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Mechanical and Thermal Properties of HDPE Thermoplastic with Oil Palm Boiler Ash Nano Filler

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ABSTRACT

This study aims to determine the mechanical and thermal properties of high density polyethylene (HDPE) thermoplastic nanocomposite with oil palm boiler ash (OPBA) filler made by coprecipitation method and synthesized with PEG 6000 surfactant with OPBA-PEG 6000 filler variations. The filler composition used was HDPE/OPBA (100/0, 98/2, 96/4, 94/6, 92/8, 90/10) wt%. Nanocomposites were prepared using a Rheomixer HAAKE Polylab OS System at 150°C and 60 rpm for 10 minutes. Mechanical properties of HDPE with increased OPBA filler content is beyond a certain threshold, the tensile strength of the HDPE composite may start to decrease. This decrease can be attributed to several factors. Firstly, as the filler content increases, the HDPE matrix may become less effective in transferring stress, resulting in reduced load-bearing capacity. This is confirmed from the SEM results that the filler agglomerates and cracks occur in the composite. The composite material may exhibit a lower Young's modulus compared to pure HDPE with low Young's modulus tend to have high elongation at break which indicatie a flexible and ductile composite. The melting point of peaks 1 and 2 on 0% filler and other fillers did not change significantly even at certain compositions the melting point decreased after adding filler.

Keywords: nanocomposite, HDPE, OPBA, filler

INTRODUCTION

In recent years, there has been a growing interest in the incorporation of nanoparticle fillers into high-density polyethylene (HDPE) to further enhance its properties. Biomass fillers are organic materials that are derived from natural sources, such as wood, plant fibers, and agricultural wastes (Jawaid et al., 2017; Bonda et al., 2015). These fillers can be added to HDPE to enhance its properties and reduce its cost. However, the addition of biomass fillers to HDPE can provide several benefits. Biomass fillers are usually less, which can reduce the overall cost of the material. Biomass fillers also can improve the mechanical properties of HDPE, such as its stiffness and strength. Biomass fillers are renewable and biodegradable, which makes them a more sustainable choice (N. Bukit et al., 2018). The addition of biomass filler to HDPE can be challenging, but if properly formulated, it can lead to composite materials with improved mechanical and thermal properties. However, there are some challenges associated with the use of biomass fillers in HDPE. For example, the filler content and processing conditions can affect the properties of the final material, and the compatibility between the filler and the polymer matrix can also impact the material's performance. Therefore, it is important to carefully select the filler and optimize the processing conditions to achieve the desired properties.

The main challenges is achieving a uniform distribution of the biomass filler within the HDPE matrix (Bartoli et al., 2022). The particle size and shape of the biomass filler can affect its dispersion within the polymer matrix and the resulting composite's mechanical properties. The thermal

properties of the composite can also be affected by the addition of biomass filler. The thermal conductivity of the composite may increase due to the higher thermal conductivity of the biomass filler, which can lead to increased heat transfer. On the other hand, the thermal expansion coefficient of the composite may decrease due to the higher stiffness of the biomass filler, which can lead to increased dimensional stability (Abdul Salim et al., 2018; Fröhlich et al., 2005; Senthivel et al., 2015).

Oil palm boiler ash (OPBA) is a biomass containing silica (SiO_2) that has the potential to be used as a filler. OPBA is an agrowaste due to burning palm oil residue in the palm oil industry. Malaysia, Indonesia and Thailand are the major palm oil-producing countries, the hallmark of the leading crop in tropical countries (Frida et al., 2022; Yahya et al., 2013; Zarina et al., 2013). Oil palm boiler ash, a byproduct of the palm oil industry, has gained attention as a potential filler material due to its abundance and low cost (B.F. Bukit et al., 2022; Frida et al., 2022; Ginting et al., 2018). By incorporating it into HDPE, researchers have sought to enhance the mechanical, thermal, and electrical properties of the resulting composite. Nanosilika, as a filler has excellent dispersion in the polymer matrix because it has a high surface energy and is free to form hydrogen bonds through the hydroxyl groups on the surface (Zulfiqar et al., 2016). Processing and utilizing OPBA powder as a primary material for engineering composites requires a lot of cost and time, but the results and benefits of future research will develop well (Farida et al., 2019; Ginting et al., 2020; Mirzapour et al., 2021; Rampe et al., 2011). Therefore this study aims to utilize palm oil mill industrial waste in the form of palm oil bollard ash as a filler in HDPE thermoplastics to obtain thermoplastic nanocomposites with increased mechanical and thermal properties.

