High-density thermoplastic vulcanizates based on LLDPE/NR for truck floor mats application

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ABSTRACT

The use of thermoplastic vulcanizates (TPVs) that was obtained from a blend of thermoplastics and natural rubbers are considered suitable for the automotive application. To modify the performance of TPVs to high-density TPVs, a high-density filler such as bismuth oxide (Bi_2O_3) was added. The focus of this study was on the development of high-density TPVs based on linear low-density polyethylene (LLDPE)/natural rubber (NR) blend with different proportions of Bi_2O_3 filler that meets with requirements for truck floor mats material. The high-density TPVs were prepared by melt blending in a kneader. Bismuth oxide filler loading was varied by 50, 150, 200, 250 phr, and 300 phr. An increase in the weight load of bismuth oxide in TPVs increases density, hardness, and volume loss but reduces the tensile properties, tear strength, and burning rate of high-density TPVs. The test results showed that the TPVs containing 250 phr Bi_2O_3 was a promising candidate for use as a truck floor base.

Keywords: high-density thermoplastic vulcanizates, Bi_2O_3 , linear low-density polyethylene, natural rubber, truck floor mats material.

INTRODUCTION

Thermoplastic vulcanizates (TPVs) are a specific group of thermoplastic elastomers (TPE) prepared by dynamic vulcanization of a thermoplastic and an elastomer, where the cross-linking of the elastomeric phase occurs during melt mixing with molten thermoplastics (Bhattacharya *et al.*, 2020). TPVs can be used in the manufacture of a variety of products and may be useful for making automotive interiors, such as instrument panels, floor consoles, and door panels.

High-density thermoplastic compounds are material have high specific gravity between 2 and 11 by adding high-density mineral filler or metal powder to the plastic matrix (Scheepens, 2016). Many high-density mineral fillers or metal powder are used to make high-density thermoplastic compounds like zinc, brass, silver, bismuth, molybdenum, solid tungsten steel, tungsten bismuth, and copper. Thermoplastic elastomer (TPE) compound with high specific gravity has a special market, such as for radiation shielding, projectiles, sporting goods, military, automotive,

packaging and noise attenuation. One of the applications of high-specific gravity material, that may be used in automotive, is for sound-proof floor mat.

Bismuth oxide is one of the high-density metal oxides with a density of 8.9 g/cc which is less than that of lead and greater than that of barite and steel and is nontoxic (Ambika *et al.*, 2017). Several researchers have studied bismuth oxide filled materials explored in various applications, such as for radiation shields (Abdalsalam *et al.*, 2020; Pavlenko *et al.*, 2019; Sheela *et al.*, 2019), biomedical applications (Jagdale *et al.*, 2021; Arnold *et al.*, 2021), electronic application (Elabbasy *et al.*, 2021; Singh *et al.*, 2019). However, there were no reports for automotive application. This paper aimed to develop high-density thermoplastic vulcanizates based on linear low-density polyethylene (LLDPE)/natural rubber (NR) blend with different proportions of bismuth oxide (Bi_2O_3) filler that meets with requirements for truck floor mats material.

MATERIALS AND METHODS

Materials

Linear low-density polyethylene (LLDPE) granules (extrusion grade, a density of 0.921 g/cm³, melt index of 1 g/10 min at 190 °C/2.16 kg) were purchased from a local supplier. Natural rubber (NR) grade SIR 20 was purchased from a local estate. The rubber ingredients used to make the rubber compounds in this research were the standard ones for the rubber industry consisted of cure activators (ZnO and stearic acid), N-Cyclohexylbenzothiazole-2-sulfenamide (CBS) as an accelerator, a filler (carbon black N330), a vulcanizing agent (sulfur), a processing oil (paraffinic oil), 2,2,4-Trimethyl-1,2-Dihydroquinoline (TMQ), N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine (6PPD), and N-isopropyl-N'-phenyl-p-phenylenediamine (IPPD) as the antioxidants, prevulcanization inhibitor (PVI). The rubber ingredients were 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. The high-density filler used in this research was a bismuth oxide (Bi₂O₃) with a density of 8.9 g/cm³.

Methods

High-density TPVs preparation

For the first step, the natural rubber was compounded with rubber ingredients by a two-roll mill for around 15 min. In this research, the composition of rubber compound consists of 100 phr of natural rubber, 5 phr of ZnO, 2 phr of stearic acid, 1 phr of TMQ, 1 phr of CBS, 1.5 phr of sulfur, 5 phr of paraffinic oil, 1 phr of 6PPD, 1 phr of IPPD, and 0.1 phr of PVI.

