

HDPE Recycled Plastic Composite with Cellulose Fiber Filler Based on Oil Palm Empty Fruit Bunches

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Abstract

Composite is an advanced material product that has better mechanical properties than a single material and has high tensile strength; it can be applied to the construction of boat walls, car dashboards, electronic casings, and others. This study examines the composite quality of HDPE recycled plastic using cellulose fiber-based on Empty Palm Oil Bunches (TKKS), which is processed through an extruder and injection melt blending system. The method used is mixing plastic and fiber materials with a roll mill and blending with an extruder. Cellulose fiber was dissolved with compatibilizer maleic anhydrous (MAH) 2.5% as much as 20 mL, stirred until smooth, and HDPE plastic was dissolved with benzoyl peroxide (BPO) 0.5% 10 mL. The composite was molded with hot pores at 160-170 °C. The tensile test shows the strength of HDPE composite (90: 10) is 27.0 Mpa with maximum load reaching 274 N, HDPE composite (80: 20) is 26.0 MPa with full load 258 N, HDPE composite (70: 30) is 320.0 MPa with a maximum load of 316 N. SEM shows the surface structure of the fiber-compatible HDPE composite. Functional group analysis showed that HDPE composites had O-H and C-O-C functional groups derived from lignocellulosic OPEFB material. The appearance of these two peaks indicates a compatible interaction between the HDPE matrix and cellulose. DSC shows that HDPE composites have a degradation temperature of up to 260 °C and a decomposition temperature of 493.45 °C.

Keywords: Plastic, Filler, EFB, composite, HDPE.

1. Introduction

High-Density Polyethylene (HDPE) plastic waste is polymer waste that is not easily decomposed by decomposing microorganisms, cannot absorb water, and cannot rust. The accumulation of plastic waste in the environment continues to increase along with the increasing human needs. This plastic waste can be recycled or reshaped by heating. HDPE is a type commonly found in everyday life, including plastic bottles, plastic buckets, plastic bags, motor vehicle spare parts, electronic materials, household appliances, and others that are disposable [1]. HDPE plastic can be made as a composite with palm empty fruit bunch (OPEFB) fiber reinforcement for motorized vehicle applications [2] [3].

Oil Palm Empty Bunches (TKKS) is the pulp of fresh oil palm fruit with solid fiber and is not easily decomposed. If used as a filler in composites, this fiber lasts a long time because it is wrapped in plastic so that spoilage microorganisms cannot enter, and it has also been moistened with a chemical (wetting agent) so that the fiber becomes more resistant [2] [4].

Thermoplastic composites made from lignocellulosic materials such as wood and cellulose are being developed. Lignocellulose is a filler that has many advantages over inorganic fillers: low density, large deformability, flexibility, does not cause heat to the equipment during the process, low price, and comes from renewable resources. Thermoplastic materials and fillers from plant fibers are principally incompatible due to differences in polarity, so process modifications are necessary, such as in situ crosslinkinthe g, the addition of compatibilizers, and copolymerization functional groups on polymers and fillers [5]. Some of the chemicals developed for the compatibility between the two materials are maleic anhydrate modified-polypropylene, poly [methylene (polyphenyl isocyanate)], poly (propylene acrylic acid), and silane [6].

Composite fabrication of used plastics such as PP and HDPE with OPEFB fiber fillers can be done by the screw extruder method. This method is principally mixing the plastic matrix with the stuffing. The matrix is wetted with 0.25% benzoyl peroxide then the filler is

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mixed in 2.5% anhydrous maleates until homogeneous. Then, the mixture is processed in the extruder. The result obtained is a composite in the form of a hard pallet with a high strength [7] [3].

LDPE matrix composites with a ratio of 70:30 and fiber size of $63 \mu m$ provide a high tensile strength of 12,496 MPa and an elastic modulus of 5,019 MPa [8]. However, this mechanical strength is still low and has not been able to equalize the mechanical strength of modified composites. Therefore, a combination of materials and a good mix of coupling agents or compatibilizers is needed. If the filler ratio is added to the matrix, the matrix cannot flatten the entire filler surface so that it can reduce the tensile strength value [8].

Modification of reactive process techniques to improve the compatibility of the PP matrix with lignocellulose-OPEFB fillers. The resulting flexural, thermal, and tensile strength properties have not shown uniformity between several mixing ratios [9] [3].

Treated cellulose has the potential to be used as a thermoplastic filler compared to OPEFB treated with PP. The tensile and flexural properties of treated cellulose were higher than that of untreated TKS. The mechanical properties of this composite were improved by using coupling and bonding agents maleic anhydride grafted polypropylene (MAPP) and multi-functional acrylates (MFA) to increase the bond between cellulose and polymer matrix [10].

