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An Facile High-Density Polyethylene - Exfoliated Graphite - Aluminium Hydroxide Composite: Manufacture, Morphology, Structure, Antistatic and Fireproof Properties

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Graphite intercalation compounds (GIC) and exfoliated graphite (EG) as raw materials were prepared with flake graphite, concentrated sulphuric acid (H_2SO_4), potassium bichromate ($K_2Cr_2O_7$) and peracetic acid (CH_3CO_3H) and characterized. Then, high-density polyethylene-exfoliated graphite (HDPE-EG) composites were fabricated with HDPE and EG via in situ synthesis technique in the different mass ratio, and their resistivity values (ohms/sq) were measured. Based on the resistivity values, it was discovered that HDPE-EG composite with the antistatic property could be fabricated while the mass ratio was 5.00 : 0.30. Last, HDPE-EG-aluminium hydroxide (HDPE-EG-$Al(OH)_3$) composites were manufactured with HDPE, GIC and $Al(OH)_3$ via the in situ synthesis-thermal expansion technique, and their resistivity values and limiting oxygen index (LOI) values were measured. Based on the resistivity values and LOI values, it was discovered that HDPE-EG-$Al(OH)_3$ composite with the antistatic and fireproof property could be manufactured while HDPE, GICs and $Al(OH)_3$ of mass ratio was 5.00 : 0.30 : 1.00. Otherwise, the petal-like morphology and structure of HDPE-EG-$Al(OH)_3$ composite were characterized, which consisted of EG, HDPE and $Al(OH)_3$.

Keywords: graphite intercalation compounds, high-density polyethylene-exfoliated graphite composite, high-density polyethylene-exfoliated graphite -aluminium hydroxide composite, antistatic property, fireproof property.

1. INTRODUCTION

Polyethylene (PE) is an insulative and flammable macromolecule. In order to modify its electrical properties, so far polyethylene/polyamide composite coated with silver [1], polyethylene/graphite composites [2], carbon nanotubes filled polyethylene films [3], polyethylene//graphite/carbon fiber composites [4] and polyethylene terephthalate/graphene nanocomposites [5] have been manufactured. In order to modify its fireproof property, so far EVA/LDPE composites [6], polyethylene/fire retardants composites [7], LLDPE/EAA/MH composites [8], the composite of LDPE and zinc chelate complex containing both phosphorus and nitrogen [9], wood flour-polyethylene composites [10] and montmorillonite//magnesium hydroxide/HDPE composites [11] have been manufactured. However, it is not discovered that the electrical property and fireproof property on PE are modified synchronously.

GIC is a functional carbon material. Due to GIC can be exfoliated into EG, it is also known as expandable graphite [12 – 16]. EG is a loose porous carbonic material [17, 18]. So far it has been discovered that both GIC and EG may be used to modify the antistatic property [19 – 22] and thermal stability [23] of macromolecule. However, it is not discovered that both GIC and EG are not used to modify the electric property and fireproof property of HDPE synchronously.

Based on the above reports, it was considered that the electrical property and fireproof property on HDPE modified synchronously should be a new approach. Recently, the invention was finished in our laboratory, which included the oxidation and intercalation, the in situ synthesis and the thermal expansion presented in Fig. 1.

