The Effect of Fibre Composition on the Strenght of Microcomposite from Recycled Polypropilene Plastics

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ABSTRACT--_This research is related to the making of composites from polypropilene (PP) plastic waste material using a micro palm empty fruit bunch (EFB) filler, the fillers used are 63 and 90 micrometers in size with PP : fiber ratio (70:30 and 60:40). The method used by the process of screw extruder, plastic material and filler is dissolved with the compatibilizer then mixed in the extruder by giving heat / temperature around 160 - 170 ^0C. The product of this research is in the form of granules or composite pellets which are then printed with a hot press to be formed as specimens or modeling according to industry requirements. In this study the target application is as a wood substitution material for interior needs, furniture, electronic casing and electronic board / board elements. The products of this research are composite granules or pellets that are ready to be printed or modeled as needed. The target of this research application is as a wood substitution material for interior needs, furniture, electronic casing and electronic element boards._

**Keywords** --Composite, EFB, Filler, HDPE, OP, Plastic and PP

I. INTRODUCTION

As per the estimates of the Indonesian Plastic and Olefin Industry (INAPlas), the plastic demand of the Indonesian people in 2002 was approximately 1.9 million tons and then increased to 2.1 million tons in 2003. While the domestic plastic demand in 2004 was estimated to reach 2.3 million tons. This means that tens of tons of plastic have been produced and used by the community. Plastic has become a necessity of life that continues to increase in number

Efforts to reduce environmental pollution can be done by recycling plastics by depolymerizing certain types of plastic into other products, for example polyethylene terephthalate (PET) types into terephthalate acid and ethylene glycol to make fibers, molding materials, and plastic cans. PET, High-density polyethylene (HDPE), and Polyethylene (PP) type, which is usually used for packaging milk into plastic bottles, toys, pipes, and other products (Martaningtyas, 2004).

Thermoplastic composites made from lignocellulosic filler materials such as wood fiber and oil palm bunches are currently being developed. Lignocellulose as a filler has many advantages over inorganic fillers, among others, low density, large deformability, flexibility, does not cause heat to the equipment during the process, low prices, and comes from renewable resources. Thermoplastic and filler materials from plant fibers are

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principally incompatible materials, due to differences in polarity, so it is necessary to modify processes such as in situ cross-linking, addition of compatibilizers and copolymerization of functional groups in 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 xylane (Rozman, et, al., 2002).

The maleic anhydrate (MAH) reaction with OPEFB has been observed with FTIR analysis showing the nature of the MAH peak in the OPEFB filler. Composites with MAH-treated fillers exhibit high flexural and impact properties. SEM shows good adhesion and compatibility between OPEFB and PP matrix as a result of chemical modification using MAH (Rozman, et al., 2002).

Potentially treated cellulose is used as a thermoplastic filler compared to untreated OPEFB against PP. The tensile and flexural properties of cellulose treated are higher than untreated EFB. The mechanical properties of these composites are improved by using maleic anhydride grafted polypropylene (MAPP) couplings and bonding agents (MFA) to increase bonding between cellulose and polymer matrix. (Khalid, et, al., 2007).

In this research, a composite was made with PP and HDPE as a matrix, and OPEFB waste as a filler. The OPEFB waste comes from the palm oil processing industry (CPO). OPEFB solid waste has the potential to be used as filler because of its organic fiber content (cellulose, hemicellulose, and lignin). The size of the filler greatly determines the results of the composite, the smaller the particle size of the filler, the greater the interface area so that the interaction is stronger and the better the composite properties. Refinement of micrometer-sized OPEFB fibers will improve mechanical properties, thermal properties, surface morphology, and degradation of composites is difficult.

Polypropylene is a thermoplastic plastic, which can be easily reshaped and processed into other forms. In general there are four requirements so that a plastic waste can be processed by an industry, including waste must be in a homogeneous form as needed (seeds, pellets, powder, fractions), the waste must be homogeneous, not contaminated, and endeavored not to be oxidized. To overcome this problem, before it is used plastic waste is processed through simple steps, namely separation, cutting, washing and removal of substances such as iron and so on.

