1st International Conference on Multidisciplinary Engineering (ICoMdEn)
Advancing Engineering for Human Prosperity and Environment Sustainability
October 23-24, 2018, Lhokseumwe - Aceh, Indonesia.

e-ISSN 2656-7520

MICROCOMPOSITES FROM RECYCLED POLYPROPYLENE PLASTIC WITH REINFORCEMENT FIBER OF OIL PALM EMPTY FRUIT BUNCHES

Zulnazri,[1], Rozanna Dewi[2]*
[1,2]Chemical Engineering Department, Malikussaleh University, Lhokseumawe, Aceh-Indonesia
*Corresponding Author : zulnazri@unimal.ac.id

ABSTRACT

This study describes the processing of thermoplastic polypropylene from used plastic packaging with filler of palm oil bunches as a composite. Polypropylene material is crushed and melted using xylene solvent and then mixed homogeneously with micrometer-sized filler, then presses using a hot press at a temperature of 165^{o}C. Methods of physical and chemical modification of fibers with polypropylene matrix and the addition of several additives were carried out to improve compatibility, mechanical properties, morphology, electrical properties and biodegradability of composites. Analysis of mechanical properties obtained tensile strength of 13.053 MPa, elastic modulus of 455.252 MPa, impact of 3019.1 J/m^2. Infra reed (FT-IR) analysis shows that the -OH functional group of fibers places a position adjacent to the polypropylene functional group. Surface structure analysis (SEM) showed that the fiber bond strength between the matrix and the analysis of thermal properties showed a high melting temperature of 488.33^{o}C.

KEY WORDS: fiber, polypropylene, composite.

INTRODUCTION

Oil palm empty bunches (OPEFB) are abundant waste in the environment. More than 250 tons of OPEFB are produced every day from CPO milling, OPEFB can be used as a filler in composite manufacturing. TKKS contains cellulose until it reaches 44.40%, so it is very potential to add composite tensile strength when used as a filler (Anggraini, et al., 2011 and Aliabadra, et al., 2013).

Thermoplastic composites made from OPEFB-based lignocellulose material are currently being developed. Lignocellulose as a filler has many advantages over inorganic fillers, including: low density, large deformability, flexibility, does not cause heat to the equipment during the process, low prices, and comes from renewable resources. The principle of thermoplastic material and filler of OPEFB fiber is incompatible material, due to differences in polarity, so it is necessary to modify processes such as in situ crosslinking, addition of matching materials (compatibilizer) and functional group copolymerization to polymers and fillers. (Basuki, et al., 2004).

Some chemicals developed for compatibility between the two materials are: maleic anhydrate modified-polypropylene, poly [methylene (polyphenyl isocyanate)], poly (propylene-acrylic acid) and xilane (Rozman, et al., 2002). If lignocellulose is used as a fiber in the composite it will form hydrophilic properties (hydroxyl groups in cellulose), good interfacial adhesion properties, and low resistance to moisture absorption. For this reason the OPEFB-based lignocellulose fibers are processed into composites. (Rozman, et al., 2001).

Polypropylene (PP) is a thermoplastic that can be easily reshaped and processed into other forms, this material is widely used everyday as film bags, plastic bottles and various packaging materials in the lives of modern society, so that thermoplastic waste is very abundant in the environment (Martaningtyas, 2004). Recycled plastic polypropylene can be made as a composite with OPEFB fiber fillers. The composite maximum tensile strength is obtained when the filler content is 20% (weight) even though elongation shows a decreasing trend with an increase in filler. Addition of acrylic acid obtained compatibility between fillers and polypropylene. (Basuki et al., 2004). The reaction of maleic anhydrate (MAH) with OPEFB was observed by FTIR analysis showing the peak properties of MAH in the OPEFB filler. Composite with MAH-treated filler shows high flexural and impact properties. SEM shows adhesion and good compatibility between OPEFB and PP matrix as a result of chemical modification using MAH. (Rozman, et al., 2002).

