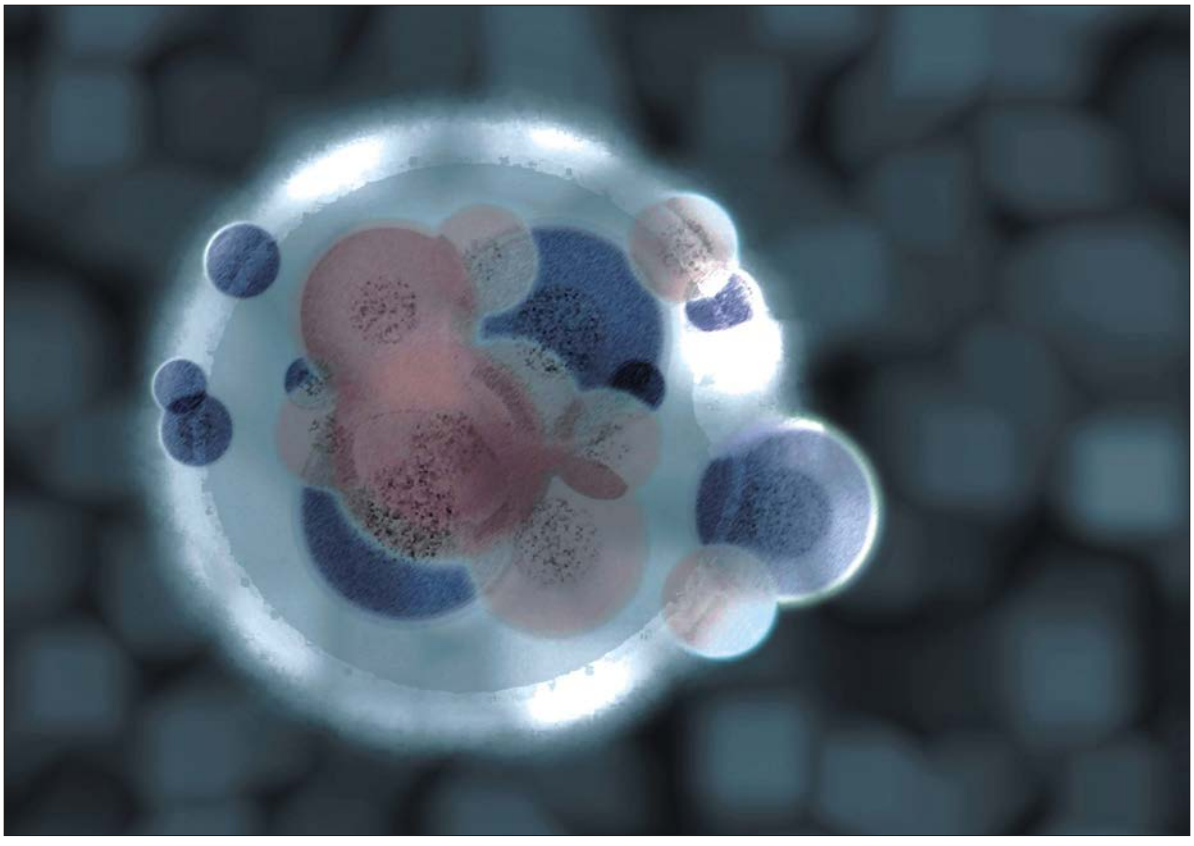

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Influence of Wood Flour and Modifier Contents on the Physical and Mechanical Properties of Wood Flour-Recycle Polypropylene Composites

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Abstract: The objectives of this research was to evaluate the physical and mechanical properties of composites of Wood Flour (WF) and Recycle Polypropylene (RPP) prepared under various WF contents, WF sizes and modifier contents. WF from mixed-soft wood species and *Eucalyptus deglupta* Blume species were used as filler. The composites composed of 0-70% WF, 0-70% Polypropylene (PP) or RPP, various sizes of WF (60-80; 80-100 and < 120 mesh), various contents of MAH modifier (0, 1, 2.5, 5, 7.5 and 10%) and 15% Dicumyl Peroxide (DCP) initiator (based on MAH weight). Kneading conditions were set at 170°C, 10-50 rpm for 15 min. The physical and mechanical properties of composites were greatly affected by WF content, WF size and modifier content. The greater the WF loading resulted in the greater was the reduction of tensile strength and breaking elongation values and at the same time the greater was the increasing of Young's modulus value. The smaller the WF size resulted in the greater the tensile strength of composites. Addition of MAH modifier improved the physical and mechanical properties of composites. WF-RPP composites with 120 mesh WF size and 2.5% MA modifier had tensile strength, breaking elongation and Young's modulus about 2.15, 2.27 and 1.18 times, respectively higher compared with composites free-MAH modifier and absorbed considerably less water. Scanning Electron Microscope (SEM) indicated that addition of MAH improved the adhesion between WF and RPP.

