Effect of Bi_2O_3 on the properties of linear low density polyethylene (LLDPE)/natural rubber (NR) composites

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ABSTRACT

The use of thermoplastic natural rubber (TPNR) has spread for many applications such as in footwear, hoses, seals, and automobiles. This increase is in line with the environmental awareness to produce materials that can be recycled. In this paper, the making of TPNR for car floor mat was studied. Bi_2O_3 filler was added to modify the performance of TPNR for car floor mat. In this study, the effects of Bi_2O_3 filler on the properties of linear low-density polyethylene (LLDPE)/natural rubber (NR) composites had been investigated. The weight ratio of LLDPE/NR was varied at 90/10; 80/20; 75/25; and 70/30. Bi_2O_3 filler loading was varied at 0; 20; 40; and 50 phr. The increase in NR and Bi_2O_3 filler reduced the tensile strength, elongation at break, and tear resistance, but increased the hardness and density of the composites. Compared to similar imported products, the samples prepared in this study showed higher values for all mechanical properties (tensile strength, elongation at break, tear resistance) but lower values in density. Scanning electron microscopy (SEM) micrograph of LLDPE/NR 75/25 composites either with or without the addition of 20 phr Bi_2O_3 filler displayed homogeneity of the mixture.

Keywords: bismuth trioxide, filler, LLDPE/NR composite.

INTRODUCTION

The use of thermoplastic natural rubber (TPNR) is spreading to many applications, such as in footwear, hoses, seals, automobiles, and marine engineering with the increasing interest in current environmental issues (Homkhiew *et al.*, 2018). Thermoplastic natural rubber (TPNR) can be obtained from a blend of thermoplastics and natural rubbers. Among the thermoplastics, one of the most used in the world industry is linear low-density polyethylene (LLDPE). LLDPE is used in the industry of packaging (Cozzi *et al.*, 2018), electrical insulating materials in power cables (Arief *et al.*, 2016), and automobiles (Ramkumar *et al.*, 2014). In this paper, the making of TPNR for car floor mats was studied. Car floor mats are generally made from natural rubber and synthetic rubber, then vulcanized to provide products that are difficult to recycle. The development of LLDPE/NR based TPNR materials was aimed to provide recyclable, high-density, and thermoformable car floor mats. Currently, high density TPNR is an

imported product.

The blend of LLDPE/NR has been widely studied by some researchers because LLDPE is suitable for blending with natural rubber attributable to its low melting temperature which can prevent the thermal degradation of rubber. However, the nature of TPNR, as a polymer, limits in several physical and mechanical properties, particularly hardness and stiffness. The addition of filler can improve the performance of TPNR. Arief *et al.* (2016) studied composites of linear low-density polyethylene/natural rubber (LLDPE/NR) with nano-silica and micro-scale Palm Oil Fuel Ash (POFA) fillers, and they found that the tensile strength of LLDPE/NR raised with the increasing of nano-silica content composition, but decreased with the increasing of POFA weight percentage.

The use of high-density metal oxide fillers in thermoplastic material formulations are developed for a diverse specific end-use, for instance radiation shielding, projectiles, electronic components, and packaging, offers benefits, such as sustainable,

non-toxic formulations, an excellent balance of properties, and the ability to be melt-processed using common methods. The amount of high-density metal oxide filler to be mixed into the thermoplastic material formulations depends on the presence or absence of other fillers, as well as the desired physical properties of the resulting material. Generally, silica (SiO_2), alumina (Al_2O_3), titania (TiO_2), and zinc oxide (ZnO) are common high-density metal oxides used as filler in thermoplastic elastomer (Abdullah & Ibrahim, 2014; Ramlee *et al.*, 2015; Villani *et al.*, 2020; Simões *et al.*, 2017). Bismuth trioxide (Bi_2O_3) fine powder material is one of the metal oxides that is selected to substitute for lead (Pb) in gamma shielding materials because it is non-toxic compared to lead metal. Some researchers had reported the application of Bi_2O_3 as a filler in the manufacture of gamma shielding materials based on natural rubber (Toyen *et al.*, 2018), on EPDM (Poltabtim *et al.*, 2018), and polyimide (Pavlenko *et al.*, 2019). In thermoplastic polyurethane (TPU), Bi_2O_3 was used as a laser-marking additive. The reason for choosing Bi_2O_3 was due to its high density, high melting point, low conductivity, and its availability in fine powder form (Ambika *et al.*, 2017). However, there is still no study reported on the application of Bi_2O_3 as a filler in the LLDPE/NR composites.