The use of lignocellulose fibers influences the increase of mechanical properties (tensile strength, modulus of elasticity, hardness) of composite polymers. Processing with agents results in good results between chemical bonds with cellulose fibers in improving mechanical properties. Immersion in water of polymer composites influences the electrical properties of composites. The polymer composite sample treated with cellulose treated gave resistivity after immersion in water. The processing of cellulose fibers with agents increases chemical bonds (especially amino groups) to develop mechanical and chemical properties of composite polymers which immerse in water. (Notingher, et al., 2006). Compared to biodegradable inorganic fillers, lignocellulose filler has low density, great deformability, small abrasive properties, high stiffness, reduced dermal and iritic respiratory properties, good thermal properties, improved energy recovery and low cost. (Khalid, et al., 2007). Process modifications from polymers using reactive process techniques have been reported by several researchers. The final result of the modification of maleic anhydrate reaction with OPEFB microfiller fiber has increased the compatibility of low density polyethylene (LDPE) plastic material with fiber, high impact strength and tensile strength obtained with a mixture ratio of 70; 30 (LDPE: OPEFB) with 63 µm fiber size (Zulnazri, et al., 2017). In this study modification of the mixture of compatibilizer and OPEFB filer can improve the mechanical properties of PP composites.

METHODS

Experimental

Raw material for OPEFB fiber is prepared, milled with grinding mill and sieved mesh up to size 63, 75, 90 and 103 µm. Recycled polypropylene plastic up to 1-2 cm in size.

Prepared a set of blending tools: hot plate, triple neck flask, water bath, condenser, stirrer and thermometer, polypropylene plastic was put in a flask, added 70% 10 ml xylene solvent, heated at 40 °C while stirring, added maleic anhydride polyethylene (MAPE) 8% (0.8 gr in 10 mL xylene). Then in hot conditions mixed with OPEFB filler and mixed until homogeneous. The mixture is poured in aluminum foil and left to room temperature. The mixture is printed (according to testing standards) with a hot press for 20 minutes, at a temperature of 145 °C, a pressure of 10 kg / cm^2. The composite is left to room temperature and ready for testing.

Characterization

Tensile Strength test

Cut composite board parts in accordance with ASTM D638-72 type IV specimens, then attached to the Tensile test tool. After that is withdrawn and the value of Tensile Strength is recorded and calculated by equation (1), then the value of elongation can be calculated by equation (2).

$$\sigma = \frac{P}{A} \qquad (1)$$

$$\Sigma = \frac{\Delta L}{L} \text{ x } 100\% \qquad (2)$$

Impact test

Impact tests are carried out according to ASTM D256. The Izod method is performed using unnotched samples with dimensions of 60 x 15 x 3 mm (P x L x T), using Zwick impact pendulum tester model 5101

Scanning Electron Microscopy (SEM) analysis

The crack surface (fracture) of the composite was studied with SEM (Cambridge Leica S-360 model). Samples mounted on aluminum pieces using double-side tape and then gold-coated with a unit palaron SEM coating to prevent electrostatic loads during testing.

Chemical structure analysis with FT-IR

Analysis of functional groups was carried out using the Nicolet FTIR spectrophotometer (Avatar system 360). All spectra are recorded in transmittance in the range 4000-400 cm^{-1}, with a resolution of 4 cm^{-1}. There are 32 scans for each spectrum.

Analysis of thermal properties with DSC

Analysis of thermal stability is done by Differential Scanning Calorie meter (DSC). Samples were heated from 25 °C to 400 °C at a heating rate of 20 °C min-^{1} below the nitrogen atmosphere with a flow rate of 30 mL min^{-1}. The samples were heated from 25 °C to 500 °C at a heating rate of 25 °C min^{-1} under a nitrogen atmosphere with a flow rate of 30 mL min^{-1}.

DISCUSSION

Tensile Strength

Figure 1 shows the load given to the time of each microcomposite with a matrix: filler ratio (70: 30), fiber size: (1). 63 µm, (2). 75 µm, (3). 90 µm, (4). 106 µm. The graph shows the highest load obtained in the composite (1). 63 µm, the high load shows the higher tensile strength of a composite

Table 1 shows the tensile strength, elongation and elastic modulus values of the composites: 63 µm, 75 µm, 90 µm and 106 µm. The tensile streng value is obtained based on the amount of load that can be given by the composite when it is stretched to the end of the break time.