MATERIALS AND METHODS

Material

Oil palm boiler ash from palm oil plant, 5M HCL, NH_4OH Emsure merck, PEG 6000 Sigma Aldrich, High Density Polyethylene (HDPE) with Melting Point 100–135 °C, Ethanol absolute Emsure merck.

Methods

Synthesis of OPBA nanoparticles with coprecipitation method and addition of PEG-6000

Synthesis was carried out by calcination of OPBA in a furnace at 500°C for 5 hours to separate the water content contained in OPBA, then milled with Ball-Mill for 10 hours at 250 rpm rotation. The coprecipitation process was done by mixing OPBA with 5M HCL in a ratio (1:4) with a magnetic stirrer at 70°C for 4 hours at 400 rpm, then filtered using filter paper. After filtering, OPBA was mixed with NH_4OH at a ratio of 1:3 for 4 hours at 70°C using a magnetic stirrer speed of 400 rpm. OPBA solution is washed with distilled water until it produces a neutral pH. It was then dried at 150°C for 5 hours. The resulting OPBA was added to PEG 6000 1:4 (OPBA: PEG 6000) using a magnetic stirrer for 4 hours at 70°C. Then filtered again and dried at 100°C for 4 hours.

Preparation of nanocomposites

Preparation of nanocomposites using the HAAKE Polylab OS Rheomixer with various compositions as shown in Table 1. HDPE and OPBA filler were mixed at 150°C and 60 rpm for 10 minutes. For mechanical testing, samples were formed using an injection molding tool based on ASTM 638 type V with a temperature of 150˚C and a pressure of 750 bar.

RESULTS AND DISCUSSION

Fourier transform infra-red characterization

The molecular structure of the -(CH_2-CH_2) n-repeat unit of HDPE has been studied using Fourier transform infra-red (FTIR). Frequency assignment has been carried out for all fundamental vibration modes (Charles & Ramkumaar, 2009). These peaks usually include a broad band around 2900–3000 cm^{-1}, which corresponds to the stretching vibrations of

Table 1. The composition of OPBA on HDPE

HDPE (wt%)	OPBA (wt%)
100	0
98	2
96	4
94	6
92	8

CH_2 groups in the polymer backbone. Another prominent peak appears around 1470 cm^{-1}, corresponding to the bending vibrations of CH_2 groups (Booth, 1992).

Figure 1 shows strong CH_2 stretching, rocking and scissoring vibrations methylene groups in all nanocomposite samples in the absorption band. A moderate intensity band occurs around 1090 cm^{-1} which is associated with the C-C stretching vibration of the bending vibration. similarity of absorption bands on HDPE thermoplastic samples without and with filler due to the overlapping peaks between matrix and filler of the nanocomposite (B.F. Bukit et al. 2022). The FTIR spectrum can provide information about possible interactions between HDPE and the ash filler. HDPE with OPBA filler shown changes in peak intensities may indicate interactions or chemical reactions between the polymer and the filler (Ferreira et al., 2015). These changes can provide insights into the compatibility and interfacial interactions within the composite material.