Processing of cellulose fibers increases the mechanical properties (tensile strength, modulus of elasticity, hardness) of the composite polymer. Treatment with these agents resulted in good chemical bonding with cellulose fibers in improving mechanical properties [11].

Several researchers have reported process modification of polymers using reactive process techniques. The end resof the modificmodichangemprove the compatibility of polymeric materials using lignocellulosic fillers. Impact strength, dynamic fracture, the effect of water on electrical properties, flexural al, and tensile properties of polypropylene polymers with OPEFB fiber reinforcement have been reported by several researchers. In this study, HDPE composites with cellulose fibers will be tested for tensile strength to see the tensile strength of the composite.

2. Literature Review

A Composite is a material with two or more different components to produce specific properties and characteristics, which cannot be achieved by one piece without combining with other materials. There are many composite materials for various applications, including OPEFB fiber-reinforced composites with a micrometer size [2]. These fibers differ from other particles because they impart anisotropic properties to the product when aligned. Such reinforcement also has an important influence on the fracture strength of the composite. The bond strength between the various components in a composite significantly influences its properties [12].

OPEFB has the characteristics: hydrophilic (hydroxyl groups in cellulose, lignocellulose, and hemicellulose), good interfacial adhesion properties, and low resistance to moisture absorption when lignocellulose is used in composites. For this reason, lignocellulosic fibers are treated with suitable chemicals [6]. Compared with inorganic biodegradable fillers, lignocellulosic fillers have low density, large deformability, low abrasive properties, high stiffness properties, reduced dermal and respiratory irritation, good thermal properties, increase energy recovery, and are inexpensive [10]. The chemical composition of OPEFB can be seen in Table 2.1 below:

No.	Parameters (unit)	Value
1	Average fiber length (mm)	1.20
2	Outer fiber diameter (mm)	15.01
3	Fiber content (%)	72.67
4	Nonfibre content (%)	27.33
5	Mass density (Kg/m ³)	177.98

Table 1. Chemical Composition of OPEFB: Wirjosentono, B, 2004

HDPE matrix processing using OPEFB fiber on a laboratory scale with the solution technique method, the tensile strength of the composite increases with the addition of OPEFB fiber above 20% (weight) with the addition of 3% (weight) acrylic acid and 0.01 mol ratio di cumyl peroxide. At the same time, the elongation properties show a downward trend. This observation also occurs when using a single screw extruder [10].

HDPE composite with chemical modification of OPEFB filler with MAH showed high flexural properties and impact strength compared to OPEFB that was not treated with MAH. This is due to the adhesion between OPEFB treated with MAH and HDPE matrix, as demonstrated by the SEM study. Infra-red analysis (FTIR) clearly showed the C=C and C=O group bonds of MAH at 1630cm-1 and 1730cm-1 [13].

Composite quality can be improved by adding a coupling agent, increasing the interface adhesion between the matrix and the fiber. The composite is more substantial; the coupling agent will form cross-links between one type of material and another. Maleic anhydrous is one of the coupling agents that will form cross-links between polymers and other polymers; this cross-link can be created with the help of peroxide compounds as an initiator, which helps open the C-H bonds in PP to form +R-OH radicals [14].

3. Methods

HDPE type used plastic is sorted, washed and dried, crushed to a size of 1-2 cm. OPEFB fibers were extracted with 5% NaOH solution for 30 minutes for the delignification process. Cellulose fibers are ground again with a grinding mill to a uniform size and weighed according to the composition ratio [15].

HDPE plastic raw material-cellulose fiber is mixed with a 70: 30 g, moistened with 70% xylene solvent as much as 10 ml, stirred until evenly distributed, cellulose filler is mixed with MAH compatibilizer 2.5% as much as 20 ml, stirred until smooth. HDPE mixed with 0.5% 10 mL BPO. These two ingredients are mixed to make the amount 100 g or 100%; after homogeneous mixing, the concentration and MAH become 0.5%, and BPO becomes 0.05%. The mixture was processed in an extruder at a temperature of 150 °C with a residence time of 15 minutes. Then the resulting pellets are printed according to test standards by hot pressing for 20 minutes, a temperature of 145 °C, and a pressure of 10 kg/cm² with a hot press. The composite was allowed to cool to room temperature and was ready for testing [16].

