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Modification of Recycled HDPE Composite with OPEFB Microfibers Through The Melt Blend Extruder Process

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ABSTRACT

Oil palm empty fruit bunches (OPEFB) have not been used extensively as a material with an economical value. OPEFB fiber content can be used as a filler for composites fabrication. The fiber contained in the OPEFB can increase the composite's quality because it has high thermal and mechanical properties. SEM analysis shows that OPEFB waste with a size of 63 and 90 µm is suitable to be used as a filler because of the compatibility of the fiber and matrix. As the results of the DSC analysis, the melting point of HDPE-TKKS composite with 63 µm fiber size reaches 133.58°C while the melting point of HDPE-TKKS composite with 90 µm fiber size reaches 134.41°C. The FTIR test results also show that the indications of the two samples have the same chemical composition, namely lignocellulose. The best tensile strength obtained is 21.11 MPa and the force required to break the composite is 391.56 N. These results were obtained from composites with a ratio of 60:40 (matrix:filler) and 90 µm fiber size.

***Keywords**: Composite; Fiber; HDPE; OPEFB; Plastic.*

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ABSTRAK

Tandan kosong sawit (TKS) belum banyak digunakan sebagai bahan bernilai ekonomis. Kandungan serat TKS dapat digunakan sebagai pengisi untuk pembuatan komposit. Serat yang terkandung dalam TKS dapat meningkatkan kualitas komposit karena memiliki sifat termal dan mekanik yang tinggi. Analisis SEM menunjukkan bahwa limbah TKS dengan ukuran 63 dan 90 µm layak digunakan sebagai pengisi karena adanya kompatibilzer antara serat dan matrik. Seperti halnya dengan hasil analisis DSC, titik lebur komposit HDPE-TKS dengan ukuran serat 63 µm mencapai 133,58°C, sedangkan titik lebur komposit HDPE-TKS dengan ukuran serat 90 µm mencapai 134,41°C. Hasil uji FTIR juga menunjukkan bahwa indikasi dari dua sampel memiliki komposisi kimia yang sama yaitu lignoselulosa. Kekuatan tarik terbaik yang diperoleh sebesar 21,11 MPa dan gaya yang diperlukan untuk memutuskan komposit sebesar 391,56 N. Hasil ini diperoleh dari komposit dengan perlakuan rasio 60:40 (matrix: filler) dengan ukuran serat 90 µm.

Kata kunci: HDPE; Komposit; Plastik; Serat; TKS.

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INTRODUCTION

The plastics used today are synthetic polymers, made from chemicals that cannot be degraded by microorganisms, including high-density polyethylene (HDPE), low-density polyethylene (LDPE), polypropylene (PP), polyethylene terephthalate (PET), and others. The plastic is disposable and cannot be reused so that it becomes waste in the environment. This plastic has the potential to threaten the survival of living things on earth.

Efforts to reduce environmental pollution can be done by recycling plastics by depolymerizing the plastics into other products. For example, polymers of the polyethylene terephthalate (PET) type can be depolymerized into terephthalic acid and ethylene glycol. It can be used as fiber, molding materials, and plastic cans. The use of HDPE, LDPE, and PP polymer materials is also widely applied as packaging materials, such as bags, packaging bottles, containers, and other packaging materials in modern society. Most of these HDPE and PP waste can be reprocessed into plastic drums, toys, pipes, and other products [1]. Besides that, HDPE plastic can also be made as a composite with palm empty fruit bunch (OPEFB) fiber reinforcement for motorized vehicle applications [2]. OPEFB is waste that is abundant in the environment. So far, it has not been used as a material with economical value. This waste causes problems for the environment. OPEFB is produced more than 250 tonnes per day which comes from the milling of palm oil. So far, the handling uses the incinerator combustion technique. Whereas, OPEFB can be reused in various aspects including as fiber to increase the strength of composites [3].