After scaling down, the PP is then heated to its melting point, then processed to pellet shape. Before being used as a composite matrix, a differential thermal analysis (DTA) was performed. In the two-stage process, the pellet is blended in advance with a coupling agent that functions as a compatibilizer in making composites.

Polypropylene has a melting point of $\sim 160\,°C$ ($320\,°F$), as determined by Differential Scanning Calorimetry (DSC). The MFR (Melt Flow Rate) and MFI (Melt Flow Index) are an indication of the molecular weight of PP and determine how easily the melted raw material will flow during processing. Higher MFR PP will fill plastic molds more easily during the injection and blow molding production process. But when the melt flow increases, some physical properties, such as the impact strength, will decrease.
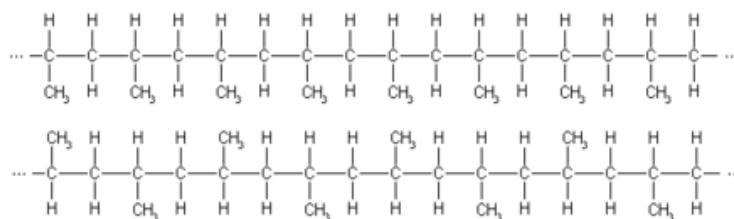
Figure 1: 'Structure of PolyPropilene'

To produce elastic polypropylene, catalysts that produce isotactic polypropylene can be made, but with an organic group that affects tacticality held in place by a relatively weak bond. After the catalyst produces a short polymer that is able to crystallize, light with the right frequency is used to break this weak bond, and eliminate the catalyst selectivity so that the remaining chain length is atactic. The result is a material which is generally amorphous with small crystals embedded in it. Because one end of each chain is in a crystal while most of its length is amorphous and soft, the crystalline region has the same usefulness as vulcanization.

II.   METHODS OF RESEARCH

Palm Oil Empty Fruit Bunch Fiber (EFB) is cleaned of dirt that sticks. OPEFB cut into pieces with a knife and scissors to measure ± 1-5 cm. Crude fiber is soaked with plain water for 24 hours. Fiber dried in the sun to dry. Boil with 5% NaOH solution for 30 minutes for the delignification process. Washed with water and dried. Smoothed again by grinding mill and sieved to micro size (size varied), the filler is weighed according to the composition ratio and ready to use.

A set of extruder, hot plate, three-neck flask, water bath, condenser, stirrer and thermometer were prepared. A 70 gram plastic matrix was mixed with a 0.25% BPO initiator as much as 20 mL stirred until evenly mixed. The 30 gr TKKS filler was mixed with a 20% MAH 2.5% compatibilizer, stirring until blended.

The two ingredients are mixed so that the amount becomes 100 g or 100%, after homogeneous mixing the concentration of BPO becomes 0.05% and MAH to 0.5%.

The mixture is extruded in a single screw extruder. Optimized variables in this process are: temperature, residence time adjusted to the properties of the material, the ratio of the matrix to the filler. After the reaction process is complete the product (polymer mixture) is in the form of pellets. The results are included in specimen molds that are sized according to ASTM standards

Then the mold was put in compression molded at 145-180℃ for 3 minutes without pressure, and the addition of 10 minutes at a pressure of 100 kN, to obtain composite specimens, for microscopic, mechanical and homogeneity testing.

2.1 Pull Test with Tensile Strength

1.   Parts of composite specimens that have been printed according to ASTM D-638-72 type IV, are attached to the Tensile Strength tool.

2.   After being drawn then the pull strength number comes out on the monitor of the tool, enter it in the formula (equation 1) to get the tensile strength value and (equation 2) to get the estrangement value.

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

When :

σ = Pull of Streght (Kgf/cm^2)

A= Surface (cm^2)

P = Maks Pressure (Kgf)

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

When :

$\sum$ = estrangement (%)

ΔL = different height after pull test (cm)

L   = initial height (cm)

Cracked (broken) surfaces of composites were studied by Scanning Electron Microscopy (SEM) (Leica Cambridge S-360 model). The sample is mounted on a piece of aluminum using double-side tape and then gold-coated with a SEM coating of palaron unit to prevent electrical loads during testing.