4. Results and Discussion

4.1 Tensile Test

Parts of composite specimens that have been printed according to ASTM D-638-72 type IV are attached to the Tensile Strength tool [17]. Figure 1 (a) shows the tensile test graph of HDPE composite with a matrix-filler ratio of 90: 10, which explains that the maximum tensile strength at that ratio is 27.0 Mpa and the strain is 27 mm/mm, with a maximum load of 274 N. Figure 1 (b) The tensile test graph for the HDPE composite with a ratio of 80: 20 shows that the maximum tensile strength of the composite is 26.0 MPa and the strain is 30 mm/mm, with a maximum load of 258 N. Figure 1 (c) Graph of the tensile test for the HDPE composite with the ratio 70: 30 indicates that the maximum tensile strength of the composite is 32.0 MPa and the strain is 36 mm/mm, with a maximum load of 316 N [18]. Based on the above analysis, the highest tensile strength is shown in HDPE: cellulose 70: 30 composites, which indicates the greater the number of fibers used as filler, the stronger the composite and the longer it takes to break and can withstand a maximum load of 316 N. Overall, all composites show high tensile strength, where the bonds between polymer components in the HDPE chain can create strong cross-links so that they are challenging to break. This indicates that the MAH compatibilizer and benzoyl peroxide initiator can open the polymer chain bonds to bind to cellulose. According to Kalpakijan, all components of composite materials have a significant binding



Fig 1. Tensile strength of composite, (a) HDPE : cellulose (90:10), (b) HDPE : cellulose (80:20), (c) HDPE : cellulose (70:30)

4.2 Surface Structure Analysis

Scanning Electron Microscopy (SEM) determined the surface structure and dimensions of the HDPE composite after being mixed with cellulose fibers were determined by Scanning Electron Microscopy (SEM). Figure 2 shows the overall surface structure of HDPE looks regular in shape with a width from 1 to 11 mm. Scanning electron micrographs show the morphological changes of the composites because they are influenced by the material composition, size, and shape of the fibers. So that the design of the material and the form of the thread need to be considered to produce a good surface structure; as shown in Figure 2, the composite ratio (70: 30) gives a better and more stable system which shows that the surface structure tends to be uniform and the combined breaks with brittle form. The composite also indicates that the fiber arrangement is compatible with the HDPE matrix, so the composite strength is also higher [19].



Fig 2. The surface structure of HDPE composite: cellulose (70: 30)

4.3 Thermal Analysis

The investigate the degradation temperature and composite decomposition, a Differential Scanning Calorimeter (DSC) was used. Figure 3 shows that the composite can withstand heat until it is degraded at a temperature of 260 $^{\circ}$ C. The melting point describes the phase change from solid to liquid without changing the composition. The melting temperature is also a critical temperature at which the overall crystallinity is lost. Composites decrease, and the degradation pressure occurs because the polymer chains are squeezed by bonds that damage the polymer molecular chains. The thermogram in Figure 3 shows that the first peak at 260 $^{\circ}$ C indicates the melting point temperature of the composite. In comparison, the second peak at 493.45 $^{\circ}$ C shows the decomposition temperature of the HDPE-cellulose composite, which is a typical characteristic of the HDPE composite thermogram. The shape of the molecular symmetry influences the melting point and the molecular weight of the polymer compound, and the degree of crystallinity of the composite material; the higher the melting point of the composite material, and vice versa [20].



Fig 3. Graphics of HDPE composite DSC thermal test with a comparison (70:30) %

4.4 Functional group analysis by FT-IR

The spectrum of Figure 4 shows that the broad absorption peak is located at 3500 cm-1, which is a stretch of the -OH group of cellulose, and the absorption peak in the region of 2800-2900 cm⁻¹ is related to the -CH₂ group, which is derived from saturated hydrocarbons of HDPE polymer and fused cellulose, where the absorption peak of 2900 cm⁻¹ is the overlap of the -CH₂ band originating from EFB-based cellulose fibers. According to Kumar et al., the absorption band in the 1400-1500 cm⁻¹ region is related to the intermolecular hydrogen in the C₆ group of the aromatic ring [21]. The absorption peaks in the wave number region of 1050 cm⁻¹ and 1150 cm⁻¹ are caused by the C-O-C stretching of alkyl aryl in lignin. The appearance of these two peaks indicates a compatible interaction between the HDPE matrix and cellulose.



Fig 4. FTIR spectrum of HDPE composites: cellulose with a ratio (70:30) %

5. Conclusion

The composite HDPE (70: 30) has better quality than the HDPE (80: 20) and HDPE (90: 10) composite with a maximum tensile strength of 32.0 MPa. The full force required to pull the HDPE composite to break (70: 30) reached 316 N. SEM showed a compatible surface structure between HDPE and fiber and brittle fracture composite. Functional group analysis showed that HDPE composites had O-H and C-O-C functional groups derived from lignocellulosic OPEFB material. The thermal stability of HDPE composites is very high, with a degradation temperature of up to 260 °C and a decomposition temperature of 493.45 °C.

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