HDPE thermoplastic composites made from lignocellulosic fillers such as oil palm bunches 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 equipment during processing, low price, and comes from renewable resources. Thermoplastic and filler materials made from plant fibers are principally incompatible materials due to differences in polarity. So, it is necessary to modify processes such as in situ crosslinking, the addition of compatibilizers, and copolymerization of functional groups in polymers and fillers [4]. Some of the chemicals developed for compatibility between the two materials are maleic anhydrate modified-polypropylene, poly [methylene (polyphenyl isocyanate)], poly (propylene-acrylic acid), and xylene [5]. Cellulose fiber processing affects increasing the mechanical properties of composites such as tensile strength, modulus of elasticity, and hardness. Treatment with this material resulted in good results between chemical bonding with cellulose fibers in improving mechanical properties [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 filler. The matrix is wetted with 0.25% benzoyl peroxide then the filler is 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 pallet that is hard and has high strength [2]. LDPE matrix composites with a ratio of 70:30 and fiber size of 63 μm provide a high tensile strength of 12,496 MPa and an elastic modulus of 5,019 MPa [3]. 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 an adequate mix of coupling agents or compatibilizers is needed. If the filler ratio is added to the matrix, the matrix is unable to flatten the entire filler surface so that it can reduce the tensile strength value [3].

Zulnazri et al. have reported a modification of reactive process techniques to improve the compatibility of the PP matrix with lignocellulose-EFB fillers. The resulting flexural, thermal, and tensile strength properties have not shown uniformity between several mixing ratios [7]. This is because the mixing process with a single screw extruder is still not homogeneous and unable to increase the compatibility of the two materials. Therefore, in this study, HDPE Recycle with micro filler OPEFB fiber was developed into a composite product through the Melt Blend Extruder process which aims to mix HDPE and filler matrices perfectly and produce crosslinking and high compatibility.

LITERATURE REVIEW

Composite Manufacturing Techniques

Composite manufacturing can be done through a two-stage process. In the first stage, the plastic raw material is modified first. Then, the filler is mixed together in the kneader and formed into a composite. Generally, this two-stage process results in a better product. The operating conditions that are most influential in making composites are temperature, rotation rate, and kneading time. The more contact surface area of the mixed material or the smaller the particles, the better the mixing process and the higher the composite strength because the motion of the particles in the mixing is easier and more evenly on the surface [8].

The solution technique, which is used on a laboratory scale, is done by dissolving polypropylene in xylene and adding fillers with various compositions and particle sizes. The mixture is refluxed with time variations and additions with and without dicumyl peroxide as the initiator and acrylic acid as a compatibilizer to determine optimum compatibility. After the evaporation of xylene, the chemical composition of the mixture is analyzed and its physical properties are ready to be characterized. The variables tested were: temperature, residence time, die diameter, initiator concentration, the content of the compatibilizer agent, and filler content. After the reaction process, the polymer mixture is in the form of a lump or pellet and then put in a compression molded at 180°C for 3 minutes without pressure, and an additional 3 minutes with a pressure of 100 kN to form a polymer film, for microscopic, mechanical, and homogeneity testing [4].

Rozman et al. performed a chemical modification of OPEFB with maleic anhydrous (MAH) dissolved in dimethylformamide at 90°C. The MAH mixture and the mixed filler are processed in a Haake Twin-screw extruder (counter-rotating). Dicumyl peroxide 2% was added during the mixing process, operating temperature 165–180°C starting from the feed zone and the end zone, with a screw speed of 35 rpm. The mixture is then extruded and pelleted. Pellets were printed with dimensions of 17.0 × 17.0 × 0.3 cm, then preheated for 10 minutes at 180°C followed by hot pressing for 10 minutes. Cooling was carried out for 5 minutes before testing the flexural and impact properties [5].

Khalid et al. processed cellulose and OPEFB fiber mixed in various ratios above 50% with PP matrix which has been treated using Brabender twin-screw compounder at a temperature of 180°C for 20 minutes, with a roller speed of 50 rpm. The composite sheet that has been printed with a thickness of 1.2.3 mm is produced at a temperature of 190°C and a pressure of 150 kg/m^2. This process occurs preheating for 5 minutes and 3 minutes, complete pressing in a hot press followed by cooling for 3 minutes under the pressure of equipment equipped with a chiller. Then the mechanical and morphological test composites [9]. The tensile strength of PP composites increased with the addition of OPEFB fibers above 20% and 3% acrylic acid and 0.01 mol ratio of dicumyl peroxide. Meanwhile, the nature of elongation shows a downward trend. This observation also occurs when using a single screw extruder [9].