Key words: Wood flour, recycle polypropylene, composite, modifier

INTRODUCTION

It is not too exaggerated to say that wood is one of the most important products of nature. Wood is not only a renewable and biodegradable resource, but also predominant source of lignocelluloses materials most extensively used in decoration and in the furniture industries. The insatiable world demand for good quality timber has led to destructive logging of tropical hardwood forest in many developing countries and gave rise to serious global concern. As rain forest supplies shrink while development accelerates, it is expected to lead to a sharp rise in the cost of natural timber products in the near future. Thus, there is an increasingly urgent need for developing high quality timber substitute products that can be of benefit to the global industrial and business development, with due regard for the world environment.

Wood Plastic Composites (WPC) are a new group of materials that are generating interest in many applications. The term WPC covers an extremely wide range of composite materials using plastics ranging from PP to Poly Vinyl Acetate (PVA) both virgin or recycles and fillers ranging from WF to agriculture fibers. Compared with reinforced thermoplastic products, WPC have many advantages such as high specific strength and modulus, low cost, low density and low friction during compounding. Unlike wood composites, WPC have excellent dimensional stability under moisture exposure and better fungi and termite resistance. For WPC, one of the most attractive features is that it can help recycle thermoplastic and wood

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wastes. Therefore, WPC have developed quickly in the last two decades (Youngquist, 1995).

It is known that affinity and adhesion between thermoplastic matrices and wood are poor; the former are hydrophobic and the latter contains hydrophilic cellulose and hemicelluloses in quantities of 60-80%. Accordingly, these blended composites show disadvantages in thermal flow ability and moldability and in unsatisfactory reinforcement. Gross segregation of the blend component forming a heterogeneous structure and the lack of interfacial adhesion resulted in an inability to transfer imposed stresses between these phases (Scheiner and Brebner, 1985; Kishi *et al*., 1989; Febrianto *et al*., 1999). It has been reported in numbers of literatures that MAH can be introduced into polymers, such as PP, Polyethylene (PE) and acrylonitrile butadiene styrene copolymer (ABS) through radical coupling addition to form MAH-modified polymer in the presence of peroxide (Scheiner and Brebner, 1985; Kishi *et al*., 1989; Han *et al*., 1989; Gatenholm *et al*., 1993; Febrianto *et al*., 1999). Then, the MAH-modified polymer can further react with the polymer having -OH or NH_2 end-groups to form long chain graft copolymers that can be used as compatibilizers of the polymer blends. MAH introduced in this way into synthetic polymers has been proved to form ester linkages when reacting with OH-groups on the WF interface. Thus, composites resulted from blends of thermoplastic or elastomer matrices and woody filler in the presence of compatibilizer have been proved to have excellent and mechanical (tensile strength, breaking elongation and Young's modulus) and physical (water absorption) properties (Han *et al*., 1989; Gatenholm *et al*., 1993; Febrianto *et al*., 1999).

Thermoplastic also bring a serious problem to environment after service because of its decomposition difficulty under natural conditions. Accordingly, thermoplastic wastes cause serious environmental pollution all over the world every year. For example the amount of plastics annually consumed in the United States is around 100 kg per person. Among plastics, the big four plastic products (Polyethylene, poly propylene, poly vinyl chloride and poly styrene were accounted about 92% by volume of the whole plastics consumption in the United States. Therefore, to reduce the negative impact of thermoplastics waste to environment, it is urgently need to utilize the waste plastics as a matrix resin for WPC (Chernier, 1992).

In this article, virgin and first recycle of polypropylene were used as matrices resin for Wood Flour (WF)-Polypropylene (PP)/ Recycle Polypropylene (RPP) composites. Maleic Anhydride (MAH) as a modifier together with Dicumyl Peroxide (DCP) as an initiator were directly introduced to the molten matrix when blending WF-RPP composites. The effect of WF content, WF size and modifier content on mechanical (tensile strength, breaking elongation and Young's modulus) and physical (water absorption and thickness swelling) properties of composites were investigated.