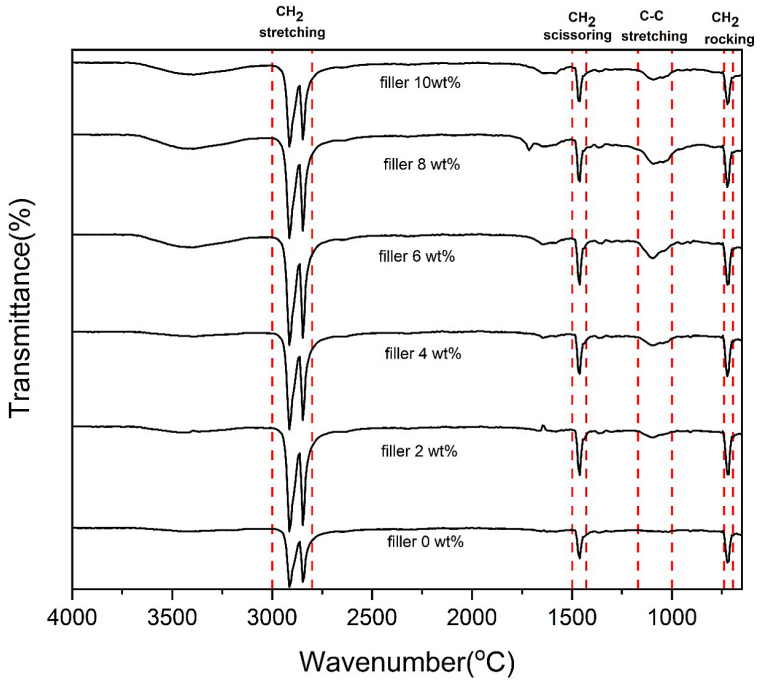

Figure 1. FTIR of thermoplastic HDPE with OPBA filler (0, 2, 4, 6, 8,10)

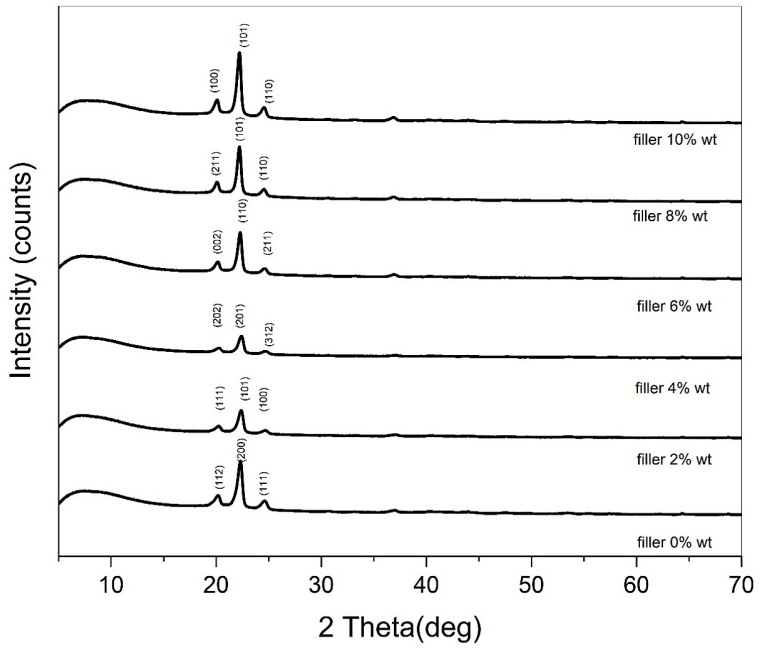

Figure 2. XRD of thermoplastic HDPE with OPBA filler (0, 2, 4, 6, 8,10) wt%

X-ray diffraction characterization

The X-ray diffraction (XRD) patterns of HDPE and HDPE incorporated with OPBA are illustrated in Figure 2. In terms of XRD analysis, HDPE typically exhibits a semi-crystalline nature. The XRD pattern of HDPE will typically show several characteristic peaks corresponding to the crystal lattice structure. The most prominent peak is usually observed around $2\theta = 22.5°$. The intensity and position of these peaks can provide information about the degree of crystallinity in the materia (Bartczak et al., 1999; Veiskarami et al., 2022). By comparing the intensities of the crystalline peaks with the amorphous baseline, it is possible to estimate the crystallinity percentage of HDPE.