FTIR analysis occurs using Nicolet FTIR spectrophotometer (Avatar system 360) and KBr method. All spectra are recorded in transmittance in the range 4000-400 cm^{-1}, with a resolution of 4cm^{-1}. There are 32 scans for each spectrum.

Analysis of thermal properties with DSC

1. Curved the corner of the sample approximately 2 grams.
2. After that it is put into printed form.
3. Then monitored will be obtained graphical form to know the melting point of the composite board

III.   RESULT AND DISCUSSION

The stages in this research include the processing of used plastics (PP and HDPE) with the crusser method, the processing of OPEFB fiber as a filler by the grinding mill method, and the process of mixing the two materials with the Screw extruder method. In the OPEFB processing obtained in the form of filler products with sizes 63 and 90 micrometers. The process of making composites by the extruder method is done by mixing a plastic matrix with a filler, where previously the 70 gram PP matrix was moistened with 0.25% benzoyl peroxide as much as 20 ml and stirred until it was evenly distributed to the entire matrix surface, then the 30 gr filler was mixed in anhydrous maleate 2.5% as much as 20 ml until homogeneous, then the matrix and filler are mixed and stirred until homogeneous, then the mixture is put in a screw extruder and operated for 20 minutes at 160℃. The result of this extruder is actually also called a composite in the form of hard and rigid pellet beads. Then the composite in the form of pellets is printed as a specimen by the hot press method. The results are in the form of thin plate specimens used for testing and characterization.

From the blending test results of a mixture of PP and filler matrices that have been carried out in the first year, imperfect composites are produced (the composite is not yet homogeneous). So that in the second year the blending process was changed using the srew extruder method. On the production scale the process of forming composites from matrices (HDPE and PP) with fillers is carried out in a single screw extruder, as an initiator plus

benzoyl peroxide and MAH as a binding agent to make the composites more compatible. The optimized variables in this process are temperature, residence time, initiator concentration, content of the compatibility agent and filler content. After the reaction process is complete, a pellet-shaped composite product (polymer mixture) is then put into compression molded at 180℃ for 3 minutes without pressure, and the addition of 3 minutes with a pressure of 100 kN to form a polymer film, for microscopic, mechanical and homogeneity testing.

In this extruder process, a very homogeneous composite mixture of PP and filler is obtained in the form of hard and rigid pellet beads. This result is very good when seen physically because it is not easily destroyed and not easily cracked. Then the composite in the form of pellets is printed as a specimen by the hot press method. The results of this mold are ASTM D-638 type IV specimens as shown in Figures 2 and 3, then these specimens are tested for mechanical and chemical properties.

Composites visually show clearer appearance and better homogeneity. On the other hand, the physical texture of the composite looks smoother with the finer size of the filler. Pereira [14] has reported that the ability to "wet" the dispersant is an important factor affecting the distribution of fillers in the polymer matrix (Basuki, 2004).

3.1 Tensile Test Analysis

Table 1 shows the results of the tensile strength test / tensile test of HDPE and PP composites in various treatments.

Table 1: Results of PP composite tensile strength test

Name	Max_Force	Max_Stress	Break_Force	Break_Disp.
Parameters	Calc. at Entire Area	Calc. at Entire Area	Sensitivity: 10	Sensitivity: 10
Unit	N	MPa	N	mm
PP 60:40, size 63 μm	288.858	15.4057	285.255	1.03565
PP 60:40, size 90 μm	313.247	14.0281	155.086	1.4165
PP 70:30, size 63 μm	301.935	16.1032	234.801	1.69315
PP 70:30, size 90 μm	336.349	18.5623	322.794	1.2365