Table 1 Tensile Strength analysis of PP composites with different filler size

No.	Filler size (µm)	Tensile strength (Mpa)	Elongation at break (%)	Yield strength (Mpa)	Elastis modulus (MPa)
1.	63	13,053	4,548	12,950	455,252
2.	75	12,470	4,918	11,450	301,801
3.	90	11,263	3,074	5,692	600,912
4.	106	12,416	5,142	9,330	265,755

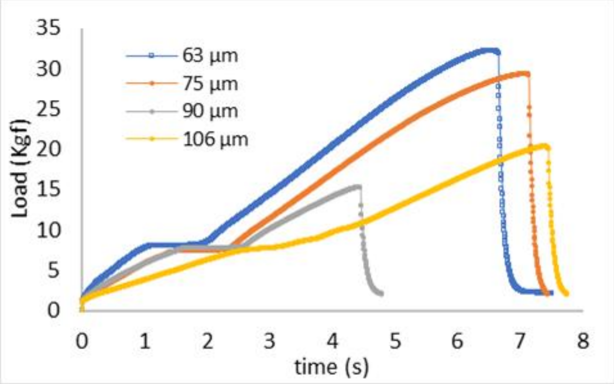

Figure 1 PP composite tensile test graph

The filler content of 30% can produce high composite tensile strength because of the presence of 8% MAPE as an initiator and peroxide as a coupling agent that can help improve composite compatibilizer so that the tensile strength is higher. The size of the fiber as a filer can also influence the composite tensile strength, the smaller the size of the fiber tensile strength increases, this can be seen in composites with 63 µm fiber size. According to khalid cellulose OPEFB which is used as a filler with a polypropylene matrix can increase tensile strength and flexural modulus. Composite mechanical properties can be developed by adding coupling or bonding agents such as maleic anhydride grafted polypropylenr and multi functional acrylates, which strengthen the bond between cellulose and the polymer matrix. (Kalid et, al., 2007).

Impack

Chemical modified composites with OPEFB filler show high impact strength properties, this is due to the adhesion between polypropylene matrix and refined OPEFB fibers in various sizes, stronger with the presence of MAPE, this statement is reinforced by SEM analysis which shows a solid and compact binding surface structure between fiber and matrix. Table 2 shows the high impact obtained on composites which have the smallest fiber size of 63 µm with impact of 3019.1 J/m^2, high impact indicates the higher resistance of a composite to impact.

Table 2 Test the impact of PP composites

No.	Filler size (µm)	W (mm)	T (mm)	Impact (J/m2)
1	63	11	2,18	3019,1
2	75	11	2,07	2863,3
3	90	11	2,11	2563,5
4	106	11	1,83	2886,2

Surface structure

The morphology and dimensions of polypropylene after mixing with OPEFB filler were determined by Scanning electron microscopy (SEM) analysis as shown in Figure 2. Overall the surface structure of the PP composite looks irregular in shape with widths from 1 to 8.5 mm. The scanning electron micrograph shows the morphological changes of the composite because it is influenced by the level of fineness of the fiber, so the size of the fiber fineness as filler needs to be considered to provide a better surface structure, as seen in figure 2 (a) the 63 µm fiber structure provides a better structure and regularly compared to figure 2 (b) the size of the fiber is 90 µm which shows the surface structure tends to be less uniform and the surface pores are more rough. Regular and uniform surface structure shows that there is compatibility between fibers and matrices so that the strength of the composite is also higher, and vice versa. This is proven by a tensile test where the 63 µm filler size shows high tensile strength.