MATERIALS AND METHODS

Materials: The matrices used in these experiments were PP (J-130 G/MI = 31) and RPP. First recycle polypropylene was collected from Bogor district, West Java, Indonesia. WF of *Eucalyptus deglupta* Blume species and mixed of softwood were used as fillers with 3 sizes: 60-80 mesh; 80-120 mesh and less than 120 mesh. The moisture content of WF was < 5%. MAH and DCP were used as a modifier and an initiator, respectively. WF-RPP composites composed of 50% WF and 50% RPP and various content of MAH ranging from 0-10% (0; 1; 2.5; 5; 7.5 and 10 %) with 15% DCP (based on MAH weight). This research was conducted in Indonesian Applied Physics Research Centre, Bandung and in laboratory of Bio-composites Forest Products Department Faculty of Forestry, Bogor Agricultural University, Indonesia, from September, 2002 to February, 2003.

Compounding the composites: PP or RPP was placed into the kneader (Toyo-Seiki Labo Plastomil LPM 18-125) at 170°C, 10 rpm for 1 min. After charging the RPP, MAH and DCP were added into the molten RPP within 1 min. After further reaction for 1 min, the WF was added within 3 min at 30 rpm. The rate of rotation was increased at 50 rpm and kneading was continued for 9 min. The total kneading time was 15 min and the amount of RPP and WF were 48 g. For composite-free MAH modifier, after charging the RPP for 1-2 min at 170°C at 10 rpm, WF was added for 3 min at 30 rpm. The rate of rotation was increased at 50 rpm and kneading was continued for 10 min.

Preparation of composites sheets: Kneaded samples were molded into sheet by hot pressing. The prescribed amount of kneaded samples (7-10 g) was placed between a pair of polyester film with 0.3 mm thick spacer. The temperature of hot press was 180°C and the samples were subjected to 0-100 kgf cm^{-2} for 1-2 min. After subsequent cold pressing at the same pressure for 30 sec, the sheets were then cooled at room temperature.

Preparation of specimen for tensile test: Strips samples were prepared from composite sheets in accordance to ISO 527-2:1993 (E) type A standard. The average values

of tensile strength, breaking elongation and Young's modulus were obtained from 5 repeated measurements.

Water absorption and thickness swelling tests: Square samples of 50.0×50.0×0.3 mm size were prepared from the composite sheets. Water absorption and thickness swelling tests were made on these square samples in three replications. The samples were dried overnight in the vacuum oven at the temperature of 60°C and then stored in a desiccator. The weight and the thickness (W1 and D1) of the samples were measured in a room adjusted to 20°C and 60% R.H. Subsequently, the samples were dipped for 24 h to water that had been conditioned at a room adjusted to 20°C, 60% R.H. for 48 h. Then, the samples were wiped and the weight and the thickness (W2 and D2) were measured. Finally, the samples were dried in the 60°C oven in vacuous to obtain the constant weight (W3). Water absorption and thickness swelling were calculated as shown in the following Eq. 1 and 2.

$$\text{Water absorption} = \frac{W2 - W3}{W1} \times 100\% \quad (1)$$

$$\text{Thickness swelling} = \frac{D2 - D1}{D1} \times 100\% \quad (2)$$

Scanning Electron Microscopy (SEM) observation: The morphology of the tensile fractures and surface of the WF-RPP composites were studied by using A JEOL JSM-T330 a scanning microscope. The samples were dried before coating to remove the sorbed water. SEM photographs were taken under the following conditions: working distance 15 mm and accelerating voltage of 10 Kv.

RESULTS AND DISCUSSION

The mechanical and physical properties of WF-PP or RPP composites under various WF contents and WF sizes: In this part of work, the composites were prepared from mixed softwood WF with the size of < 120 mesh and virgin PP. The ratios of WF and PP were set 0:100; 10:90; 20:80; 30:70; 40:60; 50:50; 60:40 and 70:30.

Figures 1-3 exhibited that the tensile strength, breaking elongation and Young's modulus of WF-PP composites were significantly influenced by WF loadings. As compared to PP film alone, addition of WF onto the matrix PP resulted in linearly decreasing both tensile strength and breaking elongation values and linearly increasing in Young's modulus value. The higher the filler loading resulted in the greater is the reduction of tensile strength and breaking elongation values. On the other hand, the Young Modulus value increased proportionally as the WF loading increased. This is the common observation with almost all filled polymer system. Reduction in tensile strength and breaking elongation were probably caused by a number of reasons including: (i) poor dispersion of the fibers in the matrix; (ii) moisture pick up, (iii) increases of interfacial defect or debonding between PP and WF. As for the first reason, WF fibers tended to cling together, due to strong inter fiber hydrogen bonding and resisted dispersion of the individual fiber as the fiber content was increased.

