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New Material Nanocomposite Thermoplastic Elastomer with Low Cost Hybrid Filler Oil Palm Boiler Ash/Carbon Black

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ABSTRACT
This study aimed to prepare thermoplastic elastomeric nanocomposites with Low-Cost Hybrid Filler Oil Palm Boiler Ash/Carbon Black as New Material. Hybrid filler composites promise to overcome the limitations of composites. The effects offered by the matrix and filler are responsible for improving the properties of the composite. The preparation of thermoplastic elastomer was carried out in two stages. The first stage involves mixing a rubber compound with filler. The second stage is blending the compound, HDPE, and PE-g-MA using an Internal Mixer. The results show that the peak intensity increased along with the amount of OPBA in TPE. The increase in peak intensity was caused by the rise in the number of crystalline phases in the nanocomposite. In general, the absorption bands are almost the same. The samples analyzed showed absorption band vibrations (Si-O-Si), in-plane strain vibrations (Si-OH), and symmetric strain vibrations (Si-O-Si), C—H deformation, –CH symmetrical stretching of the CH_2 group appeared on each sample that FTIR has analyzed. Thermoplastics show good interaction between filler and matrix, so it can be assumed that these interactions can improve the mechanical properties of TPE. Differential Scanning Calorimetry (DSC) shows an increase in the number of peaks in the sample with 60/40 phr filler.

Keywords: waste, material, silica, compound.

INTRODUCTION

The mixture of thermoplastics with elastomers (TPE) produces the materials that have properties and functions similar to those of vulcanized rubber at room temperature. The characteristics of TPE are very useful in various industrial applications. (N. Bukit, 2013). The development of the era and rapid technological advances have resulted in the emergence of non-consumable objects, such as waste that can be used for various purposes, such as low-cost fillers.

The effects offered by the matrix and filler are responsible for improving the overall performance and properties of the composite. However, the advantages of composites reinforced with hybrid fillers depend on the filler content, filler dispersion, and surface properties. Therefore, problems are related to poor interfacial quality between filler and polymer, poor stress, transfer efficiency, and high water permeability when large amounts of filler are introduced. Recent research on the properties of a new class of hybrid composites has shown that it is likely to play a positive role in certain applications (Sarifuddin & Ismail, 2018).

Oil Palm Boiler Ash (OPBA) based nanocomposite material engineering is still rarely done. Processing and utilizing OPBA as a primary material for carbon composite engineering is beneficial for future research and will develop well (N. Bukit et al., 2019a; Ginting et al., 2020; Ginting et al., 2018). OPBA has a silica content of up to 50% which has the potential to be used as a filler (B.F. Bukit et al., 2022a; B.F. Bukit, et al., 2022b; N. Bukit et al., 2019b; Ginting et al., 2020). Both amorphous and crystalline silica, pure or functional, with non-porous or porous nano- or microparticles, are the best fillers for various polymers. Differently functionalized silica nanoparticles can be well distributed in the polymer matrix, giving good quality composite properties with linear, branched, crosslinked, polar/nonpolar, ionic, and other polymers (Gun'ko, 2018).

In the rubber manufacturing industry, carbon black (CB) is the most widely used reinforcing filler to increase abrasion, tensile and tear strength, modulus and hardness. However, CB relies on an unsustainable supply of petroleum. In recent decades, researchers have been looking for new, inexpensive and environmentally friendly reinforcing fillers. Hybrid fillers, by mixing CB with silica, show superior mechanical properties compared to CB as single-phase fillers (Rattanasom et al., 2007; Zhao et al., 2012).

MATERIAL AND METHOD

Material

High density polyethylene (HDPE) with melting point 100–135 °C, specific gravity at 200 °C, water = 0.94–0.958. Natural rubber (SIR-20), oil palm boiler ash, carbon black type N330 with a diameter of 31 nm, sulfur as curative agent, ZnO (zinc oxide) as activator, stearic acid as activator, wax as antiflux, BHT as antioxidant, ZDEC as accelerator.

Method

Sample preparation was carried out in two stages. The first stage involves mixing a rubber compound with filler. The rubber compound consists of SIR-20, a mixture of filler OPBA and CB, stearic acid, zinc oxide, MBTS, and sulfur made using two roll mixing mill. The manufacturing of the mixing process is shown in Table 1. The second stage is blending the compound 1, 2, 3, HDPE, and PE-g-MA using an internal mixer with a capacity of 50 g, with the composition of the mixture (19, 30, 1) g. The mixing process is carried out at a temperature of 150 °C, and rotor speed of 60 rpm. Furthermore, the hot and cold presses are carried out to obtain elastomeric nanocomposites in sheet form.