Tabel 1 shows the maximum tensile strength results of PP composites at a ratio of 70:30 with 90 μm filler size with max sterss of 18.5623 MPa and the composite will break with elongation of 1,2365 mm.
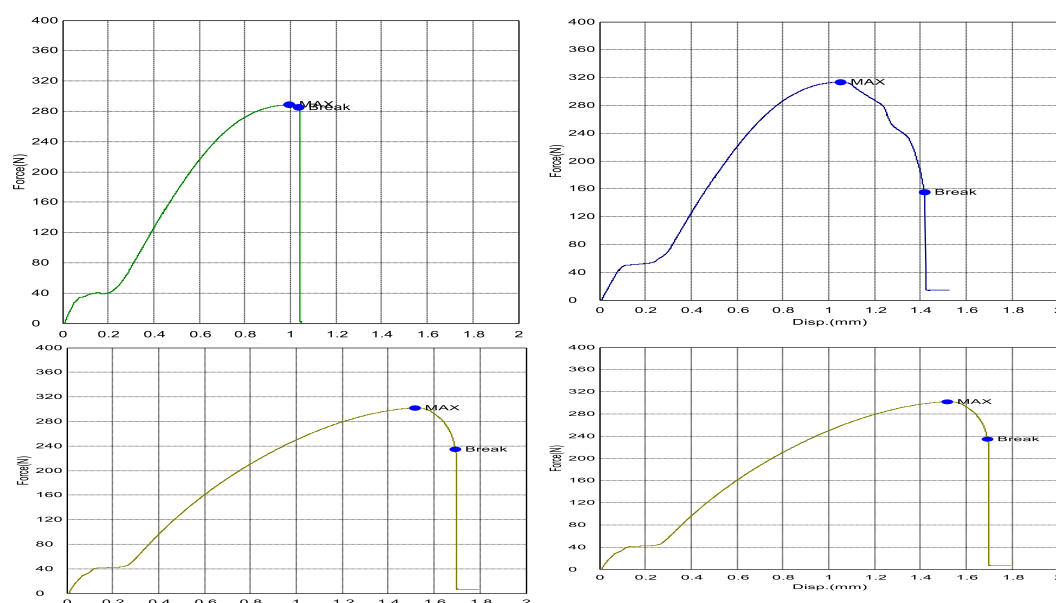
**Figure 2:** 'Composite tensile test graph, (a) PP: filler (60: 40) 63 μm, (b) PP: filler (60: 40) 90 μm, (c) PP: filler (70: 30) 63 μm, (d) PP: filler (70: 30) 90 μm'

Figure 2 (a) shows the graph of PP composite tensile test results with a matrix / filler ratio of 60: 40 and filler size of 63 μm. Table 1 and Figure 2 (a) explain that the maximum tensile strength of the treatment is 15.41 MPa and the composite will break at 285.26 N. Figure 2 (b) shows the graph of the PP composite tensile test results with a matrix / filler ratio of 60 : 40 and the size of the filler is 90 μm. Table 1 and Figure 2 (b) explain that the maximum tensile strength of the treatment is 14.03 MPa and the composite will break at 155.09 N. Force Figure 2 (c) shows a graph of the results of the PP composite tensile test with a matrix / filler ratio of 70 : 30 and filler size 63 μm. Table 1 and Figure 2 (c) explain that the maximum tensile strength of the treatment is 16.10 MPa and the composite will break at a force of 234.80 N. Figure 2 (d) shows a graph of the PP composite tensile test results with a matrix / filler ratio of 70 : 30 and filler size 90 μm. Table 1 and Figure 2 (d) explain that the maximum tensile strength in the treatment is 18.56 MPa and the composite will break at 322.79 N.

Based on the above analysis it can be seen that the maximum tensile strength possessed by HDPE composites with a 60: 40 matrix: filler ratio treatment with a 90 μm filler size of 21.11 MPa and the composite will break at 391.56 N. The PP composite analysis shows that the maximum tensile strength is owned by PP composites with a 70: 30 matrix: filler ratio treatment with a 90 μm filler size of 18.56 MPa and the composite will break at 322.79 N. This is clearly seen in Figure 2. It turns out that HDPE composites are more flexible than PP composites in which if the composite is given a load the composite will experience flexibility before breaking. This shows that the bond strength between various components in the composite has a significant influence on its properties. (Kalpakjian 1984). The bond between the HDPE matrix is stronger when compared to the PP matrix bond with the OPEFB filler.