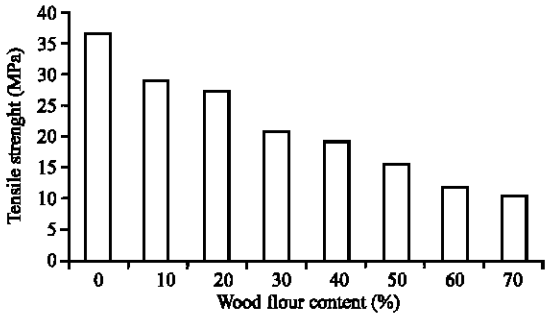

Fig. 1: Tensile strength of WF-PP composites under various WF contents

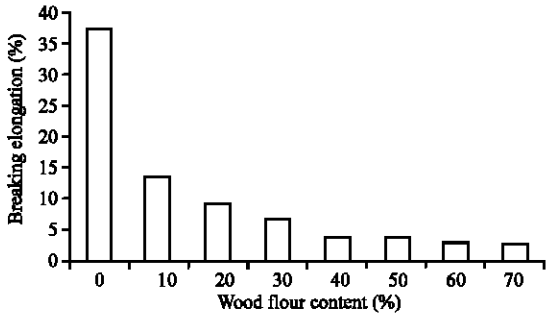

Fig. 2: Breaking elongation of WF-PP composites under various WF contents

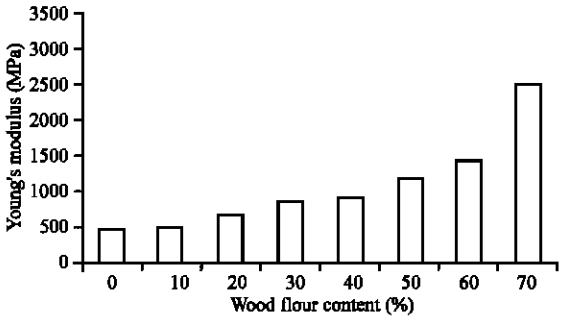

Fig. 3: Young's modulus of WF-PP composites under various WF contents

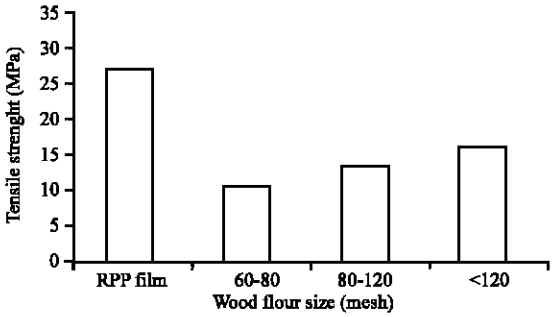

Fig. 4: Tensile strength of WF-PP composites under various WF sizes

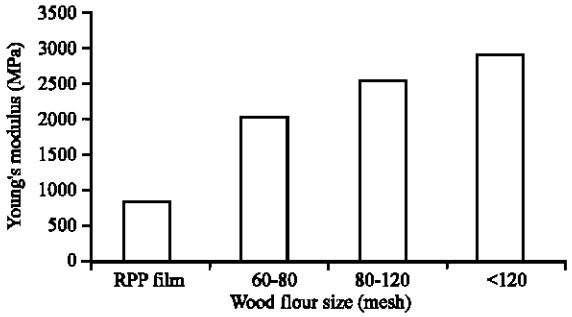

Fig. 6: Young's modulus of WF-PP composites under various WF sizes

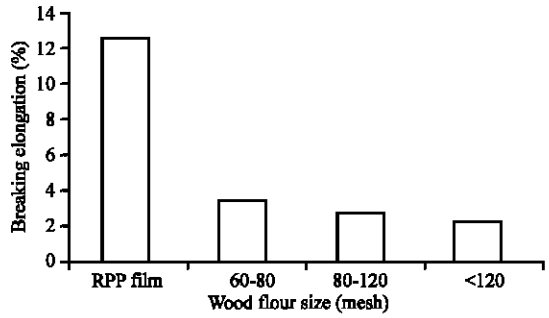

Fig. 5: Breaking elongation of WF-PP composites under various WF sizes

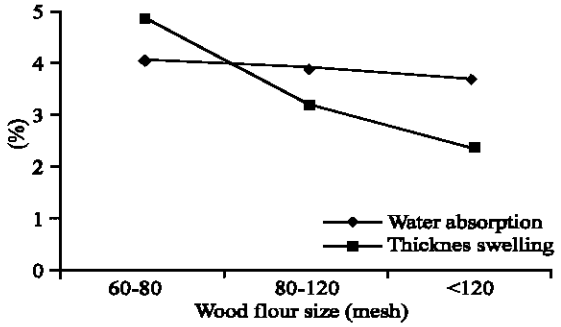

Fig. 7: Water absorption and thickness swelling of WF-RPP composites under various wood flour sizes

In the second case, since the WF is hydrophilic in nature and was chemically untreated (in this part of the experiment), the fibers may have picked up moisture during storage, processing and testing. It was thought in this work that the moisture may have interfered with the adsorption effects by reducing the effect of physical bonding and potentially acting as a lubricant between the fiber surface and PP phases. The last reason for interfacial defects and debonding between PP matrix and WF filler can be proven using a SEM micrograph, which is shown in Fig. 12A clean fiber surface is seen from the fracture surface, which indicates no interfacial interaction between the PP matrix and WF filler. On the other hand, substantial improvement of the Young's modulus was found as the WF added. This is also common phenomenon. i.e., WF addition resulted in greater modulus. This is probably caused by the fact that the WF is more rigid that PP matrix polymer. Similar results phenomena had been published by Han *et al.