HDPE XRD pattern with filler (0.2, 4.6, 8.10) wt% composite showed no change in peak position, confirming no effect on HDPE's chemical structure and crystal. While the intensity is changed depending on the wt% filler in HDPE composite (Obeid et al., 2022). X-ray diffraction pattern is different for each different phase of the composite. This confirmed that the silica from OPBA was physically adsorbed

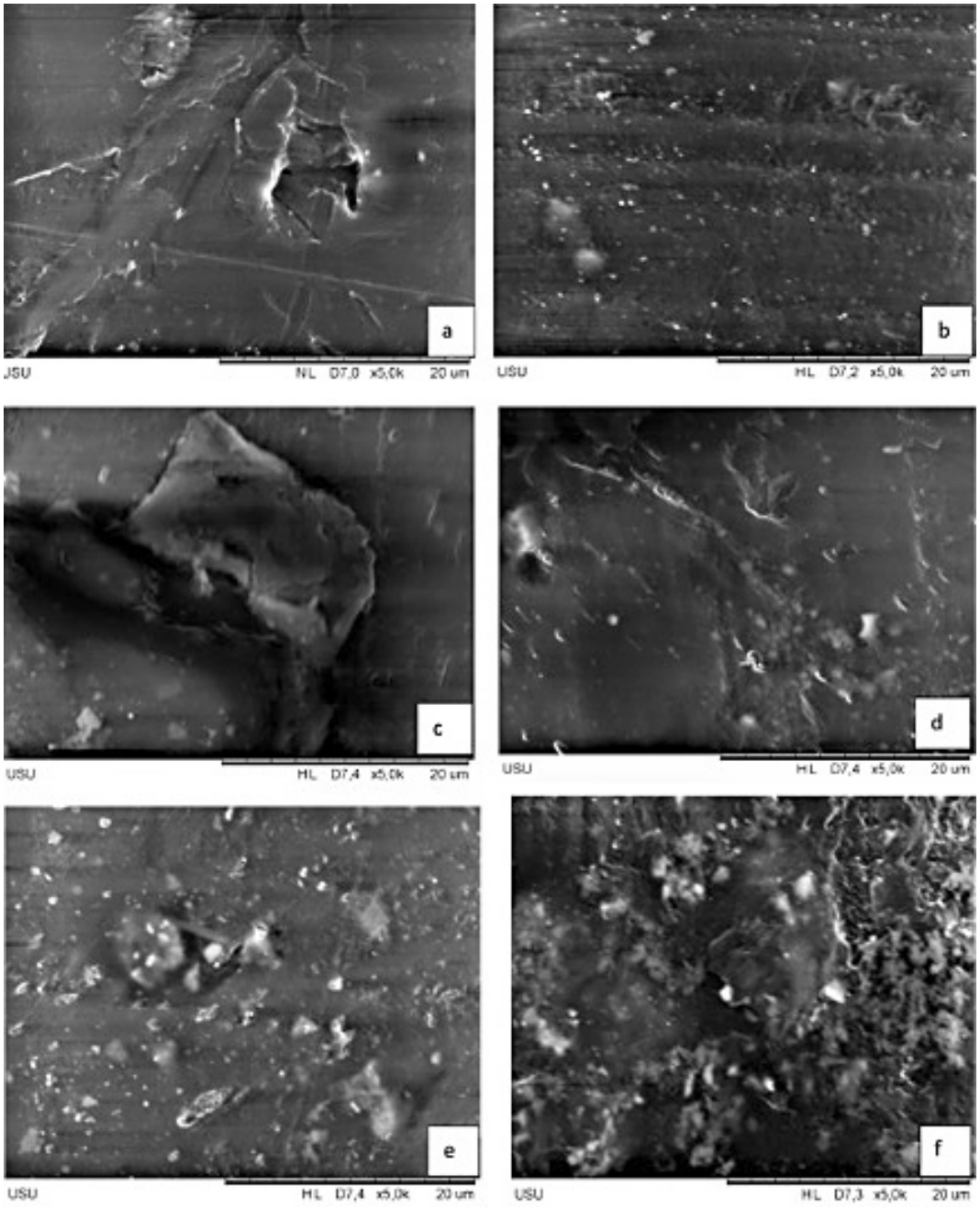

Figure 3. Morphology thermoplastic HDPE with OPBA filler: (a) 0 wt%, (b) 2 wt%, (c) 4 wt%, (d) 6 wt%, (e) 8 wt%, (f) 10 wt%

on surface polymer matrix without chemical interactions. In addition, the main characteristic peak of the composite not overlap, leading to a very accurate identification of composite crystal structure. From previous research get no substitutions occur in mixed positions while the width and different peak intensities depending on weight sample fraction (Yao et al., 2017). The relative intensity of the characteristic peaks of HDPE is attenuated with increasing particle loading and became weaker at 4.6.8 wt% filler while at 10 wt% filler the intensity increased again (Abdalsalam et al., 2020; Joshi et al., 2006; Mahmoud et al., 2018).