In this paper, LLDPE/NR/ Bi_2O_3 , a novel reactive material, was designed and prepared for the first time. The effect of Bi_2O_3 content and the ratio of LLDPE/NR on the physical and mechanical properties of composites were studied. This study was aimed to investigate the effects of Bi_2O_3 filler loadings and the ratio of LLDPE/NR on the physical and mechanical properties of LLDPE/NR composites for car floor mats.

MATERIALS AND METHODS

Materials

Natural rubber (NR) compound with a density of 1.12 g/cm^3 , a hardness of 56.6 Shore A, a tensile strength of 232 kg/cm^2 , elongation at break of 467%, a tearing strength of 101 N/mm , and a 30% compression set of 2.654% was used in this study. Linear low-density polyethylene (LLDPE) granules (Asrene UF 1810T, extrusion grade, a density of 0.921 g/cm^3 , a tensile strength of 244 kg/cm^2 , elongation at break of 816%, and a melt index of 1 g/10 min at $190 \text{ }^\circ\text{C}/2.16 \text{ kg}$) was purchased from a local supplier. Maleic anhydride-

graft-polyethylene (MA-g-PE) as a compatibilizer and the commercial antioxidant (Irganox 1010) were purchased from Sigma Aldrich supplier.

Methods

Composite preparation

LLDPE/NR composites were prepared from LLDPE, NR compound, Bi_2O_3 , compatibilizer, and antioxidant in a Haake Rheomix at $130 \text{ }^\circ\text{C}$ (Zailan *et al.*, 2018), a rotor speed of 40 rpm for 15 minutes. The weight ratio of LLDPE/NR was varied from 90/10; 80/20; 75/25; and 70/30. Bi_2O_3 filler loading was varied by 0; 20; 40; and 50 phr (per hundred resin). Maleic anhydride-graft-polyethylene (MA-g-PE) as a compatibilizer and antioxidant Irganox 1010 content was fixed at 5 phr and 0.1 phr, respectively for all formulations.

Characterization

The mechanical properties of the composite samples were tested using a Universal Testing Machine (UTM) Tinius Olsen-H25K) to observe the tensile strength, elongation at break, and tear strength. The tensile strength and elongation at break tests were carried out according to ISO 37:2017 (ISO, 2017). The samples were cut into a dumbbell shape type 2 with the length of a narrow portion of $25 \pm 0.1 \text{ mm}$ and a test length of $20 \pm 0.5 \text{ mm}$. The tests were performed at laboratory temperature ($23 \pm 2 \text{ }^\circ\text{C}$) and a crosshead speed of 500 mm/min . Three specimens of each formulation were tested and the average values were calculated. Tear strength testing was carried out according to ISO 34-1:2015 (ISO, 2015) with an angle-type specimen. The hardness of the composites was tested in accordance to ISO 48-4:2018 (ISO, 2018a) using Shore A durometer with a specimen thickness of 12 mm . The density was tested with an Electron Densimeter (Mirage EW-200SG) using the procedure of ISO 2781:2018 (ISO, 2018b) Method A by comparative measurement of samples' mass in air and water. The surface morphology of composites was observed using an electron microscope (SEC-SNE 3200M). Before testing, the samples were sputter-coated with a thin layer of gold.