The role of MAPE 8% and xylene has improved the compatibility of the composites which showed the presence of surface pores caused by strong bond marks when tensile tests were carried out. According to Zulnazri, et. Al., 2017, the presence of MAP and xylene can improve the compatibility of composites by increasing the adhesion style of the interface to the matrix and filler. Basuki explained the results of surface structure analysis showed compatibility between OPEFB and Polipropiline with the addition of acrylic acid compatibility. (Basuki et, al., 2004).

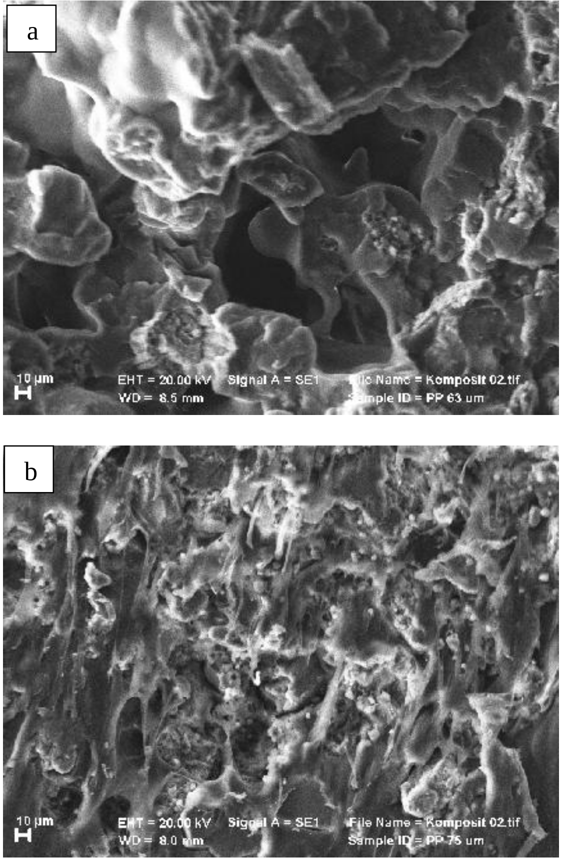

Figure 2. Composite surface morphology of fiber size (a) 63 µm, (b) 75 µm.

Chemical structure

Characteristic of the spectrum of microcomposite is shown in Figure 3. All samples display two main absorption regions, namely in the highest wave number area 2800-3500 cm^{-1} and the lowest wave number 500-1700 cm^{-1}, as shown by Haafiz et, al. 2013. The spectrum image shows that the broad absorption peak located at 3000-3500 cm^{-1} is a stretch of the -OH group which is thought to originate from the cellulose-TKKS functional group and the absorption peak in the area 2800-2950 cm^{-1} is related to the -CH$_2$ group while at peak 2846 and 2914 cm^{-1} uptake are overlapping of the -CH$_2$ band derived from lignocellulose filler with OPEFB and polypropylene. The area absorption band 1470-1585 cm^{-1} is related to hydrogen between molecules of the aromatic ring group. The absorption peaks that appeared in the area of 1018 cm^{-1} and 1049 cm^{-1} wave numbers were strongly suspected by the C-O-C stretch of alkyl aryl contained in lignin. The appearance of these two peaks in the spectrum has shown a compatible interaction between the PP matrix and OPEFB, which is influenced by the presence of MAPE as a compilizer and xylene as the initiation of coupling formation.

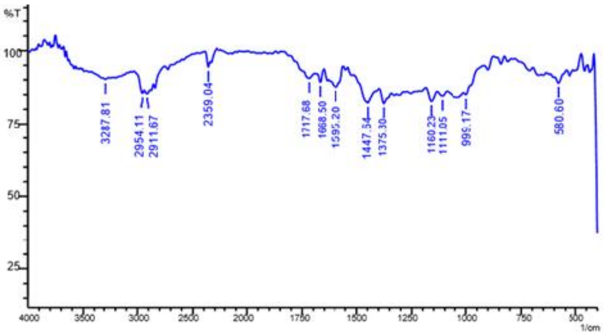

Figure 3. Spectrum FT-IR composite, 63 µm fiber size

Thermal analysis with DSC

Figure 4 shows the DSC thermogram of PP microcomposite with 63 µm filler size. Melting temperature describes the phase change from solid to liquid without experiencing a change in composition, where the polymer loses its overall crystallinity. After passing the melting temperature, the composite will experience decomposition which is indicated by changes in the composition and chemical properties of the composite. PP has a melting point of 165ºC, while the PP-OPEFB composite after heating is decomposed at a temperature of 488.33ºC, this shows the decomposition temperature of the composite is very high. High temperature degradation indicates that composites have good thermal stability and are resistant to heat.