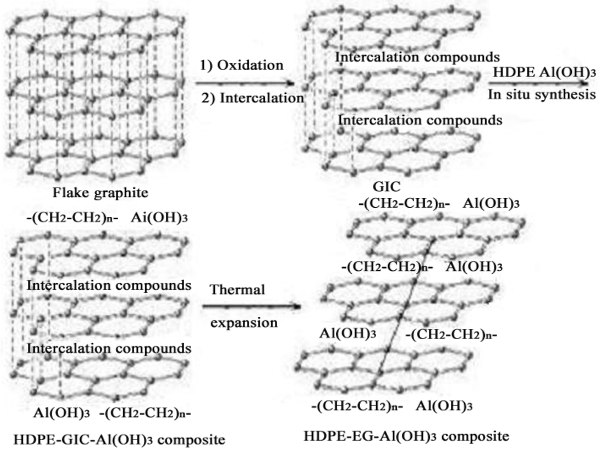

Fig. 1. Sketch map of HDPE-EG-$Al(OH)_3$ composite

In the first procedure, GIC was prepared with flake graphite, H_2SO_4, $K_2Cr_2O_7$ and CH_3CO_3H. In the second procedure, HDPE-GIC-$Al(OH)_3$ composite was fabricated with GIC, HDPE and $Al(OH)_3$ in toluene solvent. In the third procedure, HDPE-EG-$Al(OH)_3$ composite was manufactured with HDPE-GIC-$Al(OH)_3$ composite. Here it should be pointed out that GIC inside HDPE-GIC-$Al(OH)_3$ composite could be exfoliated into EG inside HDPE-EG-$Al(OH)_3$ composite, which could modify the electrical property and decrease the resistivity value. Based on the cogitation, HDPE-EG composites were firstly fabricated with HDPE and EG via the in situ synthesis technique

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before manufacturing HDPE-EG-Al(OH)$_3$ composite, and then the resistivity values of HDPE-EG composites were investigated. On this basis, GIC dosage in HDPE, GIC and Al(OH)$_3$ of mass ratio was determined in manufacturing HDPE-EG-Al(OH)$_3$ composite via the in situ synthesis-thermal expansion technique, which had the antistatic property.

2. EXPERIMENTAL

2.1. Materials

Flake graphite (diameter ≥ 320 μm) was supplied by Graphite Products Co. Ltd. (Baoding, China). Analytical grade concentrated sulphuric acid (H_2SO_4), potassium bichromate ($K_2Cr_2O_7$) and peracetic acid (CH_3CO_3H) were purchased from Hong Guang Chemicals Co., Ltd. (Beijing, China).

2.2. Preparations of GIC and EG

In the light of 1 : 0.13 : 1.84 : 0.50 of mass ratio, flake graphite, $K_2Cr_2O_7$, H_2SO_4 and CH_3CO_3H were added in a three-neck round bottom flask, and then the flask thermometer, muddler and round condenser were apart fixed on three necks. Subsequently, the reactants were continually stirred in 45 ºC of constant temperature bath. After 60 min of stirring, the resulting GIC was washed repeatedly until the neutrality (pH = 7). Last, the resulting GIC was dried at 50 ºC – 60 ºC. After 30 min of drying, GIC could be obtained.

GIC was added in an evaporating dish. Then, the evaporating dish was placed into Muffle, which had 200 ºC of furnace temperature. After 0.5 min of heating, the evaporating dish was taken out and placed until room temperature. Last, EG could be obtained.

2.3. Characterization of GIC and EG

In the light of the previous works [12 – 16], the characterization of GIC generally included morphology, structure and intercalation compounds, and the characterization of EG generally came down to morphology and structure. The morphology and structure of GIC and EG were characterized with SEM micrographs of flake graphite, GIC and EG, which were obtained from a scanning electron microscope (S-570, Japan); meanwhile, the intercalation compounds inside GIC were characterized with the chemical method, UV-visible spectrometer (Beijing, China) and Acidometer (Beijing, China). The characterization process of the intercalation compounds came down to the following procedures. Firstly, 1.00 g of GIC was soaked in 100.00 ml of distilled water. After 24 h of soaking, the aqueous solution was separated into three shares. Secondly, one share was used to detect SO_4^{2-} in the aqueous solution ($BaCl_2$ aqueous solution was added into the aqueous solution, and the solution was vibrated, and whether a precipitate formation was observed.); another share was used to detect CH_3CO_2H in the aqueous solution (the aqueous solution was added in a colorimetric ware, and the aqueous solution absorbances were determined under the different wavelengths, and the correlation curve of absorbance and wavelength was drawn); third share was used to detect the acidity of the aqueous solution (The aqueous solution was put into 50 ml of beaker, and the pH value of the aqueous solution was determined.).