RESULTS AND DISCUSSION

Mechanical Properties

The test results of mechanical properties such as tensile strength, elongation at break, tear resistance, and hardness of LLDPE/NR composites

as a function of Bi_2O_3 content are shown in Figure 1. Figure 1 shows that all mechanical properties of the composites decreased with the increasing of NR and Bi_2O_3 fillers content, except for shore hardness. Shore hardness slightly increased with the enhancement in the NR and Bi_2O_3 loading.

The tensile strength of the composites was concerned, they decreased with an increase of NR loading (Figure 1a). This might be due to the value of tensile strength of the rubber compound used (232 kg/cm^2) was lower than the LLDPE tensile strength value (244 kg/cm^2). A similar trend was studied by Wickramaarachchi *et al.* (2016) and Mastalygina *et al.* (2020), who reported that the tensile strength of polyethylene/natural rubber blends increased with the addition of polyethylene loading. The highest tensile strength (205 kg/cm^2) was achieved for LLDPE/NCR 90/10 composite without Bi_2O_3 filler. Figure 1a also shows the effect of the presence of Bi_2O_3 filler on the tensile strength for each composition. The rose of Bi_2O_3 filler loading range from 20-50 phr decreased tensile strength (Figure 1a). This was probably caused by poor adhesion between Bi_2O_3 particles and the LLDPE-NR matrix, thus weakened the interfacial zone between the polymer and Bi_2O_3 particles. This weak zone increased with the enlarging of filler content and reduced the tensile strength, elongation at break, and tear resistance of the composite (Al-Mattarneh & Dahim, 2019). When 20 phr of Bi_2O_3 filler was added to LLDPE/NR 70/30 composite, the tensile strength decreased from 185 kg/cm^2 to 95 kg/cm^2 (kg/cm^2) (decreased 48.6%). The tensile strength of LLDPE/NR 90/10 composite (205 kg/cm^2) decreased with the addition of 2 phr of Bi_2O_3 particles to 164 kg/cm^2 (decreased 20%). All the LLDPE/NR composites of this study have higher tensile strength when compared to the imported thermoplastic elastomer (TPE) for the same application which has a tensile strength of 23.7 kg/cm^2 .

Elongation at break is a description of the ductility of a material, which is the opposite of brittle (Onuoha *et al.*, 2017). As shown in Figure 1b, the elongation at break decreased gradually with the enhancement of NR and Bi_2O_3 fillers loading. The elongation at break of LLDPE/NR composite decreased with the addition in NR contents. The reduction of elongation at break was due to the value of elongation at break of rubber compound (467%) was lower than LLDPE (816%) and occurrence of stiffening of the polymer

matrix by the filler. A similar trend was studied by Okele *et al.* (2018) who reported that hardness of polypropylene/natural rubber blends increased with the increment of polypropylene. The LLDPE/NR composite of 90/10 ratio has 650% of elongation at break, it is higher about 17.7% compared to the composite ratio of 70/30 that has 535% of elongation at break. According to Mastalygina *et al.* (2020), NR addition to PE reduced the elongation at break. Moreover, it can be seen that the elongation at break of the composites declined as Bi_2O_3 filler loading increased in the LLDPE/NR composites. The increase in filler loading causes the ductility of the matrix decreases, and the elongation at break is a reflection of the ductility of a material. The addition in 20 phr of Bi_2O_3 filler loading led to elongation at break reduction from 650% to 513% for LLDPE/NR 90/10 and from 535% to 387% for LLDPE/NR 70/30 composites. Onuoha *et al.* (2017) also reported that elongation at break decreased steadily with filler loading and particle size. Arief *et al.* (2016) reported that the higher loading of the palm oil fuel ash (POFA) filler reduced the volume of the matrix of LLDPE-NR composite, which led to the decreasing trend of elongation at break (%). The elongation at break of all the LLDPE/NR composite is higher than the elongation of break of the imported TPE for the same application that has value 23.3%.