RESULTS AND DISCUSSION

XRD characterization

In order to investigate the possible interaction effect between OPBA and CB, characterization is carried out using XRD. The XRD used is the Shimadzu 6100 type (40 kV, 30 mA), an angle range of 2Θ = 10–70°, Cu-Kα1 = 1.5405 = 0.15406 nm. Figure 1 show the XRD of TPE. The 2θ angle of TPE 1 are at 21.98°, 24.34°, 44.40° with d_{hkl} {001, 171, 170}. Meanwhile, the 2θ angle of TPE 2 are at 2 22.05°, 24.41°, 44.43° with d_{hkl} {011, -112, -103}. The 2θ angle of TPE 3 are at 22.27°, 24.65°, 36.93° with d_{hkl} {012, 110, 200}. The peak intensity increased along with the amount of OPBA in TPE. The increase in peak intensity was caused by an increase in the number of crystalline phases in the nanocomposite (Li et al., 2009).

Table 1. Composition of compound formula with OPBA and CB filler in phr

No	Material	Compound formula (per hundred rubber)		
		Compound 1	Compound 2	Compound 3
1.	Natural Rubber SIR-20	92	92	92
2.	OPBA/CB nanoparticle	40/60	50/50	60/40
3.	Wax	1,5	1,5	1,5
4.	ZnO	5	5	5
5.	Stearic Acid	2	2	2
6.	Sulfur	3	3	3
7.	BHT	2	2	2
8.	ZDEC	4	4	4

According to the Bragg equation, this means increasing the distance between the galleries. The increased viscosity in the CB-containing nanocomposites enhances the shear transformation of the matrix towards OPBA, leading to enhanced breakdown of the OPBA stack. However, XRD results are only partially reliable in determining the exfoliation state and homogeneity of nanocomposite dispersions. Therefore, using SEM with XRD results leads to a better understanding of the morphology of nanocomposites (Barkoula et al., 2008).

FTIR characterization

To determine the functional group of thermoplastic elastomer was given filler. Then, characterization was carried out by FTIR. The FTIR used is the Agilent Cary 630 FTIR. This flexible benchtop FTIR instrument offers high performance and exceptional ease of use in an ultra-compact design. In general, the absorption bands in Figure 2 are almost the same. The absorption band at 729 cm^{-1} is associated with the presence of asymmetric strain vibrations (Si-O-Si), in-plane strain vibrations (Si-OH), and symmetrical strain vibrations (Si-O-Si). It is related to the similarity of the type of filler used in different amounts (Rezaei et al., 2014). The graft reaction between each group leading to this characteristic graft band overlaps with the C—H deformation at 1080–1472 cm^{-1} (N. Bukit et al., 2022; Kaynak & Meyva, 2014). The peaks at 2847–2914 cm^{-1} correspond to the –CH symmetrical stretching in the CH_2 group.

SEM characterization

Dispersion of OPBA and CB is one of the most significant factors determining the quality of TPE, which is fed with filler. By using SEM, the visualization of the dispersion filler on the TPE can be observed. SEM characterization was carried out using the SEM TM3030 model. Hitachi High-Tech has provided a "5 kV mode" that allows for sharper observations of the surface structure of the finest samples, which cannot be observed at high accelerating voltages. The morphology of TPE is shown in Figure 3.

The morphology in Figure 3 shows that the thermoplastic elastomer appears dark in color, but the carbon black and OPBA particles appear as bright spots. In the picture with the composition of OPBA and CB 50:50%, it can be seen that the number of bright spots increased compared to other compositions. It is also seen that OPBA and CB are well dispersed in the matrix. In addition, the smooth TPE surface is also visible. It was proven that the interaction between filler and matrix is good, so it can be assumed that the interaction can improve the mechanical properties of TPE.