To improve the quality of the composites, a coupling agent is added to the thermoplastic polymer which aims to increase the interface adhesion between the thermoplastic and the fiber. The use of a coupling agent will form a cross-link between the polymer and the fiber so that the composite is stronger. The coupling agent that readily forms cross-links between polymers is anhydrous maleate (MAH). MAH is a compound that will form cross-links between polymers and other polymers. Cross-links can also occur between glass fibers with PP and HDPE. This cross-linking can occur well if peroxide compounds are added as initiators that help open CH bonds in PP to form radicals +R-OH [10].

METHOD

HDPE plastic as a matrix was smoothed with a crusher, the OPEFB fibers as filler were smoothed by grinding mills to sizes of 63 and 90 µm. Modification of making composites by mixing the matrix with fiber, 70 g of the matrix moistened with 20 ml of 0.25% BPO, stirred until homogeneous throughout the surface of the matrix, filler 30 g moistened with 20 ml of 2.5% MAH. Furthermore, the matrix and filler are mixed and stirred until homogeneous, the mixture is put in a screw extruder and heated for 20 minutes at a temperature of 160°C. The result is hard pellets, then molded by hot press molding, made into ASTM D-638 type IV specimen [11]. The composite quality tests carried out include Tensile Strength, SEM, FTIR, and DSC.