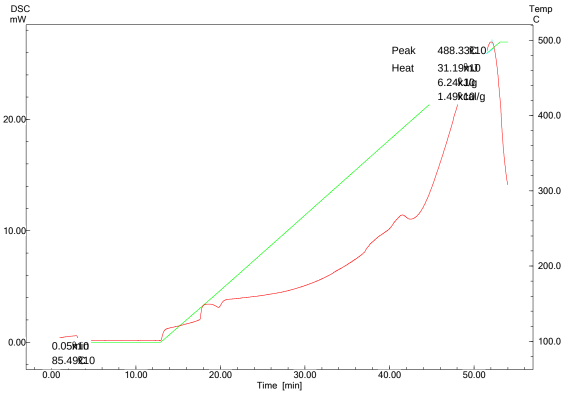

Figure 4. DSC composite thermogram with 63 µm fiber size

CONCLUSIONS

Polypropylene microcomposite with OPEFB filler with a ratio (%) 70:30 has good compatibility with strong cross ties between PP and OPEFB filler after addition with 8% MAP. The MAPE reaction with OPEFB was analyzed by FT-IR showing the presence of MAPE peaks overlapping with OPEFB fibers. PP-TKKS composite with 63 µm filler size shows high tensile strength and impact. SEM shows adhesion and good compatibility between TKKS and PP matrices.

REFERENCES

Anggraini, D. and Roliadi, H. (2011). Manufacture of pulp from oil palm empty fruit bunches to cartons at small scale businesses, Forest Products Research, 29 (3), 211-225.

Aliabadra P., Saputra, H.A. and Nasikin, M. (2013). *World Appl Sci J.***,** 22 (5), 747-754

Basuki, W., Guritno, P., Ismail, H. (2004). Oil Palm Empty Fruit Bunch Filled Popypropylene Composites, Int. J. Polymeric Materials, 53: 295-306.

Rozman, H.D, Tay, G. S., Kumar, R. N., AbuSamah, A., Ismail, H. and Ishak, Z. A., (2002). Flexural and impact properties of oil palm empty fruit bunch (EFB)-polypropylene composites-the effect of maleic anhydride chemical modification of EFB, J. Polymer Testing, 22: 335-341.

Rozman, H.D, et al. (2001). Polypropylene-oil palm empty fruit bunch-glass fibre hybrid composites: a preliminary study on the flexural and tensile properties, European polymer journal, 37: 1283-1291.

Martaningtyas. (2004). *Potential Plastic* "*Biodegradable*". Institut Pertanian Bogor, Bogor.

Khalid, M, et.al., (2007). Comparative study of polypropylene composits reinforced with oil palm empty fruit bunch fiber and oil palm derived cellulose, J. Material design, 29: 173-178.

Notingher, P.V, (2006). The effect of water on electrical properties of polymer composites with cellulose fibers, journal of optoelectronics and advance materials, 8: 687-689.

Zulnazri and Sulhatun, (2017). Effect of Palm Oil Bunches Microfiller on LDPE-recycled Composite Tensile Strength through Melt Blending Process, Emerald Reach Proceedings Series, Vol. 1, pp. 547–553.

ASTM (2000). American Society for Testing and Materials Information Handling Services. *Standard Test Method for Tensile Properties of Plastics*. (D 638-99): 1-13.

Haafiz, M. M. K., Eichhorn, S. J., Hassan, A., Jawaid, M. (2013). Isolation and Characterization of Microcrystalline Cellulose from Oil Palm Biomass Residue, *Carbohydrate Polymers,* 93, 628– 634.