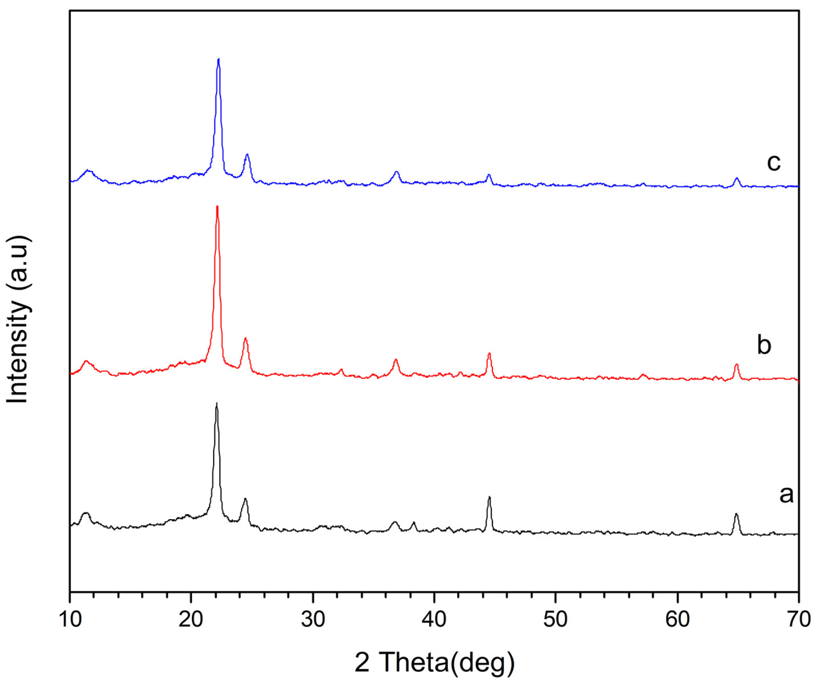

Figure 1. XRD spectrum of thermoplastic elastomer: (a) TPE 1, (b) TPE 2, (c) TPE 3