* (1989) using virgin PP matrix and 3 types of WF contents (0, 30 and 50%) and mixing with Toyo-Seiki Labo Plastomil LPM 18-125; by Febrianto *et al.* (1999) using *trans*-1.4-isoprene rubber matrix and 8 type WF (0, 10, 20, 30, 40, 50, 60, 70%) and mixing with similar labo plastomil and by Sombatsompop *et al.* (2005) using virgin PP matrix and 4 types wood sawdust filler (0, 10, 20 and 30%) and mixing with twin screw extruder (Haake Polylab-Rheomix CTW 100P, Germany). Based on the above results, for obtaining good dispersion of WF filler in the PP matrix using Toyo-Seiki Labo Plastomil LPM 18-125 mixer, the ratio of filler and matrix was 50:50.

The physical and mechanical properties of WF filler and thermoplastic or elastomer matrices composites were also much affected by WF size. Hence, in this part of experiment, the effect of 3 types of *Eucalyptus deglupta* Blume species WF (i.e., 60-80 mesh; 80-120 mesh and less than 120 mesh) size on the physical and mechanical properties of WF-RPP composites was observed. The ratio of WF and RPP was set 50:50. Figures 4-6 showed the tensile strength, breaking elongation and Young's modulus values of WR-RPP composites under various WF sizes. It is also clear from Fig. 4-6 that tensile strength, breaking elongation and young's modulus values varied with WF sizes. The smaller the WF size resulted in the greater the values of tensile strength, Young's modulus and breaking elongation. Similar results have been reported using virgin polypropylene matrix (Han *et al.*, 1989; Zaini *et al.*, 1996). The smaller the WF size resulted in the more homogeneous dispersion of the filler in the matrix polymer and the greater the contact between WF and polymer matrix. As a result the composites have better tensile properties.