3.2 Study of Scanning Electron Microscopy (SEM)

The morphology and dimensions of PP composites, determined by SEM analysis. Overall the surface structure of the composite looks regular in shape with widths of 10, 11, 12, and 14 mm. Micrographs show changes in the morphology of composites because they are influenced by the size and degree of fiber fineness. So that the size of the fineness of the fiber as a filler needs to be considered to provide a better surface structure, as shown in figures 3 (a) and 3 (b) 63 μm fiber size gives a better flat structure and smoother surface pores compared to Figure 3 (c) and 3 (d) fiber size of 90 μm which shows the surface structure tends to be less uniform but looks more rigid and strong, this is consistent with the results of tensile tests which show stronger than the fiber size of 63 μm. Although the surface pores of composites (c) and (d) are coarser, the bond between the fiber and the PP matrix looks more compatible and shows good crosslinking and more dipole-dipole bonds that occur stronger. The fibers shown in fig.2 below show the typical shape of the morphology of lignocellulose.
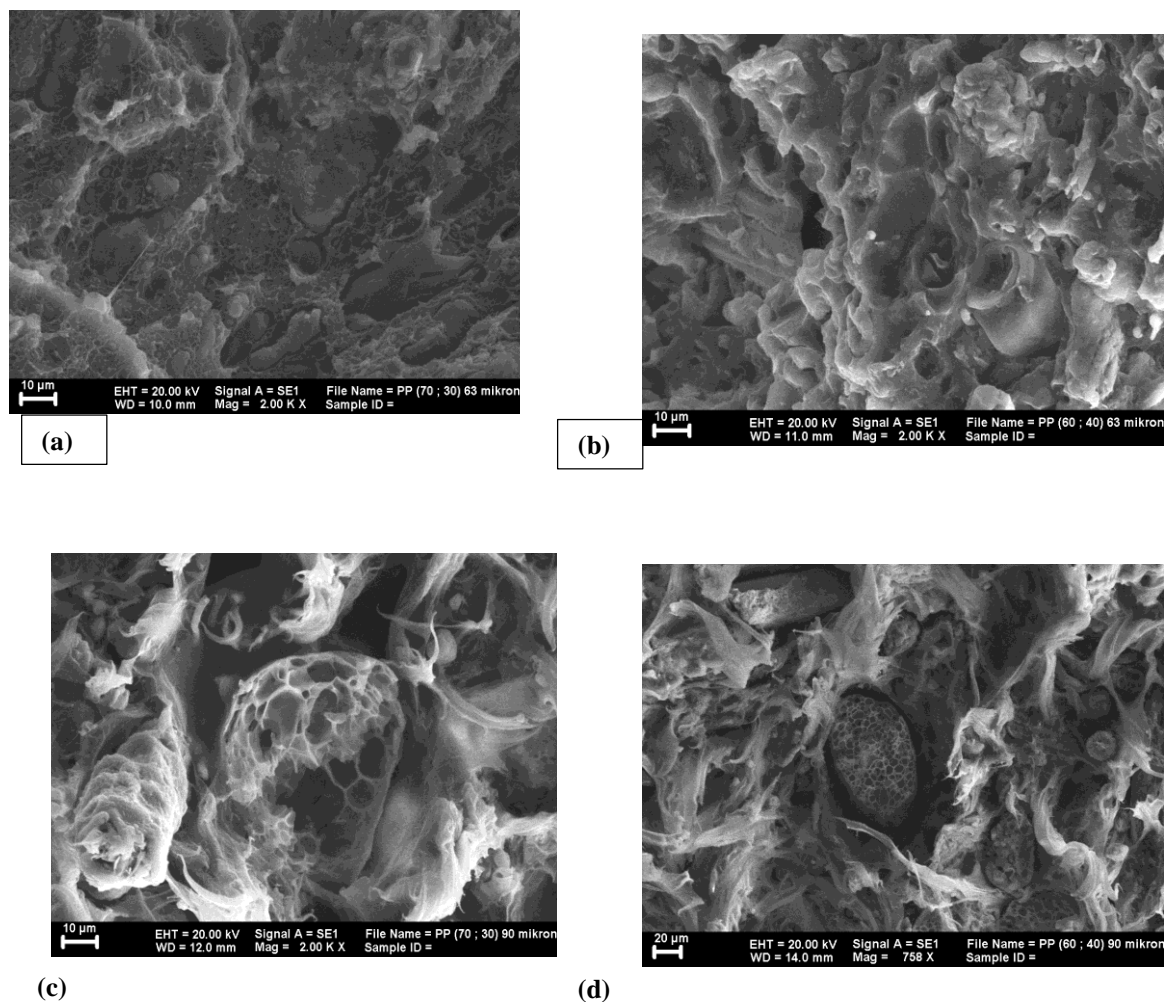