In the second step, the rubber compounds were blended with LLDPE, MA-g-PE, Irganox 1010, and Bi₂O₃ in a kneader at 130 °C, for 15 minutes, to make high-density TPVs. The composition of high-density TPVs consisted of 75 phr of LLDPE, 25 phr of a rubber compound, 5 phr of MA-g-PE, 0.1 phr of Irganox 1010, and a varied amount of Bi₂O₃ filler. Bi₂O₃ filler loading was varied by 50, 150, 200, 250, and 300 phr.

Characterization

The tensile strength, elongation at break, and tear strength of TPVs were done at room temperature (23±2 °C) with a Universal Testing Machine (UTM-Tinius Olsen H25K), before and

after aging. The tensile strength and elongation at break were tested according to ISO (2017a) using a dumbbell-type 2 specimen, an angle-type specimen used for tear strength testing according to ISO (2015). The Shore A hardness was measured according to ISO (2018a) using a Shore A durometer (Toyoseiki). Density was carried out using an electron densimeter (EW-200SG, Mirage) following ISO (2018b) Method A. All TPVs were also assessed for mechanical properties changes after thermal aging. The test was carried out by putting all the samples in an oven at 70 °C for 7 days, after which tensile strength, elongation at break, tear strength, and hardness (Shore A) tests were performed, following the same procedures and standard testing used on the samples before thermal aging.

The abrasion resistance test was done with a rotary drum abrasion tester (Bareiss) according to ISO (2017b) using the cylindrical shape specimens with a diameter of 16±0.2 mm and a height of 6 mm. The abrasion resistance test result was expressed as a relative volume loss compared to an abrasive sheet calibrated using a standard reference compound.

The flammability test was determined following ASTM (2018). TPVs samples, with dimensions of specimen size were 125±0.2 mm long, 13±0.2 mm wide, and thickness of 2±0.5 mm, were tested at a horizontal position by using a flame from a bunsen burner. During the test, time to ignition (TTI), and the time required to perform combustion of TPVs specimens as far as L (75 mm) were recorded. The burning rate was calculated from equation (1).

$$\text{Burning rate (mm/min.)} = \frac{60L}{t} \quad (1)$$

where:

L = the burned length, in millimetres = 75 mm

t = the burning time (sec.)

RESULTS AND DISCUSSION

Density Before and After Aging

High-density TPVs is a material that has a density exceeding about 2.0 g/cm³. The density of TPVs samples before and after aging are shown in Figure 1. The Bi₂O₃ filler had higher density than the other constituents of the formulation and the density increased by raising the Bi₂O₃ content in the composites, as observed in Figure 1. This

Figure 1. The density of TPVs at various Bi₂O₃ content, before and after thermal aging.

was due to the much larger densities of the Bi₂O₃ powders (8.9 g/cm³) compared to the density of NR and LLDPE. A similar trend, regarding high-density fillers, leading to an increase in composite density was reported by Poltabtim *et al.* (2018), Toyen *et al.* (2018), Al-Mattarneh and Dahim (2019), and Pavlenko *et al.* (2019).

Figure 1 shows the density of TPVs containing 50 phr of Bi₂O₃ filler is 1.26±0.016 g/cm³. When 300 phr of Bi₂O₃ filler was added to the TPVS, the density of TPVs increased to 2.82±0.032 g/cm³ (an increase of 123.81%). The density of TPVs before and after accelerated thermal aging at the temperature of 70 °C during 72 hours have range 1.26±0.013 to 2.82±0.0132 g/cm³ and 1.3±0.012 to 2.75±0.022 g/cm³, respectively. Accelerated thermal aging did not affect the density of TPVs, because the change in the average maximum density was 5.60% as indicated by the TPVs containing Bi₂O₃ filler up to 150 phr, while the change in the average density of TPVs composites containing Bi₂O₃ filler was above 150 phr, was not very significant, namely 2.48%. The density required for TPE-based trucks floor mats by one of the automotive companies is 2±0.2 g/cm³, and TPVs composite containing 250 phr of Bi₂O₃ filler can meet these requirements. However, the density of one of the commercial TPEs (1.75 g/cm³) for truck floor mats did not yet meet this requirement.

Hardness Before and After Aging

The hardness before and after aging of the obtained TPVs are presented in Figure 2. The results were depended on the content of Bi₂O₃.

Figure 2. The hardness of TPVs at various Bi₂O₃ content.