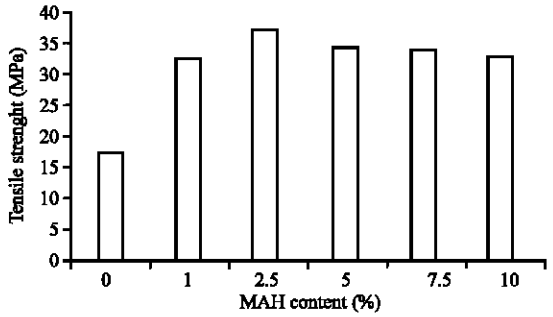

Fig. 8: Tensile strength of WF-RPP composites under various MAH contents

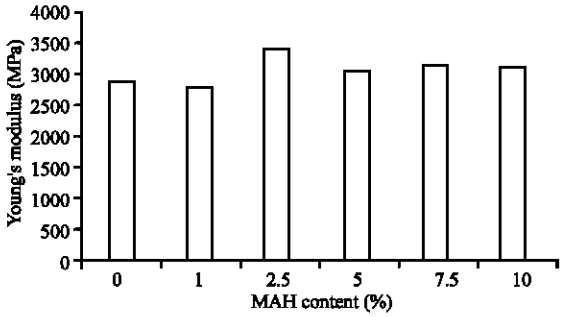

Fig. 10: Young's modulus of WF-RPP composites under various MAH contents

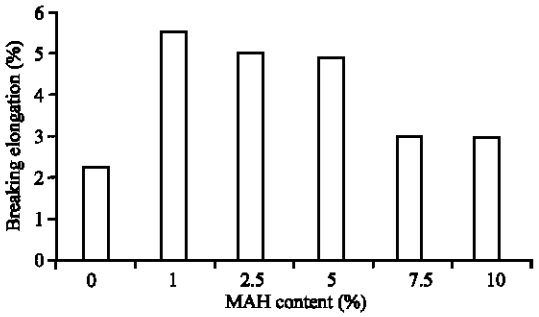

Fig. 9: Breaking elongation of WF-RPP composites under various MAH contents

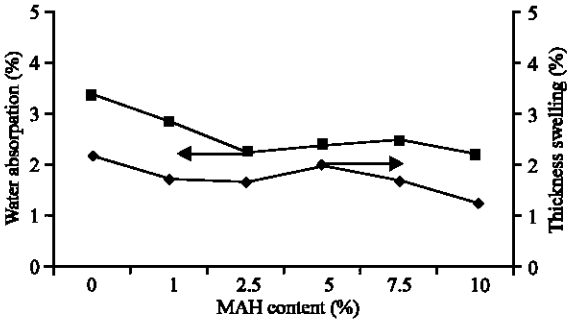

Fig. 11: Water absorption and thickness swelling of WF-RPP composites under various MAH contents

Fig. 7 showed the water absorption and thickness swelling values of WF-RPP composites under various WF sizes. Similar phenomenon with the mechanical properties occurred. The values of water absorption and thickness swelling of RPP film which is hydrophobic in nature were negligible. As a result, the values of water absorption and thickness swelling of the composites resulted from the WF filler absorbed the water. It is clear from the data presented in Fig. 7 that the smaller the WF size resulted in the lower the water absorption and thickness swelling values of WF-RPP composites. The values of thickness swelling and water absorption of WF-RPP composites were varied in the range of 2.35-4.75 and 3.62-3.98%, respectively.

Based on the above results, it can be concluded that the physical and mechanical properties of WF-RPP much affected by WF size. The best WF-RPP composites was achieved using WF size < 120 mesh. The value of tensile strength, breaking elongation, Young's modulus, water absorption and thickness swelling were 16.63 MPa, 2.20%, 3003 MPa, 3.62 and 2.35%, respectively.

Mechanical and physical properties of WF-RPP composites under various MAH contents: The mechanical (i.e., tensile strength, breaking elongation and Young's modulus) and physical (i.e., water absorption and thickness swelling) properties results discussed in Fig. 1-3 and Fig. 7 clearly indicated that introducing WF filler onto PP or RPP matrices resulted in decreases in strength and dimensional stabilization of the PP-WF composites. Therefore, this part of the experiment aimed to improve the mechanical and physical properties of composites by introducing several amount of MAH together with DCP as modifier and initiator, respectively. The composites composed of 50% *Eucalyptus deglupta* Blume species WF, 50% RPP, various amount of MAH (i.e. 0; 1; 2.5; 5; 7.5 and 10%) and 15% of DCP based on MAH weight. The results were shown in Fig. 8-11.

Figures 8-10 exhibited that tensile properties (tensile strength, breaking elongation and Young's modulus) of WF-RPP composites increases with addition MAH together with DCP. What was most important was the fact that a tremendous increase for its tensile strength was noted up to 2.5% addition of MAH modifier on the whole composite. Surprisingly, such a small addition of MAH modifier together with DCP initiator could improved tensile strength, breaking elongation and Young's modulus 2.15, 2.27 and 1.18 times higher, respectively compared with WF-RPP composites free-MAH modifier. Beyond this point, addition of MAH more than 2.5% MAH modifier resulted in a gradual decreases in the tensile strength, breaking elongation