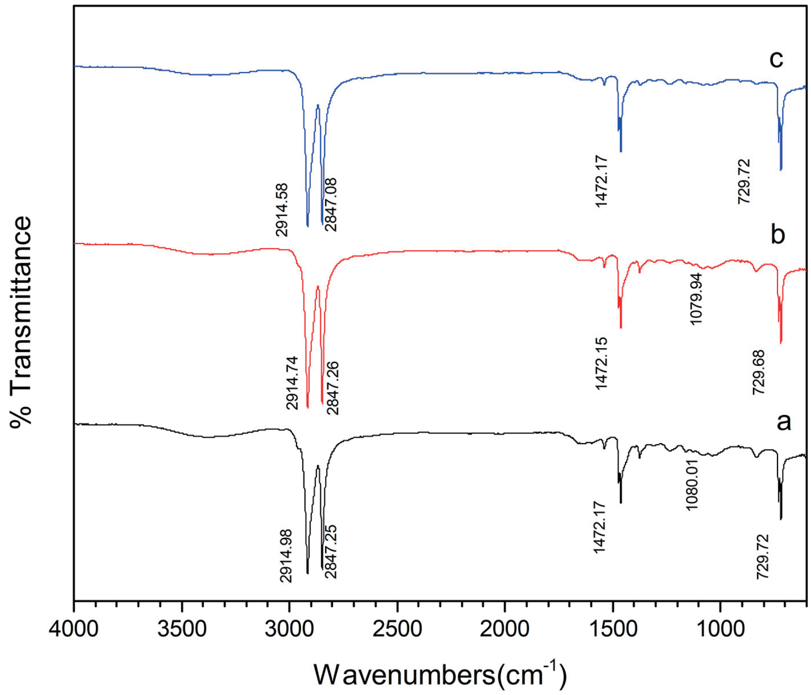

Figure 2. FTIR of thermoplastic elastomer: (a) TPE 1, (b) TPE 2, (c) TPE 3

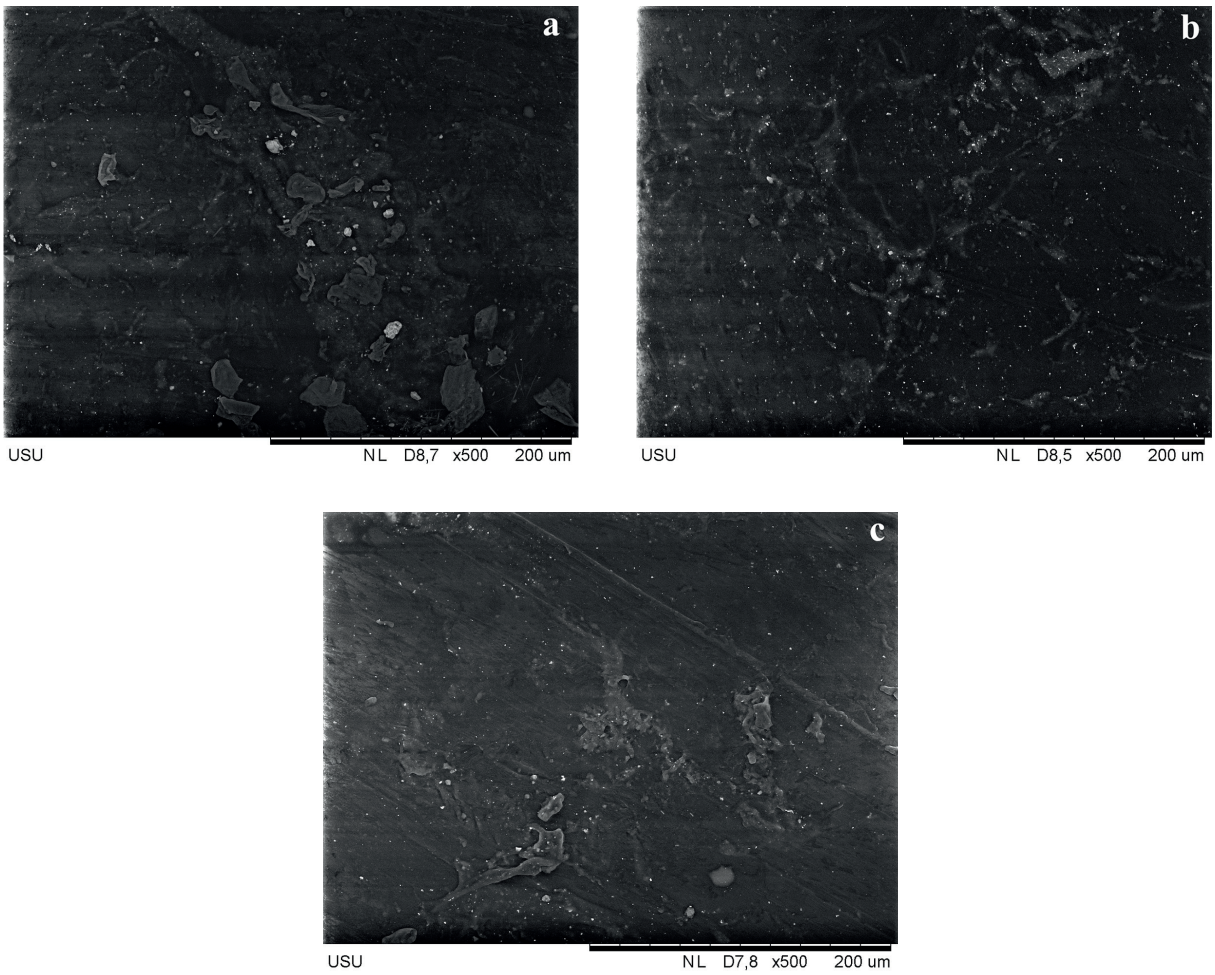

Figure 3. Morphology of (a) TPE 1, (b) TPE 2, (c) TPE 3

DSC characterization

Thermal analysis methods play an essential role for structure-property relationship characterization in individual polymers and polymer blends (Grigoryeva et al., 2006). Several techniques are widely applied to research the phase structure and thermal properties of polymer blends. DSC thermogram was performed to characterize the thermal behavior of TPE with OPBA and CB fillers. DSC analysis of TPE mixtures can be used to evaluate strengthening effect, and rubber compatibility mixture, as well as rubber-filler interactions. In this study, Shimadzu DSC-60 Plus was used to analyze the thermal properties of rubber compounds. Figure 4 shows differential scanning calorimetry analysis of TPE.

Table 2 shows an increase in the number of peaks in TPE 3 and a decrease in temperature. A glass transition temperature and a melting temperature indicate that the material is semicrystalline and can be processed. In general, polymers are semicrystalline. In other words, some polymer chains are crystalline, and some are amorphous. Melting temperature is an essential parameter for thermoplastics because it represents the minimum temperature required for polymer processing. Melting behavior is highly dependent on the chemical structure of a material, along with its size and regularity of crystallization found in the crystalline phase (Farida et al., 2019).