2.4. Fabrication of HDPE-EG composite via in situ synthesis technique

5.0 g of HDPE and a certain amount of EG were added in a flask filled with toluene solvent, and then the flask was fixed on an electromagnetic stirrer. After 60 min of stirring, HDPE and EG were dispersed completely in the toluene solvent at 100 °C. Then, the toluene solvent was separated out via the distillation method, and the resulting HDPE-EG composite was washed with the hot water and dried in a desiccation box at 60 °C. After 1 h of drying, HDPE-EG composite could be obtained.

2.5. Manufacture of HDPE-EG-Al(OH)$_3$ composite via in situ synthesis-thermal expansion technique

5.0 g of HDPE, 0.3 g of GIC and a certain amount of Al(OH)$_3$ were added in a flask filled with toluene solvent, and then the flask was fixed on an electromagnetic stirrer. After 60 min of stirring, HDPE, GIC and Al(OH)$_3$ would be dispersed completely in the toluene solvent at 100 °C. Then, the toluene solvent was separated out through the distillation method, and the resulting HDPE-GIC-Al(OH)$_3$ composite in the flask was washed with the hot water and dried in a desiccation box at 60 °C. After 1 h of drying, HDPE-GIC-Al(OH)$_3$ composite could be synthesized.

HDPE-GIC-Al(OH)$_3$ composite was added in an evaporating dish. Then, the evaporating dish was placed into Muffle, which had 200 °C of furnace temperature. After 1.00 min of heating, the evaporating dish was taken out and placed until room temperature. Last, HDPE-EG--Al(OH)$_3$ composite could be obtained.

2.6. Characterization of HDPE-EG composite and HDPE-EG-Al(OH)$_3$ composite

The specimen slices of HDPE, HDPE-EG composite and HDPE-EG-Al(OH)$_3$ composite were manufactured with Punch Machine TDP-5 (made in China). The thickness of specimen slice was 0.30 cm (l), and the area of specimen slice was 100.00 cm^2. The resistance values (ohms⁄sq) of HDPE, HDPE-EG composite and HDPE-EG--Al(OH)$_3$ composite were measured under the same environmental temperature and relative humidity conditions with an ACL Staticide (model 800, made in USA). LOI values of HDPE, HDPE-EG composite and HDPE-EG-Al(OH)$_3$ composite were measured with Oxygen Index Tester (made in China). FTIR spectra of HDPE-EG composite and HDPE-EG-Al(OH)$_3$ composite were recorded at room temperature with Perkin Elmer Spectrum GX spectrophotometer. In the manufacture of FTIR specimens, HDPE-EG composite was firstly mixed with potassium bromide, and then HDPE-EG composite and potassium bromide were rubbed together to the powder. Last, the powder of HDPE-EG composite and potassium bromide was pressured to thin slices as the specimen of HDPE-EG composite. In the light of the same procedure, the specimen of HDPE-EG-Al(OH)$_3$ composite was manufactured. SEM micrographs of HDPE-EG composite and HDPE-EG-$Al(OH)_3$ composite were obtained by a scanning electron microscope (S-570, Japan).