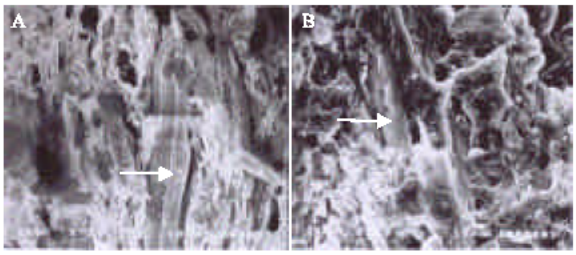

Fig. 12: SEM micrograph of tensile fracture samples of composites of WF-RPP with or without MAH modifier. A) Tensile fracture WF-RPP composite without MAH (750X); B) Tensile fracture WF-RPP composite with 2.5% MAH (750X)

and Young's modulus. From these observations, the reason for the positive role of MAH addition is apparent and attributable to the enhanced formation of MARPP (MAH-modified RPP) and acting as a compatibilizer in the composites. It was reported that addition small amount of MPP (maleated polypropylene) coupling agent onto WF-PP composites (Kishi *et al*., 1989; Han *et al*., 1989; Sombatsompop *et al*., 2005) and MTIR (maleic anhydride modified *trans*-1,4-isoprene rubber) onto TIR-WF composites (Febrianto *et al*., 1999) greatly enhanced the tensile properties of the composites.

Furthermore, the possible reason for its negative role which appeared when excess MAH (more than 2.5% MAH) was used presumably due to 1) the increased occurrence of low molecular weight substances within the composites with excess addition of MAH (MAH remained and its reactants with other additives); 2) the reduction of initiator caused by possible reaction with excess MAH; 3) the degradation of WF and RPP resulting from kneading and molding at high temperature in the presence of excess acidic MAH and so forth (Han *et al*., 1989; Febrianto *et al*., 1999).

It is remarkable that the use of small amount MAH together with DCP dramatically improve the tensile strength of the WF-RPP composites. In addition to this finding, certain physical properties, including water absorption and thickness swelling of WF-RPP composites under various MAH contents were measured (Fig. 11). It is obvious that addition of MAH onto WF-RPP composites significantly improved the water absorption and thickness swelling values of WF-RPP composites. The water absorption and thickness swelling of WR-RPP in the present of MAH was smaller that WF-RPP free-MAH. The lower the water absorption and thickness swelling values of WF-RPP composites with MAH are due to grafting through ester linkage between WF and MRPP, resulted in the composites is more hydrophobic. Similar result has been reported when using MTIR as a compatibilizer on the TIR-WF composites (Febrianto *et al*., 1999).

The scanning electron micrographs of the tensile fracture surfaces of composites samples support these arguments (Fig. 12). Due to hydrogen bond formed on the WF surface and the difference in polar character between WF filler and RPP matrix, WF tends to agglomerate into bundles and become unevenly distributed throughout the matrix (Fig. 12A). Also holes and spacing are commonly formed along the fiber and the fibers pull out, which indicated insufficient adhesion, low compatibility, poor contact and inferior stress transfer between the phases. On the contrary, Fig. 12B showed the microstructure of The WF-RPP composite in the presence of MAH modifier together with DCP initiator. The microstructure is different from that of Fig. 12A. Generally, it is more difficult to differentiate the filler particle from the RPP matrix. This may suggest that the WF filler are coated probably by the matrix and that the failure most commonly occurs in the matrix rather than at the filler surfaces. The filler surfaces are not as clean as in the Fig. 12A and the MARPP (maleic anhydride-modified recycle polypropylene) is believed to be located at the filler surfaces. The MARPP prevents hydrogen bonds among the WF from being formed and renders the properties of WF surfaces and matrix more similar. MARPP facilitates the direct contact between the WF filler and RPP phase and enhances the fiber dispersion in the RPP phase. Good adhesion between WF filler and RPP matrix in the present MARPP was expected because the composites had improved tensile strength, which means that loads can be transferred from the matrix to the fillers.

Based on the above results, it can be concluded that addition of MAH modifier together with DCP initiator directly during mixing the WF-RPP composites can resulted in improvement of physical and mechanical properties of the WF-RPP composites. The best result was achieved by adding 2.5% MAH onto the molten matrix when blending WF-RPP composites. The tensile strength, breaking elongation, Young's modulus, water absorption and thickness swelling values of the WF-RPP composites were 35.74 MPa, 5.00%, 3558 MPa, 2.40 and 1.78%, respectively.

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