Figure 1c shows the Shore hardness of LLDPE/NR composites increased with the enlargement of LLDPE loading. The highest Shore hardness of LLDPE/NR composites without Bi_2O_3 filler (85 Shore A) was achieved at 90 phr of LLDPE and 10 phr of NR. This was because the Shore hardness of LLDPE (90 Shore A) was higher than the Shore hardness of NR (56.6 Shore A). Wickramaarachchi *et al.* (2016) and Homkhiew *et al.* (2018) also found that the enhancement of HDPE in the HDPE/NR composites increased the Shore hardness of the composite. A similar trend was studied by Okele *et al.* (2018) who reported that the hardness of polypropylene/natural rubber blends raised with an increase of polypropylene. Furthermore, it was shown that the Shore hardness of the composites progressively escalated as Bi_2O_3 fillers loading increases in the LLDPE/NR composites. The reason was the metal oxide filler (Bi_2O_3) has higher hardness than LLDPE/NR composites and the addition of metal oxide into LLDPE/NR phases decreases elasticity of polymer chains, resulting in more rigid composites. The

addition of filler into a polymer matrix increases the stiffness and hardness of the composite (Onuoha *et al.*, 2017). For LLDPE/NR 70/30 composites, the addition of Bi_2O_3 filler at 20 phr increased Shore hardness from 80 to 89 Shore A, thus there was an increase of 11.25%. However, for LLDPE/NR 90/10 composites, the addition of

Bi_2O_3 filler at 20 phr increased Shore hardness from 85 to 92 Shore A, thus only it increased 8.24%. The imported thermoplastic elastomer (TPE) for the same application has a hardness of 85 Shore A, this value is lower than the hardness of all the LLDPE/NR composites containing Bi_2O_3 filler.

The tear strength of LLDPE/NR composites

Figure 1. Mechanical properties of LLDPE/NR composites as a function of Bi_2O_3 content: tensile strength (a), elongation at break (b), hardness (c), tear strength (d).

decreased with increment in the NR and Bi_2O_3 loading (Figure 1d). As shown in Figure 3d, the rise of NR loading reduced the tear strength of LLDPE/NR composites. The highest tear strength of LLDPE/NR composites without Bi_2O_3 filler (122 N/mm) was achieved at 90 phr of LLDPE and 10 phr of NR. Wickramaarachchi *et al.* (2016) also reported that the tear strength of polyethylene/natural rubber composites decreased with increasing of natural rubber. Furthermore, the addition of Bi_2O_3 filler to LLDPE/NR composites contributed in reducing the tear strength of the composites. When 20 phr of Bi_2O_3 filler was added to LLDPE/NR 70/30 composites, the tear strength decreased from 115 N/mm to 80.9 N/mm (decreased 29.6%). The tear strength of LLDPE/NR 90/10 composite 122 N/mm decreased to 109 kg/cm² (decreased 10.6%) with the addition of 2 phr of Bi_2O_3 particles. When compared to the imported TPE for the same application which has a tear strength of 27.3 N/mm, all LLDPE/NR composites of this study have higher tear strength.

Physical Property

In this study, the physical property of LLDPE/NR composites were represented by density. High-

density metal oxide fillers were used to improve some properties of the thermoplastic composition, including density. The test result of the density of LLDPE/NR composites as a function of Bi_2O_3 content is shown in Figure 2. In accordance with the rule of mixture, the density of a particulate filled composite is related to the density of its constituent particles.

As shown in Figure 2, the density increased as the amount loading of NR and Bi_2O_3 increased. The rise in density was due to the density of Bi_2O_3 (8.9 g/cm³) was much greater than the density of rubber compound (1.12 g/cm³) and LLDPE (0.921 g/cm³). Meanwhile, the density of the rubber compound was also greater than the density of LLDPE. In this study, the LLDPE/NR composites had a density range of 1.07-1.28 g/cm³. Al-Mattarneh and Dahim (2019) also reported similar phenomena where the density of TPNR increased with the increasing of barium ferrite filler content because of the higher density of barium ferrite compared to TPNR.