3. RESULTS AND DISCUSSION

3.1. Confirmation of GIC and EG as raw materials

Fig. 2 shows SEM micrographs of flake graphite (a), GIC (b) and EG (c). To compare Fig. 2, a, with 2, b, it is discovered that flake graphite and GIC have the flake morphology, and there is the multiple graphene structure inside them, and the arrangement of graphene sheets inside GIC is looser than the one inside flake graphite. These indicate that the multiple graphene structure inside flake graphite is the same as the one inside GIC, and the interlayer distances between the graphene sheet and the graphene sheet inside flake graphite are smaller than the ones inside GIC. Based on the facts, it is considered that GIC should possess the multiple graphene structure, and there are the spaces between the graphene sheet and the graphene sheet, which can intercalate some compounds. Otherwise, the characterization results for the aqueous solution show that the white deposit ($BaSO_4$) generates in the aqueous solution, and the top absorption peak of the aqueous solution appears at 204.0 nm of wavelength, and the pH value of the aqueous solution is 1.12. In the light of $Ba^{2+} + SO_4^{2-} = BaSO_4$ (white precipitate) and the correlation of the absorptive wavelength and the function group and the correlation of H^+ concentration and the pH value, it is considered that there might be CH_3CO_2H and H_2SO_4 in the aqueous solution, which originate from GIC. To colligate the above conclusions, it is considered that GIC has been prepared, which possesses the flake morphology, the multiple graphene structure and CH_3CO_2H and H_2SO_4 as the intercalation compounds inside.

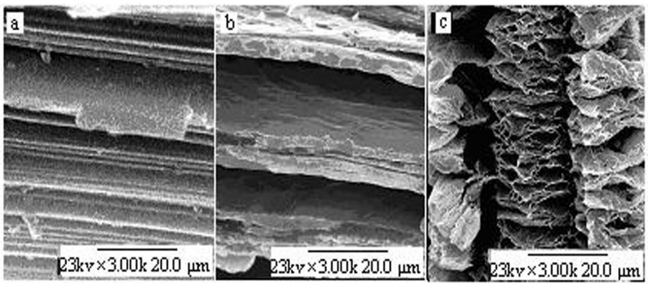

Fig. 2. SEM micrographs of flake graphite (a), GIC (b) and EG (c)

From Fig. 2, b, and 2, c, it is discovered that EG has the wormlike morphology, and the multiple graphene structure of GIC is the same as the one of EG, but the interlayer distances between the graphene sheet and the graphene sheet inside GIC are smaller than the ones inside EG. Based on the facts, it is considered that GIC has been exfoliated into EG at 200 ºC. For the exfoliation reason, it is considered that it may ascribed to the vaporization and combustion of CH_3CO_2H and H_2SO_4 inside GIC, and the interlayer distances between the graphene sheet and the graphene sheet are further extended under the expansion force generated by the vaporization and combustion of CH_3CO_2H and H_2SO_4. To colligate the above conclusions, it is considered that EG has been prepared, which possesses the wormlike morphology and the multiple graphene structure extended further.

3.2. Morphologies of HDPE-EG composite and HDPE-EG-$Al(OH)_3$ composite

Fig. 3 shows SEM micrographs of HDPE-EG composite and HDPE-EG-$Al(OH)_3$ composite. To observe them contrastively, it is discovered that HDPE-EG composite has the planar morphology, and HDPE-EG-$Al(OH)_3$ composite possesses the petal-like morphology; meanwhile, it is also discovered that there are not any pores on the surface of HDPE-EG composite, and there are many pores on the surface of HDPE-EG-$Al(OH)_3$ composite.

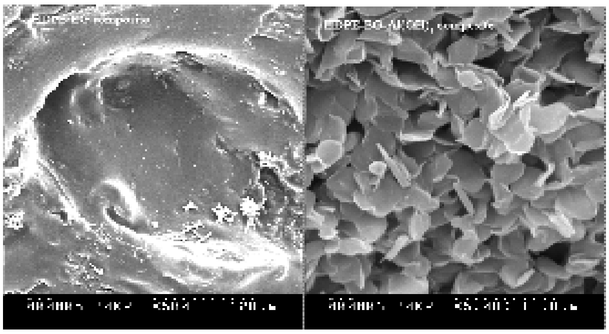

Fig. 3. SEM micrographs of flake graphite (a), GIC (b) and EG (c)

