in Figure 4. As shown in Figure 3, the MFI value of the rPP/TMPTA/AA/RHP/CPO composite is 43.7% higher than that of composites without CPO.

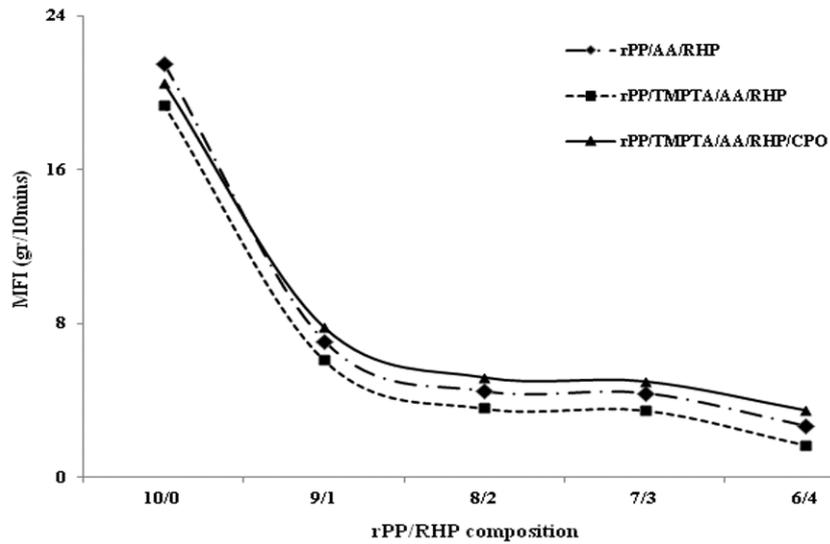


Figure 3: MFI of rPP/AA/RHP bio-composite as a function of TMPTA and CPO at various rPP/RHP ratios.

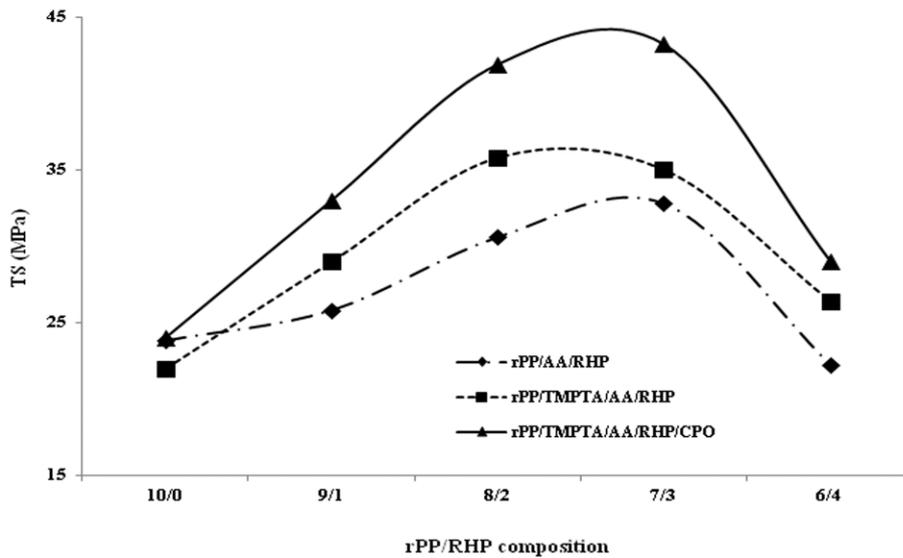


Figure 4: Tensile strength of rPP/AA/RHP bio-composite as a function of TMPTA and CPO at various rPP/RHP ratios.

It was also observed that the TS and elongation at break (Eb) of composites containing CPO was higher by 21.0% and 1.3%, respectively, compared to the composite without CPO, as shown by Figure 4 and 5, respectively. Furthermore, the Young's Modulus (YM) of rPP/TMPTA/AA/RHP/CPO (Figure 6) was slightly lower, indicating increased plasticity compared to the composite without CPO. In the presence of CPO as a plasticiser, mixing and processing of the composite was facilitated, thereby allowing for more efficient dispersion and distribution of RHP particles, and promoting greater interaction and chemical binding between RHP and rPP. Addition of compatibiliser TMPTA and coupling agent AA subsequently improved the TS and Eb and reduced the YM of the rPP/RHP composite with CPO.