Scanning electron microscope

The utilization of scanning electron microscope (SEM) to investigate HDPE composites filled with oil palm boiler ash opens up new avenues for sustainable materials with improved performance characteristics. This research not only contributes to the field of materials science but also addresses the growing demand for eco-friendly alternatives in various industries.

When using oil palm boiler ash as a filler in high-density polyethylene, it is possible to encounter issues such as agglomeration and cracking. If the oil palm boiler ash particles have a tendency to stick together due to their physical or chemical properties. Agglomeration can negatively impact the homogeneity and mechanical properties of the composite material(Ninduangdee & Kuprianov, 2016). To minimize agglomeration, it is important to ensure proper dispersion of the filler particles within the HDPE matrix during the blending process. Techniques such as adequate mixing and the use of compatibilizers or coupling agents can help to improve dispersion and reduce agglomeration. Cracking can occur in HDPE composites when the addition of oil palm boiler ash filler introduces stress concentrations within the material. The differences in the coefficient of thermal expansion and mechanical properties between the filler and the polymer matrix can lead to localized stress, which may result in cracking (Ray & Rathore, 2014). Additionally, the presence of agglomerated filler particles can act as stress concentrators, further increasing the likelihood of cracking. To mitigate cracking, it is essential to optimize the composition and processing conditions of the HDPE composite. This may involve selecting filler particles with properties (such as particle size and surface treatment) that are better matched to the HDPE matrix and incorporating appropriate processing techniques, such as melt blending and compression molding (Georgopoulos et al., 2005).

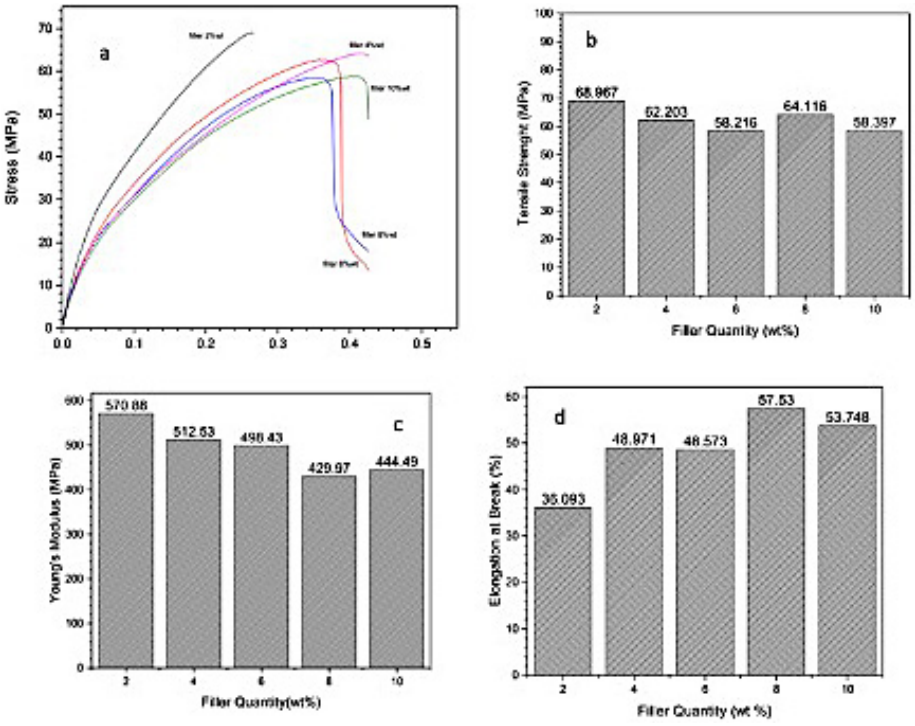

Figure 4. (a) The stress and strain relationship, (b) tensile strength, (c) Young's modulus, (d) elongation at break of thermoplastic HDPE with OPBA filler

Mechanical properties of nanocomposite

Nanocomposites were formed according to ASTM 638 type V specifications to test their mechanical properties. A stress-strain graph in Figure 4a. shows the relationship between the stress (force per unit area) applied to a material and the resulting strain (deformation) that the material experiences.