Figure 3: 'Morphology analysys with SEM composite PP:fiber, (a). (70:30), (b). (60:40) filer size 63 μm and (c). (70:30), (d). (60:40) filer size 90 μm'

3.3 Analysis of Fourier Transform Infrared (FTIR) Test

Functional group analysis is performed by FT-IR spectroscopy, the typical PP-EFB spectrum is shown in Figure 4 and the vibration area placement is summarized in Table 2 below. Composites display two main uptake regions as shown by Muhammad Haafiz et al. (2013), namely in the area of the highest wave number (2800-3500 cm^{-1}) and the lowest wave number (500-1700 cm^{-1}). From Figure 5.8 below shows the similarity between the two spectra which is an indication that both samples have the same chemical composition.

From the spectrum it is seen that a broad absorption peak located at 3000-3500 cm^{-1} is stretching the -OH group and the absorption peak at the area of 2800-2950 cm^{-1} is related to the $-CH_2$ group while at the absorption peak 2846 and 2914 cm-1 is overlapping from the band $-CH_2$ derived from lignocellulosic fillers and PP. The absorption band in the region of 1470-1585 cm^{-1} corresponds to intermolecular hydrogen in groups C_6 (aromatic ring) (Kumar, Maria De La, & Yang, 2002). Absorption peaks that appear in the wave number region 1017 cm^{-1} and 1052 cm^{-1} in PP composites are presumably caused by stretching C-O-C of the alkyl aryl contained in lignin. The appearance of these two peaks in both spectrums has shown the existence of compatible interactions between the matrix and the OPEFB.
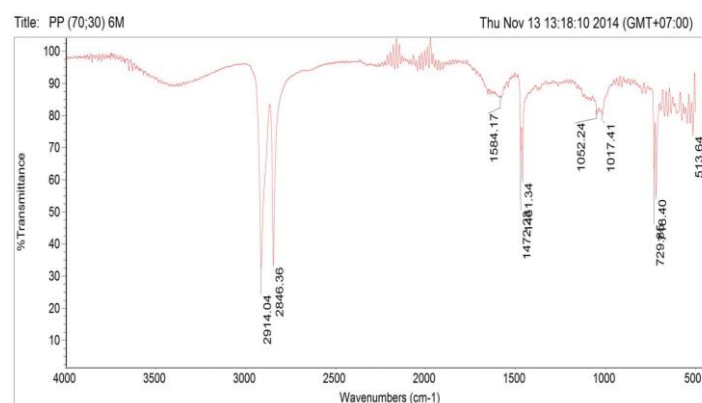
Figure 4: 'Specktrum of FT-IR composite from PP-EFB rasio 70:30 with fiber size 63 μm'

Table 2: The Peak absorbtion of FT-IR spectrum from HDPE-EFB and PP-EFB

Peak Frequency (cm^{-1})	Placed Peak
3000–3500	OH- bending (branching)
2800–2950	CH_2- groups
1470-1585	CH_2- bending (aromatics rings)
1017-1052	C-O-C stretching
729-750	C-H

3.4 Thermal Test Analysis with Differential Scanning Calorymeter (DSC)

DSC (Differential Scanning Calorymeter) testing is done to see the melting point of the composite against heat and to see the amount of heat needed to reach the melting point. The melting point represents the phase change from solid to liquid without undergoing a change in melting composition and temperature, which is a critical

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temperature at which the polymer loses its overall crystallinity. Composite material has decreased its melting point along with the addition of filler, this occurs because the polymer chain is physically degraded, meaning that the polymer chain is forced by the filler so that it damages the polymer molecular chain bonds.