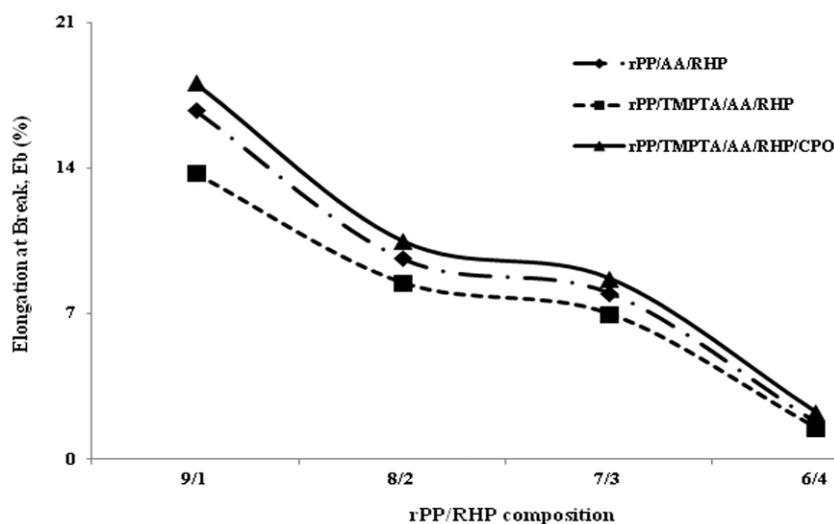


Figure 5: Elongation at break of rPP/AA/RHP bio-composite as a function of TMPTA and CPO at various rPP/RHP ratios.

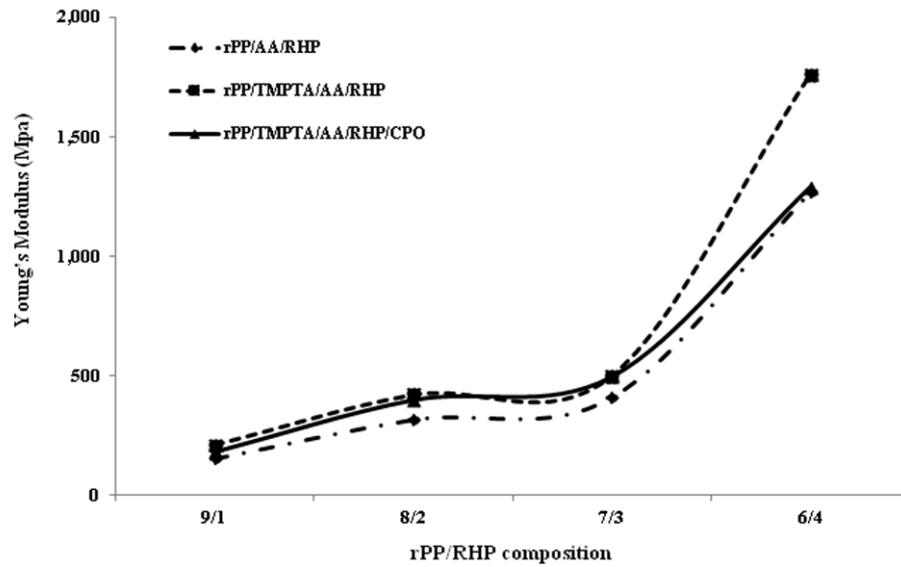


Figure 6: Young's modulus of rPP/AA/RHP bio-composite as a function of TMPTA and CPO at various rPP/RHP ratios.

### 3.3 Water Absorption Properties

The water absorption properties of raw rPP and rPP/AA/RHP composites as a function of TMPTA and CPO plasticiser content at a rPP/RHP ratio of 7/3 were studied to understand the behaviour of the composite. Thin films of each composite were compression moulded, and water absorption tests were conducted. The composite films were soaked in water for 24 h, and the percentage of water uptake by the films was determined by measuring the initial and final weights of the composite films. The presence of RHP (cellulose) improves the hydrophilicity of the rPP composite, allowing more water absorption due to ease of hydrogen bonding between RHP and water molecules. Figure 7 shows that rPP does not absorb water, while RHP-filled rPP composites absorb water depending on the amounts of the other materials added. At an rPP/RHP ratio of 7/3, the bio-composites absorb water with or without the addition of TMPTA or CPO.