RESULT AND DISCUSSION

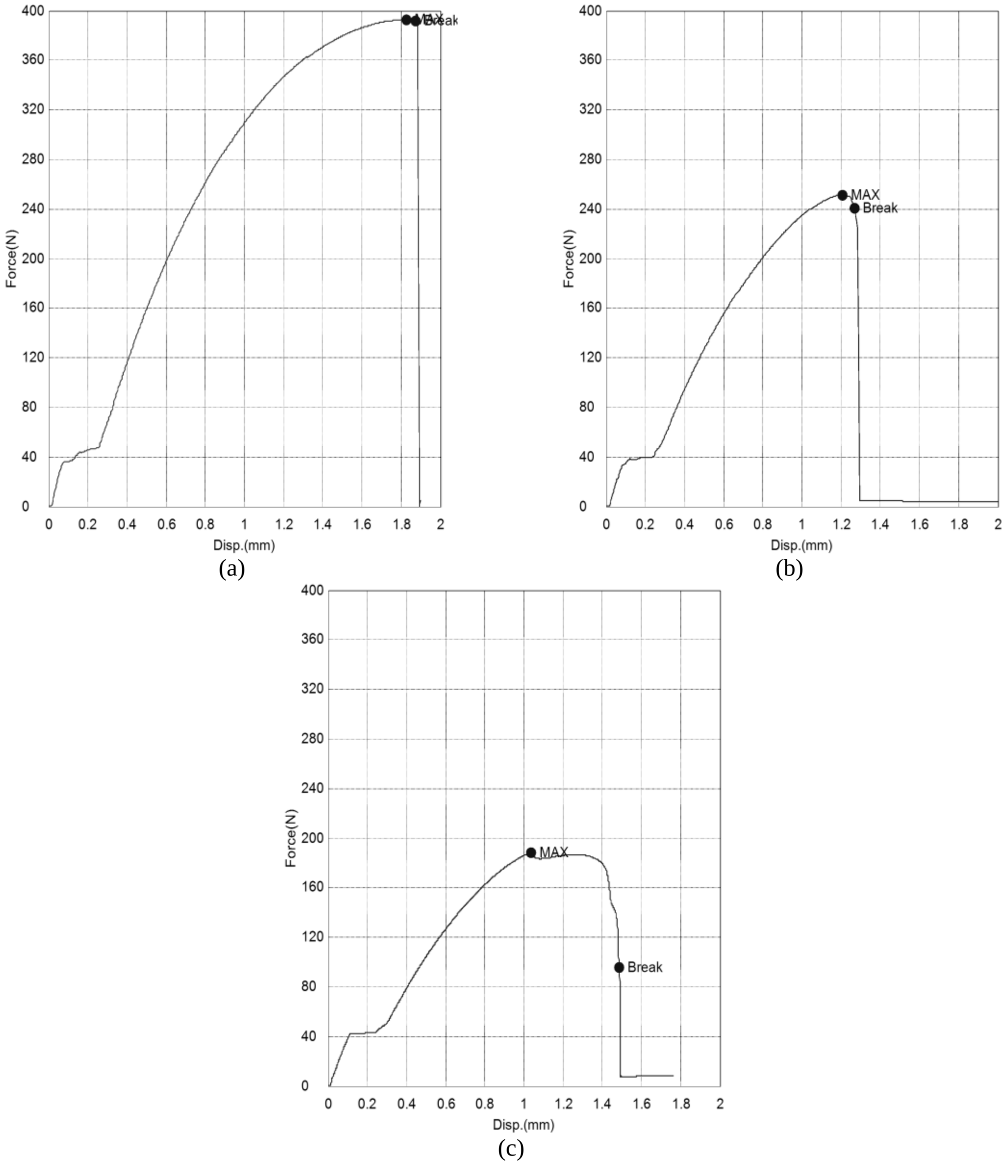
Tensile Strength

The composite visually shows a clearer appearance and better homogeneity, the physical texture of the composite looks smoother with the finer the filler size. Basuki reported that the *wetting* ability of the dispersant is an important factor affecting the distribution of the filler in the polymer matrix [4]. Table 1 shows the Tensile test results of HDPE composites with a matrix:fiber composition of 60:40 and 70:30, with a fiber size of 90 and 30 µm.

Table 1. The results of the HDPE composite tensile strength test

Name	Max Force	Max Stress	Break Force	Break Disp.
Parameters	Calc. at Entire Area (N)	Calc. at Entire Area (MPa)	Sensitivity: 10 (N)	Sensitivity: 10 (Mm)
HDPE 60:40 90 µm	392.9340	21.1084	391.5560	1.87228
HDPE 70:30 63 µm	251.3460	13.0637	240.2670	1.26898
HDPE 70:30 90 µm	187.9030	9.5820	95.6742	1.48650

Note: This data is tested in the Laboratory of Concrete and Building Materials, Department of Civil Engineering, Sepuluh Nopember Institute of Technology (ITS)



(a) (b)

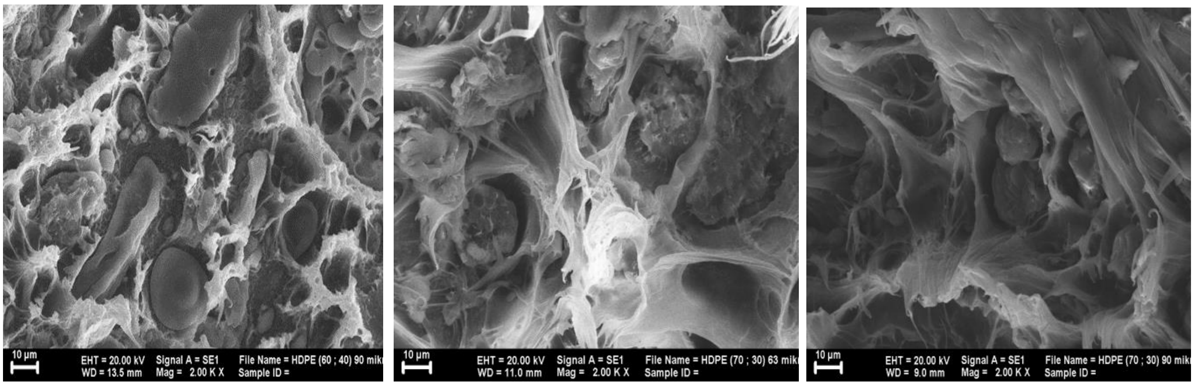
(c)

Figure 1. Composite tensile strength: (a) HDPE:fiber ratio (60:40), 90 µm fiber size; (b) and (c) HDPE:fiber ratio (70:30), 63 and 90 µm fiber sizes