Figure 4 shown when incorporating filler materials such as oil palm boiler ash into HDPE, there can be changes in the mechanical properties of the composite. The addition of filler materials to HDPE can have both positive and negative effects on its tensile strength, depending on various factors such as filler content, particle size, and dispersion. when the filler content is increased beyond a certain threshold, the tensile strength of the HDPE composite may start to decrease. This decrease can be attributed to several factors. Firstly, as the filler content increases, the HDPE matrix may become less effective in transferring stress, resulting in reduced load-bearing capacity (Fu et al., 2008). Additionally, the filler particles can create stress concentrations in the composite, leading to premature failure under tensile loading. In the specific case of oil palm boiler ash as a filler material, it is important to consider the characteristics of the ash, such as particle size, shape, and surface properties. These factors can influence the interaction between the filler particles and the HDPE matrix, ultimately affecting the tensile strength of the composite. If the oil palm boiler ash has irregular particle shapes or poor compatibility with HDPE, it can result in reduced tensile strength when added in high concentrations (Das et al., 2022). This is confirmed from the SEM results in Figure 3. It can be seen that the filler agglomerates and cracks occur in the composite. Therefore, it is crucial to optimize the filler content and processing conditions to achieve the desired mechanical properties in HDPE composites with oil palm boiler ash or any other filler material.

HDPE is a thermoplastic polymer with a high modulus of elasticity, commonly referred to as the Young's modulus. When fillers such as boiler ash are added to HDPE, they can have an impact on the mechanical properties of the composite material. The specific effect on the Young's modulus will depend on various factors, including the type and amount of filler added, the manufacturing process, and the interaction between the filler and the polymer matrix. In general, the addition of fillers to a polymer matrix can lead to a decrease in the Young's modulus. Fillers typically have lower stiffness compared to the polymer matrix, and their incorporation can disrupt the polymer's molecular structure, reducing the overall stiffness of the composite. Boiler ash, as a filler, is often composed of various mineral particles and oxides resulting from the combustion process (Bamaga et al., 2013; Lau et al., 2019). These particles can introduce defects, voids, or weak interfaces within the HDPE matrix. As a result, the composite material may exhibit a lower Young's modulus compared to pure HDPE. Materials with low Young's modulus tend to have high elongation at break Figure 4d, indicating they are more flexible and ductile. These materials can undergo significant deformation and elongation before reaching their breaking point. Materials with high elongation at break can be applied to electrical devices & displays, special vehicles, electrical components, and infrastructure (Getu et al., 2020; Mujal-Rosas et al., 2012; Song et al., 2020). However, mechanical, thermal and microstructure evaluation of HDPE after weathering was change. Gradual and progressive mechanical breakdown properties, especially impact resistance, and elongation in rest can be associated primarily with a reduction in molecular weight and, to some extent, with an increase crystallinity. Mechanically unstable HDPE after 2520 hours of exposure, changing its ductility behavior to fragile as a result of degrading growth the structure that propagates the photo-oxidation reaction (Mendes et al., 2003).

Thermal properties of nanocomposite

DSC can provide valuable information regarding the thermal properties and behavior of the composite. DSC measures the heat flow into or out of a sample as a function of temperature or time. It can detect endothermic and exothermic transitions, such as melting points, glass transition temperatures, crystallization, and decomposition temperatures. DSC of thermoplastic HDPE with OPBA filler shown in Figure 5.