The melting point and decomposition temperature of PP composites is not a single price but lies in a certain range. Figure 5 shows the first peak, giving information about the melting point of the material, while the second peak gives information about the decomposition temperature and curve character in general. The melting point is influenced by the molecular symmetry shape and molecular weight of the polymer compound and the degree of crystallinity of the composite material, the higher the degree of crystallinity of the composite material, the higher the melting point of the composite material, and vice versa.

From Figure 5, it is seen that the PP-filler composite 63 μm has a lower melting point than that of the 90 μm PP-filler composite with values of 133.57℃ and 163.81℃, respectively. This is due to the fact that PP with 63μm TKKS filler has a smaller size so that it is easy to cross-link between the filler and PP polymer, besides the addition of filler is done by heating which can cause some polymer molecular chains to become irregular and tired so it tends to be more easily degraded. When the filler is mixed with the matrix, the damage that occurs in the PP-filler molecule chain 90 μm is not as much as in the PP-filler molecule 63 μm, so that the arrangement of the polymer chains that are still regular are also more numerous. So that the temperature used to melt PP-filler composite material 90μm higher than the temperature to melt PP-filler composite material 63μm.

The decrease in decomposition temperature from the graph shown in Figure 5 (a) and (b) is not so significant, the condition is suspected to be the addition of OPEFB filler to maintain the polymer decomposition temperature. The decomposition temperature of the two PP composite materials did not show a significant difference, this was due to the addition of 63μm and 90μm EFB fillers which had the same effect in maintaining the composite decomposition temperature.
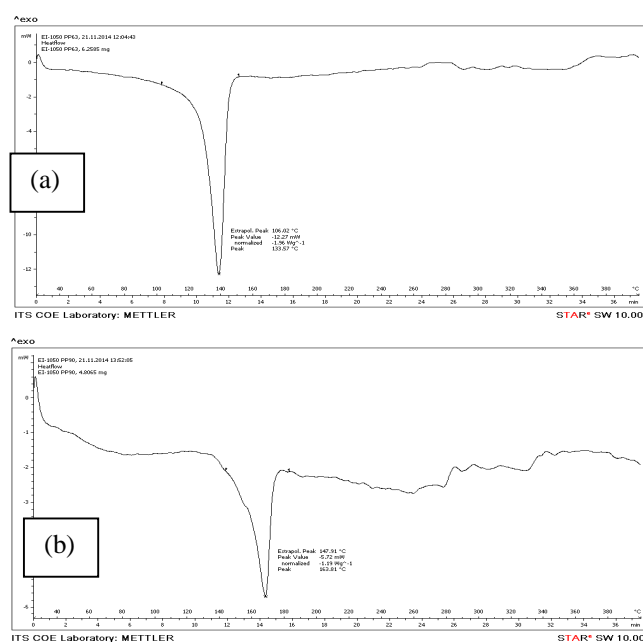
Figure 5: 'DSC thermogram PP-EFB ratio of 70:30 (a) filler size 63 μm, (b) filler size 90 μm'

IV. CONCLUSION

1. PP composite has good characteristics when viewed from tensile strength and surface structure.

2. The maximum force required to break a PP composite reaches 18.56 Mpa

3. SEM shows the composite surface structure at a ratio of 70, 30 and 60; 40 with a fiber size of 90 μm, which shows more compatibility than the fiber composite size of 63 μm.

4. Analysis of functional groups shows that PP composites have O-H and C-O-C functional groups, each of which is derived from cellulose and ligin OPEFB material, so that this composite is already very compatible between matrix and filler.

5. The thermal stability of PP composites is quite high as indicated by the melting point reaching 163.81℃.

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