The increase of Bi₂O₃ filler loading significantly increased hardness due to the high rigidity of metal oxide powder in TPVs composite. A similar trend regarding high-density fillers loading resulted in the rise in hardness was reported by Mysiukiewicz *et al.* (2020) and El-Fiki *et al.* (2015). The hardness of the TPVs samples before aging was in the range of 90±1.225 – 96±1 Shore A. The hardness of thermal aged TPVs containing Bi₂O₃ fillers up to 250 phr was higher than that of the unaged. It can be indicated that the post vulcanization reaction of natural rubber compounds during the aging process increased hardness. Meanwhile, the hardness of thermally aged TPVs containing 300 phr of Bi₂O₃ filler was lower than unaged (decreased 2.48%). According to Ali and Pajarito (2018), the decrease in the hardness of the rubber composite due to thermal aging was caused by the degradation of the natural rubber main chain. The hardness range of the TPVs samples after aging was 92±1.581 – 95±1 Shore A.

The hardness required for TPE-based trucks floor mats by one of the automotive companies is 90±5 Shore A, and TPVs containing Bi₂O₃ filler up to 250 phr can meet these requirements. The hardness of one of the commercial TPEs (85 Shore A) for the same application meet this requirement.

Tensile Properties Before and After aging

The tensile properties of before and after aging TPVs as a function of Bi₂O₃ filler loading are displayed in Figure 3. Figure 3 shows that the tensile strength and elongation at break of the TPVs decreased with the increase of Bi₂O₃

Figure 3. Effect of Bi_2O_3 content on tensile strength (a) and elongation at break (b) for TPVs before and after thermal aging.

filler loading. The same trend was observed in tear strength values (Figure 4). The addition of high amounts of Bi_2O_3 filler particles can cause agglomeration of filler due to the interaction between fillers. The decrease in tensile strength may be due to the agglomeration of metal oxide particles which resulted in the weakening of the bond between the metal oxide particles and TPVs as a matrix. The decrease in elongation at break might be due to a decrease in ductility associated with the incorporation of an excess of metal oxide fillers into the polymer matrix. Such decreasing tensile strength and elongation at break with high load filler was also reported by Datta *et al.* (2016), Yuhaida *et al.* (2016), Onuoha *et al.* (2017), Poltabtim *et al.* (2018), and Lopattananon *et al.* (2019).

The highest tensile strength ($110 \pm 2.549 \text{ kg/cm}^2$) and elongation at break ($220 \pm 1.871\%$) value were achieved by TPVs containing the lowest Bi_2O_3 filler (50 phr). On the other hand, the highest Bi_2O_3 filler content (300 phr) resulted in the lowest tensile strength ($94.8 \pm 2.280 \text{ kg/cm}^2$) and elongation at break ($55.2 \pm 2.168\%$). An increase in Bi_2O_3 filler from 50 phr to 300 phr caused a decrease in tensile strength and elongation at break of 13.82% and 74.91%, respectively.

After accelerated thermal aging, the tensile strength and elongation at break of TPVs composites containing Bi_2O_3 filler tended to decrease. According to Park (2013), the decrease in tensile properties after thermal aging could be related to the increase in brittleness of specimens

driven by chain scission and degradation during the aging process. Elongation at break became lower after thermal aging due to the high elasticity of composites.

The test results for the tensile strength and elongation at break of commercial TPE for truck floor mats before aging were 23.7 kg/cm^2 and 23.3% , respectively, and after aging were 25.3 kg/cm^2 and 10.6% , respectively. These values were lower than the tensile strength and elongation at break values of all TPVs composites produced in this study.

Tear Strength Before and After Aging

Tear strength before and after aging of the obtained TPVs composites are presented in Figure 4. The tear strength of all samples were decreased with the increase in Bi_2O_3 loading. According to Wickramaarachchi *et al.* (2020), poor interaction between filler and matrix decreased the tear strength of composites.

The highest tear strength value before aging ($86 \pm 1.581 \text{ N/mm}$) was shown by TPVs containing Bi_2O_3 filler 50 phr. The addition of Bi_2O_3 of 250 phr and 300 phr gave a decrease in the tear strength of 8.37% and 31.16%, respectively, compared to TPVs composite containing 50 phr Bi_2O_3 filler. The highest tear strength value after aging ($85.6 \pm 1.516 \text{ N/mm}$) was obtained for TPVs containing Bi_2O_3 filler at 50 phr. The tear strength of the aged TPVs was lower than that of the non-aging. This might be attributed to the increase in brittleness of TPVs driven by chain scission and degradation during

Figure 4. Tear strength of TPVs at various Bi₂O₃ content, before and after thermal aging.

the aging process. Decreasing in tear strength after thermal aging was also reported by Li *et al.* (2015). The tear strength of one of the commercial TPE (27.3 N/mm) for truck floor mats is lower than the tear strength of all TPVs composites that resulted in this study.