The melting point of the addition boiler ash filler usually does not significantly affect the melting point of HDPE, as it primarily depends on the HDPE polymer itself. The melting point of HDPE typically ranges from 120 to 180°C depending on the molecular weight and density of the polymer. It can be seen from Table 2 where the melting

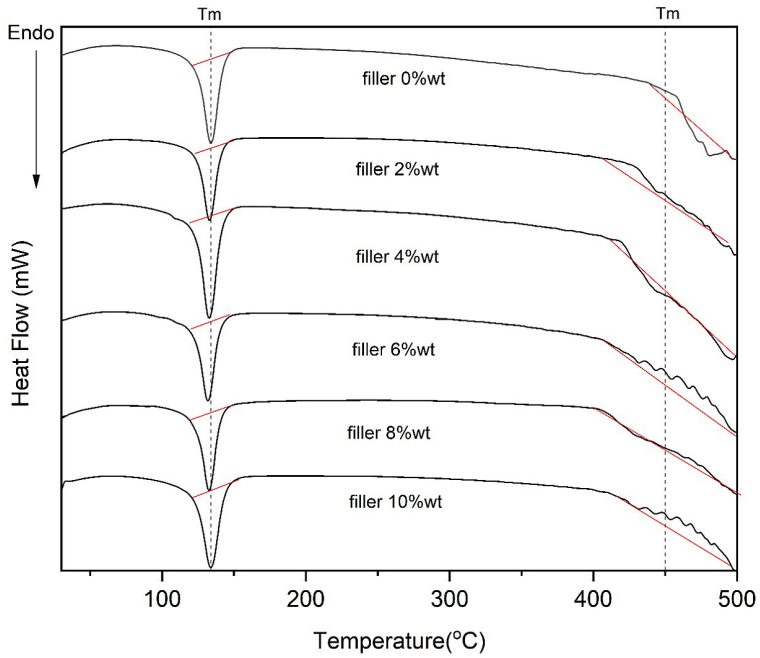

Figure 5. DSC graph of thermoplastic HDPE with OPBA filler

Table 2. DSC peak of thermoplastic HDPE with OPBA filler

Filler (%wt)	Peak 1 (°C)	Peak 2 (°C)	Onset 1 (°C)	Onset 2 (°C)	Endset2 (°C)	Endset 2 (°C)
0	134.27	379.48	124.61	455.18	143.28	480.28
2	133.39	498.59	125.05	410.34	141.40	491.75
4	132.93	496.61	124.30	419.81	142.20	556.05
6	131.83	381.37	123.28	654.06	140.74	496.82
8	132.53	380.68	124.02	380.74	141.03	497.43
10	133.67	380.73	123.53	839.69	144.05	500.60

point of peaks 1 and 2 on 0% filler and other fillers did not change significantly even at certain compositions the melting point decreased after adding filler. This decrease has also occurred in previous studies (Frida et al., 2023; Akhira et al., 2023). Changes in crystallinity can explain this decrease the thermal behavior of HDPE in the presence of ash particles, as well related to the ability of ash particles to limit the mobility of macromolecules and reduce the available space they occupy macromolecules (Ahmed, 2015; Awad et al., 2019). The thermal properties of HDPE with boiler ash filler can vary significantly depending on the filler content, particle size, distribution, and surface treatment. Therefore, the hardening effect induced by silica nanoparticles can hardly be attributed to interphase around the nanoparticles. In other words, it can be hypothesized that the hardening effect exerted by silica nanoparticles is due to the physical constraint of certain matrix parts due to nanoparticle aggregation (Dorigato et al., 2012).

CONCLUSION

Mechanical properties of HDPE with increased OPBA filler content is beyond a certain threshold, the tensile strength of the HDPE composite may start to decrease. This decrease can be attributed to several factors. Firstly, as the filler content increases, the HDPE matrix may become less effective in transferring stress, resulting in reduced load-bearing capacity. This is confirmed from the SEM results that the filler agglomerates and cracks occur in the composite. The composite material may exhibit a lower Young's modulus compared to pure HDPE with low Young's modulus tend to have high elongation at break which indicatie a flexible and ductile composite. The melting point of peaks 1 and 2 on 0% filler and other fillers did not change significantly even at certain compositions the melting point decreased after adding filler.

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