In view of HDPE-EG composite and HDPE-EG-$Al(OH)_3$ composite fabricated apart via the in situ synthesis technique and the in situ synthesis-thermal expansion technique, it is considered that the differences of morphology and structure may ascribe to the differences of two techniques. In the in situ synthesis technique, HDPE-EG composite is fabricated by HDPE and EG in the toluene solvent, and the multiple graphene structure extended further inside EG may be filled by HDPE dissolved in the toluene solvent, and HDPE may be solidified in the multiple graphene structure after the removal of toluene solvent. In the in situ synthesis-thermal expansion technique, HDPE-EG-$Al(OH)_3$ composite is manufactured by HDPE-GIC-$Al(OH)_3$ composite, and GIC covered by HDPE and $Al(OH)_3$ may be exfoliated into EG of which the multiple graphene structure is extended further, and HDPE and $Al(OH)_3$ may be solidified in the multiple graphene structure exfoliated after the thermal expansion. These namely cause that HDPE-EG composite has the planar morphology, and HDPE-EG-$Al(OH)_3$ composite possesses the petal-like morphology, and there are not any pores on the surface of HDPE-EG composite and many pores on the surface of HDPE-EG-$Al(OH)_3$ composite.

3.3. Structures of HDPE-EG composite and HDPE-EG-$Al(OH)_3$ composite

Fig. 4 exhibits FTIR spectra of HDPE-EG composite and HDPE-EG-$Al(OH)_3$ composite. According to the previous work [22], it is concluded that HDPE-EG composite is structured by HDPE and EG. To analyze

FTIR spectra contrastively, it is discovered that there are common absorption bands in FTIR spectra of HDPE-EG composite and HDPE-EG-Al(OH)$_3$ composite. These indicate that there also are HDPE and EG inside HDPE-EG-Al(OH)$_3$ composite. Otherwise, it is also discovered that there are two percent transmittances in FTIR spectra of HDPE-EG composite and HDPE-EG-Al(OH)$_3$ composite, which are apart 3.377 and 3.208 and 14.391 and 13.241. These indicate that there is a plurality of hydroxyl inside HDPE-EG-Al(OH)$_3$ composite, which can absorb 2918 cm^{-1} and 2850 cm^{-1} of infrared light. Based on the analyses, it is concluded that HDPE-EG-Al(OH)$_3$ composite should be structured by HDPE, EG and Al(OH)$_3$.

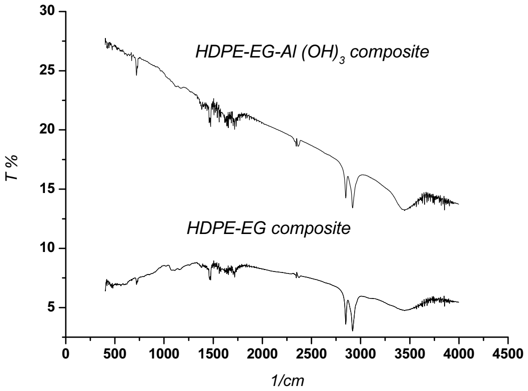

Fig. 4. FTIR spectra of HDPE-EG composite and HDPE-EG--Al(OH)$_3$ composite

3.4. Influence of mass ratio on antistatic property of HDPE-EG composite

Table 1. Influence of mass ratio on resistance value of HDPE-EG composite

Mass ratio (m HDPE : :m EG)	Resistivity value (ohms/sq)	Relative humidity (%)	Temperature (ºC)
5.00 : 0.00	1.1×10^{17}	44.40	25.80
5.00 : 0.10	1.0×10^{12}	44.40	25.80
5.00 : 0.20	1.7×10^{11}	44.40	25.80
5.00 : 0.30	6.1×10^{6}	44.40	25.80
5.00 : 0.40	2.1×10^{5}	44.40	25.80
5.00 : 0.50	1.9×10^{5}	44.40	25.80