According to Ribeiro *et al.* (2019), the density of TPE increased by raising the copper microparticle filler content in the TPE compounds because copper has a higher density than the other constituents of the formulation. The highest

Figure 2. The density of LLDPE/NR composites as a function of Bi_2O_3 content.

density (1.28 g/cm^3) was observed for composite with an LLDPE/NR 70/30 containing 50 phr of Bi_2O_3 and gave an increment of about 15.32% in density compared to the standard LLDPE/NR 70/30 without loading filler. The lowest density (1.07 g/cm^3) was shown by the composites with LLDPE/NR 90/10 without Bi_2O_3 content. The density of all LLDPE/NR composites were lower than the density of the imported TPE for the same application that has value 1.75 g/cm^3 . Therefore, to increase the density of LLDPE/NR composites to match the density of the imported TPE, the amount of loading of Bi_2O_3 filler must be increased.

Morphological Analysis of Bi_2O_3 Filled LLDPE/NR Composites

The homogeneity of the mixtures between polymer matrix and filler in the composite was analyzed by scanning electron microscopy (SEM). Figure 3 displays the SEM images of the LLDPE/NR 75/25 composite containing various loading

of the Bi_2O_3 filler. Figure 3a presents that the NR has blended well in the LLDPE matrix. A similar trend can be seen in the study by Sampath *et al.* (2019) who reported that the SEM image of NR/LDPE 70/30 composite showed that LDPE is clearly dispersed in the NR matrix and which was interpreted as good interfacial adhesions between the NR and LDPE phases.

The composite with 20 phr (Figure 3b) displayed a better dispersion of filler than composites with 40 and 50 phr Bi_2O_3 filler loading (Figure 3c and 3d, respectively). On the other hand, Figure 1b shows the ability of a mixture of both polymers and Bi_2O_3 fillers to mix well in a polymer matrix. However, as shown in Figure 3c and 3d, Bi_2O_3 filler has a tendency to form agglomeration indicating that the interaction between the polymer matrix and Bi_2O_3 fillers was weak. The effect of interfacial adhesion significantly affected the mechanical and physical properties of composites (Homkhiew *et al.*, 2018).

Figure 3. Micrograph SEM of LLDPE/NR 75/25 composites with 0 phr (a), 20 phr (b), 40 phr (c), 50 phr (d) of Bi_2O_3 filler loading (10000x magnification).

CONCLUSIONS

In this study, the LLDPE/NR-based composite for car floor mat application was developed. Compared to similar imported products, the prepared samples showed higher values in all mechanical properties, but lower in density. All mechanical properties (tensile strength, elongation at break, tear resistance), except the Shore hardness, of the composite decreased with the increasing content of both NR and Bi_2O_3 filler. Shore hardness only slightly increased with increasing NR and Bi_2O_3 loading. The highest tensile strength (205 kg/cm^2), elongation at break (650%), and tear strength (122 N/mm) were achieved for LLDPE/NCR 90/10 composite without Bi_2O_3 filler. The highest Shore hardness (92 Shore A) was achieved for LLDPE/NCR 90/10 composite with 50 phr of Bi_2O_3 filler loading. The physical properties of LLDPE/NR composites that represented by density increased with the increment of NR and Bi_2O_3 fillers loading. The highest density (1.28 g/cm^3) was observed for composite with a ratio of LLDPE/NR 70/30 containing 50 phr of Bi_2O_3 . Scanning electron microscopy (SEM) micrograph of LLDPE/NR 75/25 composites with and without the addition of 20 phr Bi_2O_3 filler displayed homogeneity of the mixture. However, increasing Bi_2O_3 filler loading tends to form agglomeration.

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