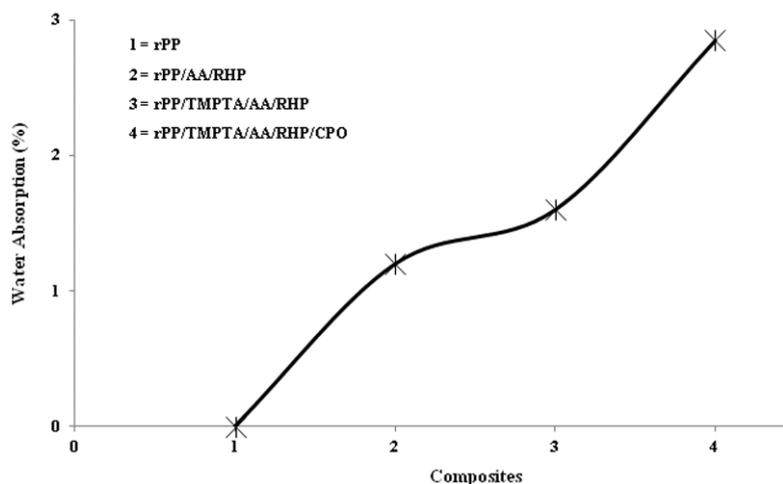


Figure 7: Water absorption percentage of raw rPP and rPP/AA/RHP bio-composites as a function of TMPTA and CPO at rPP/RHP ratio of 7/3.

The water absorption capacity of rPP was increased with the addition of RHP in the presence of AA, followed by the further addition of TMPTA. rPP/AA/RHP composites containing CPO exhibit slightly higher water absorption compared to that of rPP/AA/RHP and rPP/AA/RHP/TMPTA composites. However, the rPP/AA/RHP composites containing both TMPTA and CPO had the highest water absorption capacity, approximately 2.8% higher than that of raw rPP. The presence of the compatibiliser TMPTA and the plasticiser CPO improved the dispersion and distribution of the RHP particles in the process. This composition also increased the number of chemical reactions between the raw materials: PP matrix, TMPTA compatibiliser, RHP enforcement, and multifunctional AA, which is reactively processed by the BPO initiator. The more efficient distribution of RHP particles throughout the composite allows the RHP to absorb more water.

### 3.4 Degradation Properties of the Composites

The biodegradation properties of rPP/AA/RHP composites were studied by soil burial tests in a mixed medium of landfill soil and cattle waste (1:1, w/w) for a period of 4 months. The weight loss of the specimens after a 4 month burial period was measured according to Suharty et al.<sup>3,24,25,27</sup> which indicates the extent of bio-degradation experienced by the composites. As illustrated by Figure 8, raw rPP exhibited no reduction in weight after 4 months. However, other rPP composites containing RHP, TMPTA and CPO underwent weight loss, indicating the occurrence of degradation via microbial activities present in the soil burial medium.

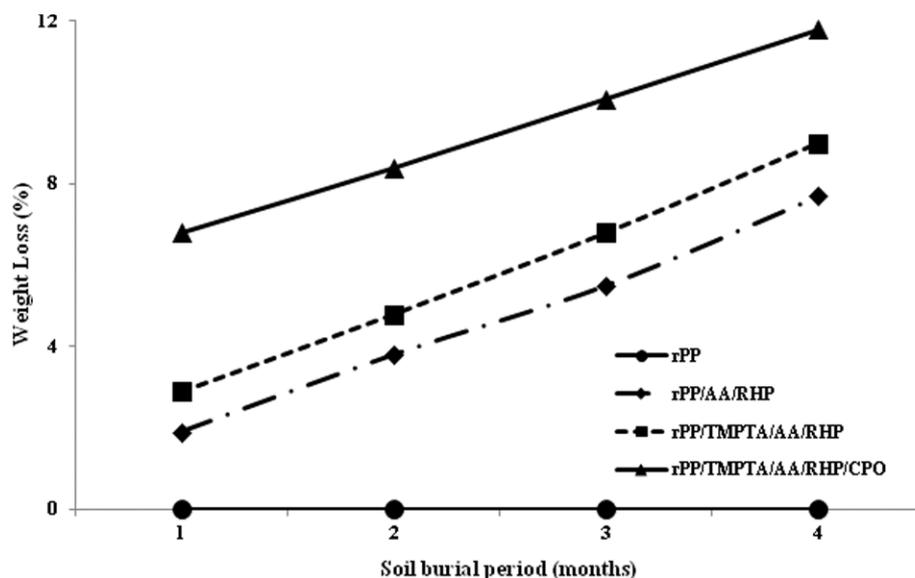


Figure 8: Weight loss percentage of raw rPP and rPP/AA/RHP bio-composites as a function of TMPTA and CPO at rPP/RHP ratio of 7/3.