As shown in Table 1, when the relative humidity and the environmental temperature are the same, the resistance value of HDPE is 1.1×10^{17} ohms/sq, and the resistance values of HDPE-EG composites will decrease from 1.0×10^{12} ohms/sq to 1.9×10^{5} ohms/sq when the mass ratios increase from 5.0 : 0.1 to 5.0 : 0.5, and there is a value of the mutation (6.1×10^{6} ohms/sq) at 5.0 : 0.3 of mass ratio. These results may be explained from following three aspects. Firstly, HDPE is a granular solid with 1.1×10^{17} ohms of resistance value. When the granular HDPE is compressed into HDPE sample slice, the conductive capacity of HDPE will not be changed without doubt. This namely causes that the resistance value of HDPE sample slice should be for 1.1×10^{17} ohms/sq. Secondly, EG is a loose and porous solid with 1.0×10^{3} ohms/sq of resistance value. When EG and HDPE mixed homogeneously are compressed into the sample slices of HDPE-EG composites, the network structure will be formed beyond all doubt. In the light of the results reported previously [23 – 26], EG may decrease the resistance values of EG-macromolecule composites via forming the conductive network inside EG-macromolecule composites. Thus, it is guessed that EG should be a conductive additive. In the syntheses of HDPE-EG composites, all of HDPE dosages in the mass ratios are 5.00 g. This namely implies that the resistance value of HDPE-EG composite will decrease along with the increase of EG dosage. Therefore, this namely causes that the resistance values of HDPE-EG composites will decrease from 1.0×10^{12} ohms/sq to 1.9×10^{5} ohms/sq when the mass ratios increase from 5.0 : 0.1 to 5.0 : 0.5. Thirdly, EG as the conductive additive may undoubtedly enhance the conductive capacity of HDPE-EG composite. This namely implies that the conductive value of HDPE-EG composite will be also enhanced along with the increase of EG dosage. However, there is a value of the mutation known as the percolation threshold in the conductive values of HDPE-EG composites. In view of the conductive value of HDPE-EG composite varies linearly with the reciprocal of the resistance value of HDPE-EG composite, it is considered that 6.1×10^{6} ohms/sq of resistance value should be the percolation threshold. Thereby, this namely causes that there is a value of the mutation (6.1×10^{6} ohms/sq) at 5.0 : 0.3 of mass ratio.

In addition, the data in Table 1 also show that the resistance values of HDPE-EG composites will be 6.1×10^{6} ohms⁄sq while the mass ratio is 5.0 : 0.3. According to the antistatic principle, a solid substance will possess the antistatic property while the resistance value is between 1.0×10^{5} ohms/sq and 1.0×10^{8} ohms/sq. It is judged that HDPE-EG composites should have the antistatic property when the mass ratio is 5.0 : 0.3.

3.5. Influence of mass ratio on antistatic property and fireproof property of HDPE-EG-Al(OH)$_3$ composite

Table 2. Influence of mass ratio on resistance value and LOI value of HDPE-EG-Al(OH)$_3$ composite

Mass ratio (mHDPE:mGIC: :mAl(OH)3)	Resistivity value (ohms/sq)	Relative humidity (%)	Temperature (ºC)	LOI
5.00 : 0.00 : 0.00	1.1×10^{17}	44.40	25.80	17.40
5.00 : 0.30 : 0.00	6.1×10^{6}	44.40	25.80	21.00
5.00 : 0.30 : 1.00	1.7×10^{7}	44.40	25.80	28.10
5.00 : 0.30 : 2.00	6.1×10^{9}	44.40	25.80	29.30
5.00 : 0.30 : 3.00	2.1×10^{10}	44.40	25.80	30.20
5.00 : 0.30 : 4.00	1.9×10^{11}	44.40	25.80	31.10