Figure 1 (a) shows the maximum tensile strength of the composite reaches 21.108 MPa and the composite will fracture at 392.934 N. Figure 1 (b) shows the maximum tensile strength of the composite reaches 13.063 MPa and the composite will break at a force of 251.346 N. The maximum tensile strength of the composite reaches 9.582 MPa and the composite will break at 187.903 N. The bonds between the HDPE-filler matrices with a ratio of 60:40 is stronger than the 70:30 ratio of filler matrices. There is a strong interaction between the matrix and the small filler, where the micrometer-sized particles affect the large surface area formed so that the cross-linking that occurs between HDPE and OPEFB fibers is stronger and increases compatibility. The analysis results also show that if the composite is given a large force it will experience flexibility before breaking. According to Kalpakjian, this shows that the bond strength between the components in the composite has a significant effect on their properties [12].

Scanning Electron Microscopy (SEM)

Overall, the surface structure of HDPE composites looks regular with a width from 1 to 13.5 mm. Micrographs show changes in composite morphology because it is influenced by the size of the fineness of the fiber, so the size of the fineness of the fiber as a filler is important to provide a better surface structure, as seen in Figure 2 (b) 63 µm fiber size gives a better and regular structure compared to the image. 2 (c) and 2 (a) the fiber size is 90 µm which shows that the surface structure tends to be less uniform. The fibers shown in the figure below show the typical shape of the lignocellulosic morphology of EFB. Meanwhile, Figures 2 (c) and 2 (a) show irregular fiber shape and surface morphology that are not compatible with the HDPE matrix, while Figure 2 (b) shows uniform fibers and their arrangement is very compatible with the HDPE matrix so that the strength of the composite is also increasingly high.



(a) (b) (c)

Figure 2. Composite SEM morphology: (a) HDPE-OPEFB (60:40) 90µm, (b) HDPE-OPEFB (70:30) 63µm, (c) HDPE-OPEFB (70:30) 90 µm

Fourier Transform Infra Red (FTIR)

Figure 3 shows the FT-IR spectrum of the HDPE-OPEFB composite with a ratio of 70:30 and a fiber size of 63 µm. FT-IR analysis was carried out to determine the appearance of cellulose-OPEFB functional groups in the composite. The spectrum shows that the broad absorption peak is located at 3,366 cm-1 which is a stretch of the -OH group and the absorption in the wavenumber area 2,800–2,950 cm-1. There are two sharp absorption peaks: 2,846 and 2,914 cm-1 which are overlapping of the -CH2 band derived from lignocellulose-OPEFB fibers and HDPE polymer groups. The absorption band in the 1,470–1,577 cm-1 region is related to the intermolecular hydrogen grouped C6 (aromatic ring). This is a correlation with the research conducted by Kumar et al. [13]. The absorption peak that appears in the wavenumbers 1,018 cm-1 and 1,049 cm-1 is strongly suspected to be caused by C-O-C stretching of the alkyl aryl contained in the lignin. The appearance of these two peaks in the spectrum indicates a compatible interaction between the HDPE matrix and the OPEFB fiber.

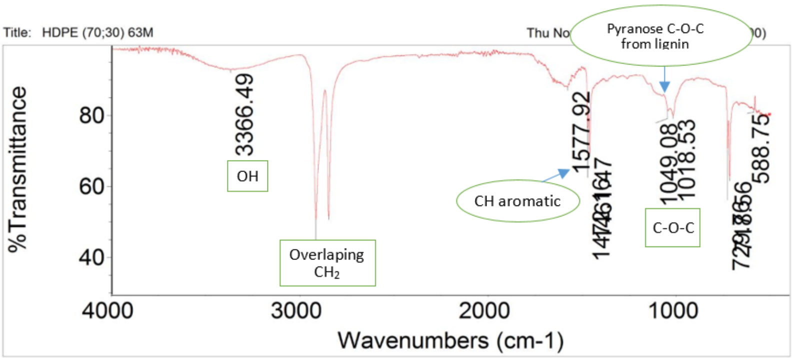


Figure 3. FT-IR spectrum of HDPE-OPEFB composite (70:30) with 63 µm fiber size

Differential Scanning Calorimetry (DSC)