From Figure 8, it can be observed that the rPP/TMPTA/AA/RHP/CPO composite exhibited the greatest weight loss, followed by the rPP/TMPTA/AA/RHP composites. In addition, the rPP/AA/RHP composite exhibited the least amount of weight loss among the composites studied. The weight loss results of the composites revealed the extent of microbial attack and degradative reactions after exposure for 4 months, resulting in the breakdown of the PP molecular chains in the presence of RHP and other materials. This attribute would be beneficial for the disposal of rPP into landfills in the future. These data are in good agreement with and are supported by the water absorption observation in Figure 7.

### 3.5 SEM Morphological Observation

The SEM morphological study was performed on the composite specimens before and after soil burial tests to determine the extent of biodegradation after exposure of the composites to degradation. The composites were transparent and white in colour with a smooth surface before degradation. After degradation, the specimens became grey in colour with a coarse surface. SEM images of the rPP/TMPTA/AA/RHP/CPO composite before and after degradation are shown in Figure 9. As shown, the specimen before degradation [Figure 9(a)] possesses a smoother surface, while the surface became brittle and rough with voids and cracks after degradation [Figure 9 (b)]. The presence of

water and cellulolytic bacteria in the mixed medium of landfill soil and cattle waste caused the bio-degradation of the composite.<sup>3,4,24,25,27</sup>

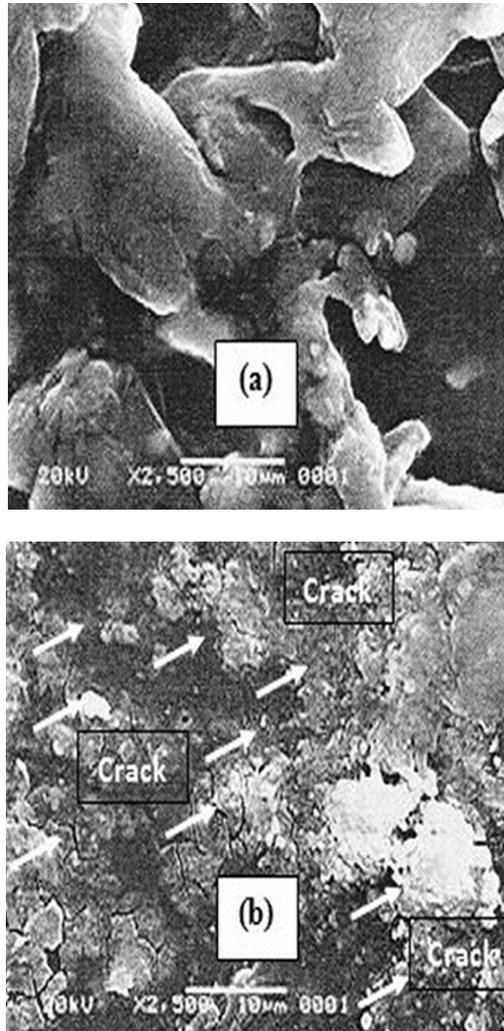


Figure 9: SEM images of rPP/TMPTA/AA/RHP/CPO bio-composite: (a) before and (b) after degradation (2,500 times magnification).

For the rPP/AA/RHP/CPO composite, the SEM morphological observations at a higher magnification factor of 20,000 appear similar to the composite in Figure 9. At a higher magnification, the number of cracks after degradation can be observed clearly, which indirectly demonstrates the lower density of the bio-composite after degradation. From Figure 10 (a) and (b), the number of cracks before and after degradation can be determined. It can be

observed that before degradation, the rPP/AA/RHP/CPO composites without TMPTA had 16 crack sites with a total crack area of  $10 \times 10^4 \text{ nm}^2$  [Figure 10(a)]. However, the number of cracks increased after the degradation process, enlarging the total crack area by 450%, to  $45 \times 10^4 \text{ nm}^2$ .