Abrasion Resistance

The abrasion resistance of the TPVs expressed as volume loss is shown in Figure 5. An increase in volume loss means a decrease in abrasion resistance. The addition of Bi₂O₃ filler to TPVs decreases the abrasion resistance. The lowest volume lost value (25.8±0.837 mm³) was demonstrated by TPV containing 50 phr of Bi₂O₃ filler. The highest volume lost value (84.4±0.894 mm³) was observed by TPVs containing Bi₂O₃ filler at 200 phr. There was an 84.28% increase compared to volume loss of the TPVs containing 150 phr Bi₂O₃ filler (45.8±0.84 mm³). The increase in volume loss indicates a decrease in abrasion resistance. According to Mrówka *et al.* (2021), the decrease in abrasion resistance for higher filler content was due to the agglomeration of the filler. During abrasion, the filler agglomerate might detach from the sample, thus increasing

Figure 5. Volume loss of TPVs at various Bi₂O₃ content.

the volume loss. Volume loss of TPVs containing Bi₂O₃ filler at 50, 150, and 300 phr were lower than one of commercial TPE (76.09 mm³) for truck floor mats. However, TPVs containing 200 phr and 250 phr Bi₂O₃ filler were 10.92% and 8.81% higher, respectively, than volume loss of one of commercial TPE.

Flammability

Burning test is important for assessing the flammability properties of truck floor mats. According to Borowicz *et al.* (2019), time-to-ignition is one of the most important flame-retardancy parameters, because it determines how fast the material combustion will occur when exposed to a heat source. Additionally, according to ASTM (2018), it is necessary to record the time required to burn the TPVs specimen as far as L (75 mm) to calculate the burning rate (BR). In this research, the addition of metal oxide filler was expected to increase the composite burn resistance. The effect of Bi₂O₃ filler loading on the flammability properties of high-density TPVs are listed in Table 1.

Table 1 shows that increasing the Bi₂O₃ content gradually increases the time to ignition

Table 1. Burning rate for high-density TPVs.

Bi ₂ O ₃ filler content (phr)	time to ignition (sec)	Time to 75 mm (sec)	Burning rate (mm/min)
50	52	100	45.00
150	60	113	40.00
200	66	120	37.50
250	70	150	30.00
300	128	234	19.23

(TTI). The longer the TTI value, the probability of fire is lower (Borowicz *et al.*, 2019). As shown in Table 1, the ignition time of composite containing 50 phr Bi_2O_3 filler is 52 seconds, and an increase in Bi_2O_3 filler from 50 phr to 300 phr caused an increase in TTI of 148.15%. According to Jagdale *et al.* (2018), Bi_2O_3 is the filler that has high impact on the ignition time of the composite, and TTI of Bi_2O_3 (1-3%) reinforced composites was increased by 15–20 seconds. This may be due to the high thermal stability of the Bi_2O_3 filler. From Table 1, it is clear that at Bi_2O_3 filler loading of 300 phr observed the lowest value of burning rate (19.23 mm/min), which indicated the sample burned slowly. From the results, it is clear that the addition of Bi_2O_3 filler in the TPVs results in decreasing the rate of burning, and hence improves flammability resistance. Test results from one of the commercial TPE showed the burning rate was 66.67 mm/min. This value was higher than the burning rate from and all of TPVS composite resulted in this study.

CONCLUSIONS

High-density TPVs were prepared by adding different weight loads (50, 150, 200, 250, 300 phr) of bismuth oxide (Bi_2O_3) filler in linear low-density polyethylene (LLDPE)/natural rubber (NR) matrix by melt blending in a kneader. The effect of the amount of Bi_2O_3 filler on the physical and mechanical properties in the 75/25 LLDPE/NR blend was investigated. An increase in the weight load of bismuth oxide in TPVs increases density, hardness, and volume loss but reduces the tensile properties, tear strength, and burning rate of high-density TPVs. The results show that high-density thermoplastic vulcanizate containing 250 phr Bi_2O_3 is a promising candidate for use as a truck floor base. The properties of this high-density thermoplastic vulcanizate before aging were density $1.92 \pm 0.01 \text{ g/cm}^3$, hardness $94 \pm 0.707 \text{ Shore A}$, tensile strength $95.8 \pm 2.95 \text{ kg/cm}^2$, elongation at break $180 \pm 2.345\%$, tear strength $78.8 \pm 2.168 \text{ N/mm}$, volume loss $82.8 \pm 0.837 \text{ mm}^3$, burning rate 30 mm/min . Properties after aging were density $1.88 \pm 0.021 \text{ g/cm}^3$, hardness $95 \pm 0.707 \text{ Shore A}$, tensile strength $94.8 \pm 2.683 \text{ kg/cm}^2$, elongation at break $162 \pm 1.871\%$, and tear strength $77.2 \pm 1.643 \text{ N/mm}$.

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