As shown in Table 2, when the relative humidity and the environmental temperature are the same, the resistance value of HDPE is 1.1×10^{17} ohms/sq, and the resistance value of HDPE-EG composite is 6.1×10^{6} ohms/sq, and the resistance values of HDPE-EG-Al(OH)$_3$ composites will increase from 1.7×10^7 ohms/sq to 1.9×10^{11} ohms/sq while the mass ratios enhance from 5.0 : 0.3 : 1.0 to 5.0 : 0.3 : 4.0. These indicate that EG exfoliated by GIC inside HDPE-EG-Al(OH)$_3$ composite can play the role of conductive additive like EG inside HDPE-EG composite, and the action of Al(OH)$_3$ is just opposite to the one of EG for modifying the electric property of HDPE. Based on the cogitation, it is considered that the resistance values of HDPE-EG-Al(OH)$_3$ composites should increase from 1.7×10^7 ohms/sq to 1.9×10^{11} ohms/sq while the mass ratios enhance from 5.0 : 0.3 : 1.0 to 5.0 : 0.3 : 4.0.

Otherwise, Table 2 exhibits that LOI value of HDPE is 17.4, and LOI value of HDPE-EG composite is 21.0, and LOI values of HDPE-EG-Al(OH)$_3$ composites will increase from 28.1 to 31.1 while the mass ratios enhance from 5.0 : 0.3 : 1.0 to 5.0 : 0.3 : 4.0. For these results, it is considered that they should have relation to the fireproof mechanisms of EG and Al(OH)$_3$. Firstly, EG is a kind of carbon materials, which originates from graphite and has the fireproof capacity, and the specific heat capacity of EG is bigger than the one of HDPE. When HDPE-EG composite combusts, EG will store and transfers the heat energy so that HDPE is not easy to attain the decomposable temperature; otherwise, EG will also combust, and the carbonic layer formed after EG combustion will intermit the combustion of HDPE through blocking up the heat energy and oxygen. These namely cause that LOI value of HDPE-EG composite (21.0) is bigger than the one of HDPE (17.4). Secondly, Al(OH)$_3$ can be decomposed into Al_2O_3 and H_2O in the process of calcination. When HDPE-EG-Al(OH)$_3$ composite combusts, Al(OH)$_3$ will imbibe a lot of heat energy to slower the calefactive and intermit the decomposition of HDPE; otherwise, Al(OH)$_3$ will also release the vapor to dilute the flammable gas. These namely cause that LOI values of HDPE-EG-Al(OH)$_3$ composites should increase from 28.1 to 31.1 while the mass ratios enhance from 5.0 : 0.3 : 1.0 to 5.0 : 0.3 : 4.0.

To calculate the antistatic property and the fireproof property synthetically, it is considered that HDPE-EG--Al(OH)$_3$ composite should have the resistance value between 1.0×10^5 ohms/sq and 1.0×10^8 ohms/sq and the bigger LOI value. To refer to the resistance values and LOI values in Table 2, it is discovered that the resistance value and LOI value of HDPE-EG Al(OH)$_3$ composite will be 1.7×10^7 ohms/sq and 28.1 while the mass ratio is 5.0 : 0.3 : 1.0. These namely imply that HDPE-EG Al(OH)$_3$ composite should have the antistatic property and the fireproof property synchronously while the mass ratio is for 5.0 : 0.3 : 1.0.

4. CONCLUSIONS

HDPE-EG composite with the antistatic property could be fabricated via the in situ synthesis technique while HDPE and EG of mass ratio was 5.00 : 0.30, which possessed the planar morphology and consisted of HDPE and EG. Otherwise, HDPE-EG-Al(OH)$_3$ composite with the antistatic property and the fireproof property could be manufactured via the in situ synthesis-thermal expansion technique while HDPE, GIC and Al(OH)$_3$ of mass ratio was 5.00 : 0.30 : 1.00, which possessed the petal-like morphology and consisted of HDPE, EG and Al(OH)$_3$.

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