DSC analysis was conducted to determine the amount of heat needed to reach the melting point. The melting point describes the phase change from solid to liquid without experiencing a change in composition. Figure 4 shows an increase in the melting point of the HDPE-OPEFB composite reaching 133.58°C, the OPEFB filler can maintain thermal stability so that the melting point obtained by the composite is high. The melting point is influenced by the shape of the molecular symmetry and the 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. The decomposition temperature is the temperature when the material changes its composition and there is a shift in the molecules contained in its constituent materials. In Figure 4, the HDPE-OPEFB composite shows its high decomposition temperature, which is due to the bonds that occur between the small and strong OPEFB fillers so that the energy required to decompose the polymer chains is also high when compared to the HDPE matrix without OPEFB fillers. According to Yu et al., the high degradation temperature of OPEFB cellulose can reduce the damage in the cellulose crystalline region and reduce the crystal decomposition [14].

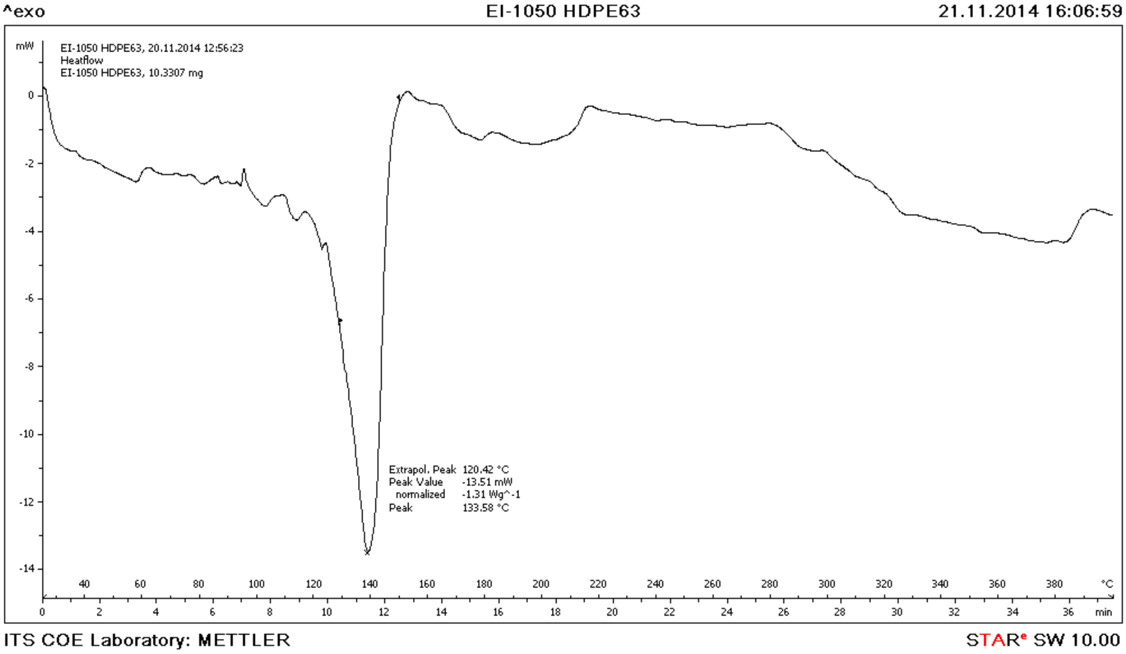


Figure 4. DSC spectrum for HDPE-OPEFB ratio (70:30) fiber size 63 µm

CONCLUSION

OPEFB fibers can improve the quality of HDPE composites because they have high thermal and mechanical properties. Composites with treatment ratio 60:40 and fiber size 90 µm showed a high tensile strength of 21.11 MPa and the force required to break the composite was 391.56 N. SEM showed OPEFB fibers with sizes 63 and 90 µm were very good to be used as fillers because of their compatibility between fiber and matrix. DSC analysis showed quite good thermal stability, where HDPE composite with fiber size 63 µm showed a melting point of 133.58°C, while HDPE composite with fiber size 90 µm showed a higher melting point of 134.41°C.

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