CONCLUSIONS

Nanocomposite thermoplastic elastomer with hybrid filler oil palm boiler ash and carbon black showed promising results. The peak intensity increased when the amount of OPBA in the TPE

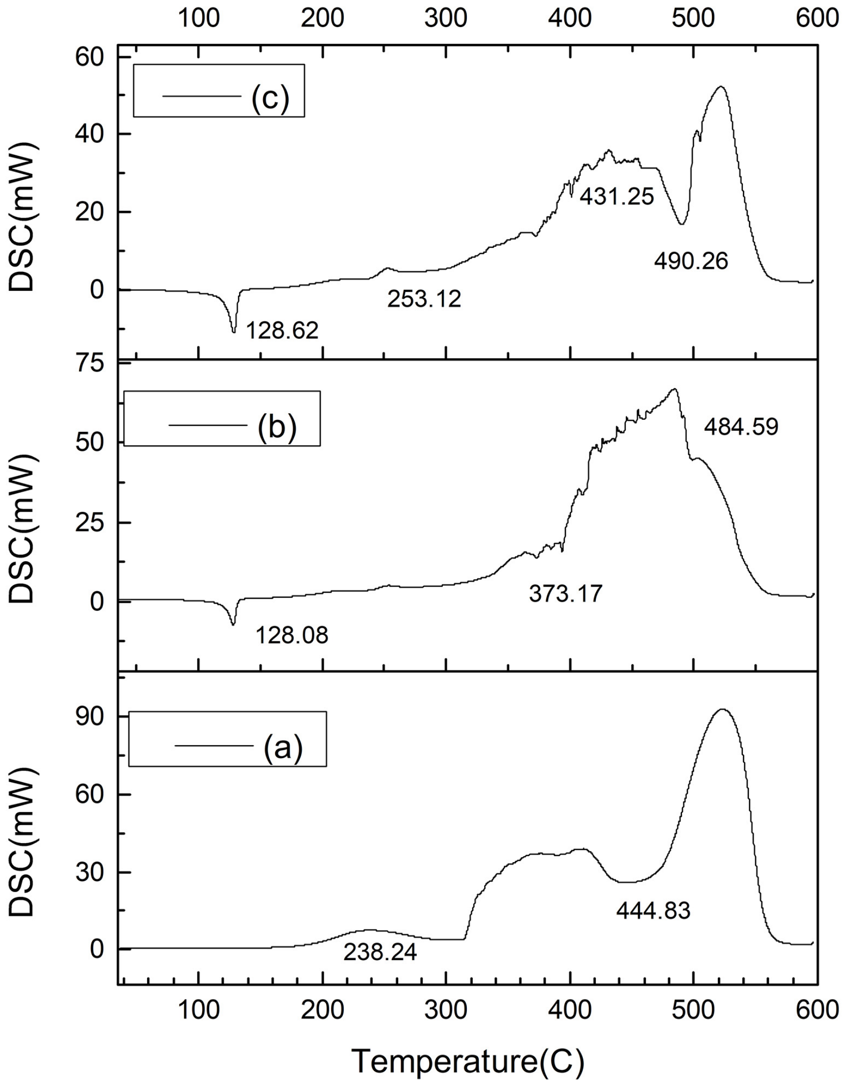

Figure 4. DSC thermogram of TPE: (a) TPE 1, (b) TPE 2, (c) TPE 3

Table 2. Differential scanning calorimetry peak list of thermoplastic elastomer

Peak list	Temp. onset (°C)	Temp. endset (°C)	Peak (°C)
TPE 1			
Peak 1	209.26	271.83	238.26
Peak 2	398.17	419.35	411.17
Peak 3	429.61	461.98	444.85
Peak 4	515.03	529.87	522.77
TPE 2			
Peak 1	122.93	131.88	128.08
Peak 2	371.67	374.54	373.17
Peak 3	477.14	489.62	484.59
TPE 3			
Peak 1	122.43	132.48	128.62
Peak 2	242.90	260.48	253.12
Peak 3	426.48	437.62	431.25
Peak 4	471.58	498.42	490.36
Peak 5	512.95	528.35	521.51

was increased. The increase in peak intensity was caused by the rise in the number of crystalline phases in the nanocomposite. In general, the absorption bands are almost the same. The samples analyzed showed absorption band vibrations (Si-O-Si), in-plane strain vibrations (Si-OH), and symmetric strain vibrations (Si-O-Si), C—H deformation, –CH symmetrical stretching of the CH_2 group appeared on each sample that FTIR has analyzed. Thermoplastics show good interaction between filler and matrix, so it can be assumed that these interactions can improve the mechanical properties of TPE. Differential Scanning Calorimetry shows an increase in the number of peaks in the sample with 60/40 phr filler.

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