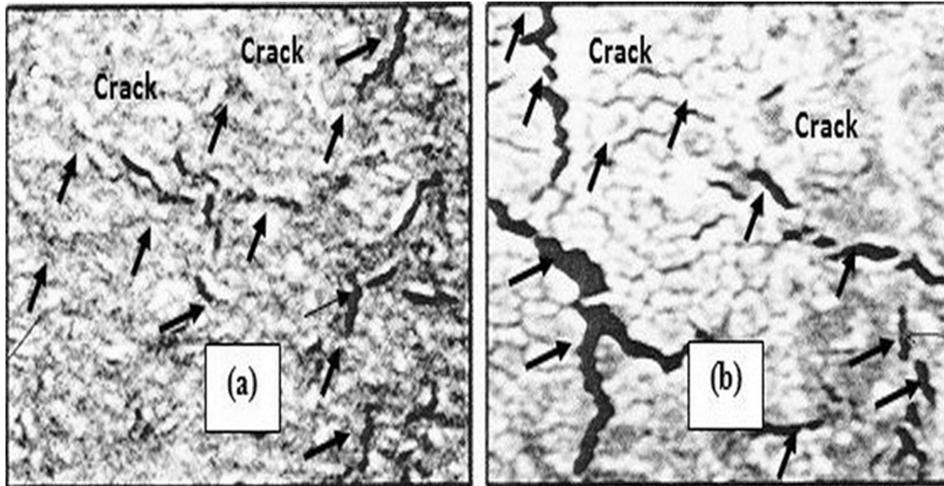


Figure 10: SEM images of rPP/AA/RHP/CPO bio-composite: (a) before and (b) after degradation (20,000 times magnification).

Figure 11 shows SEM images of the rPP/TMPTA/AA/RHP/CPO composite with TMPTA. It can be observed that with the addition of TMPTA, the number of cracks at the specimen surface was less than that of the sample without TMPTA, at which 12 crack sites with a total crack area of  $8 \times 10^4 \text{ nm}^2$  [Figure 11 (a)] were observed before degradation. This result indicates that the composite with TMPTA added is compact and has a high density. However, after the degradation process, the total crack area increased to  $61 \times 10^4 \text{ nm}^2$  [Figure 11 (b)] with an increment of 763% after degradation, which was approximately 136% higher than that of composites without TMPTA.

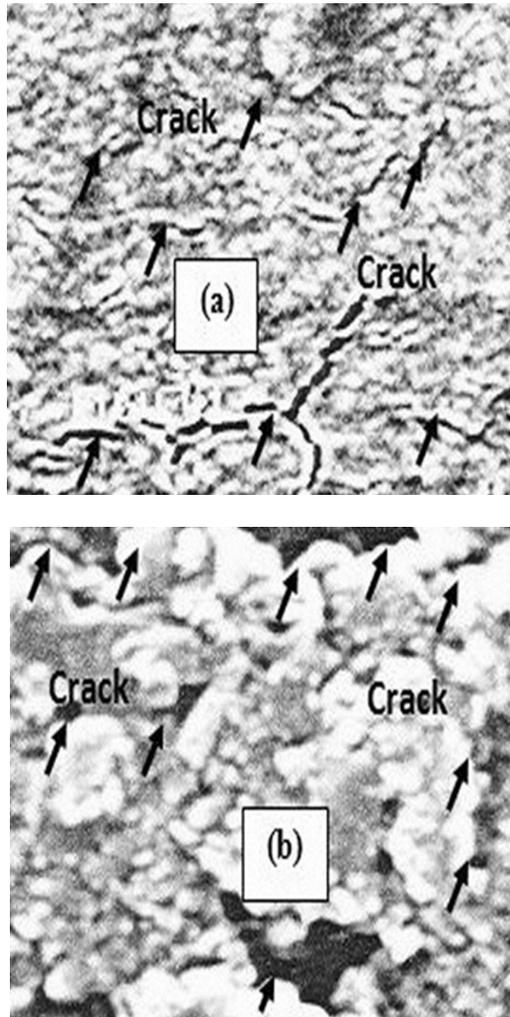


Figure 11: SEM images of rPP/TMPTA/AA/RHP/CPO bio-composite: (a) before and (b) after degradation (20,000 times magnification).

#### 4. CONCLUSION

The optimum composition weight ratio of rPP/RHP to prepare the rPP/TMPTA/AA/RHP/CPO composite with optimum properties was 7/3. The tensile strength, elongation at break and MFI of the rPP/AA/RHP composites were improved with the addition of CPO. The water absorption of the composites with CPO was higher than that without CPO, which was further increased by the addition of TMPTA. The biodegradation of the rPP/AA/RHP composites, as

determined by weight loss, was increased with the addition of TMPTA and CPO, but the greatest extent of degradation was observed with the rPP/TMPTA/AA/RHP/CPO composite.

## 5. ACKNOWLEDGEMENT

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