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RESEARCH ARTICLE

Electron beam irradiated HDPE/EVA/$Mg(OH)_2$ composites for flame-retardant electric cables

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Abstract The mechanical properties and flammability of high-density polyethylene (HDPE)/ethylene vinyl acetate (EVA) mixed with various amounts of magnesium hydroxide ($Mg(OH)_2$) as the filler in composites, irradiated with electron beam at an irradiation dose of 150 kGy, have been studied. It is found that high-energy electron beam irradiation has significant effects on the mechanical properties of the HDPE/EVA/$Mg(OH)_2$ composites. The tensile strength and elastic modulus increased greater than in the unirradiated ones. Meanwhile, with increasing the content of $Mg(OH)_2$ in the composites, the limiting oxygen index (LOI) value increased sharply. The microstructure of the caves of the unirradiated HDPE/EVA/$Mg(OH)_2$ composites show poor interface of composites compared with the irradiated ones, as observed in SEM micrographs.

Keywords electron beam irradiation, cross-linking, mechanical properties, $Mg(OH)_2$, HDPE/EVA

1 Introduction

The initiative of producing highly filled polyethylene (PE)/magnesium hydroxide ($Mg(OH)_2$) composite is to fabricate flame retardant electric cables in replacing chlorine-containing polymers, which are believed to produce large amounts of corrosive and toxic gases and smoke when they are exposed to fire. However, the use of large amounts of inorganic filler, up to 60% of $Mg(OH)_2$ in composite for obtaining the required flame resistance, drastically decreases some important mechanical properties of the material. Thus, obtaining a highly filled PE/$Mg(OH)_2$ composite with stable mechanical properties is the primary goal of this research.

Mechanical properties of polymer composites can be altered by various factors: properties of the polymer matrix, filler particle size and shape, distribution of the filler, interfacial adhesion between filler and matrix, etc. Many investigations have been reported on the irradiated polymers and polymer composites and its effect on chemical structure and its physical properties [1–7]. High-energy electron beam irradiation on polymers has been carried out in a wide range of application fields, for example, the production of heat shrinkable polyethylene films, tubes and hot water piping installation, wire and cable industries [5,8–11]. Radiation cross-linking of HDPE/EVA and LDPE/EVA can improve the blends' mechanical properties and thermal stability [12,13].

This paper reports the effect of high-energy electron beam irradiation on the mechanical properties of flame-retardant HDPE/EVA/$Mg(OH)_2$ composites with various $Mg(OH)_2$ contents. Scanning electron microscopy was used to investigate the morphology of the composites before and after irradiation.

2 Experimental

2.1 Materials

High-density polyethylene (HDPE, 5502#, MFR = 0.35 g/10 min, density = 0.955 g/cm^3) was obtained from Daelim, Korea. Ethylene vinyl-acetate copolymer (EVA, 8450#, VA = 15 wt.%, MFR = 1.5 g/10 min, density = 0.940 g/cm^3) was obtained from Nippon Unicar. Co. Ltd., Japan. The flame retardant filler magnesium hydroxide ($Mg(OH)_2$, average particle size = 2 μm) was obtained from the Dalian Yatai Science and Technology New Material Co. Ltd., China.

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2.2 Sample preparation and irradiation

HDPE/EVA/Mg(OH)$_2$ composites were prepared via melt compounding at 160°C in a ThermoHaake rheomixer with a rotation speed of 60 rpm, and the mixing time was 6 min for each sample. The mixed samples were transferred to a mold and preheated at 180°C for 15 min, then pressed at 20 MPa and then successively cooled to room temperature while maintaining the pressure to obtain the composites sheets for further measurements. Before mixing, all the components were dried in a vacuum oven at 80°C for at least 12 h. The pressed samples were irradiated by the high-energy electron beam at the same irradiation dose (150 kGy) in air at room temperature. The content of Mg(OH)$_2$ in the HDPE/EVA/Mg(OH)$_2$ composites was changed from 10% to 70%.

2.3 Measurements

2.3.1 Mechanical properties

The tensile strength (TS), elongation at break (EB) and elastic modulus were measured with a RGM-type electronic testing machine (made by Shenzhen Reger Instrument Co. Ltd., China) at a crosshead speed of 50 mm/min according to ASTM D638.

2.3.2 LOI test

The LOI measurements were carried out using a HC-2 type instrument (Nanjing Analytical Instrument Factory, China) on the sheets of 120 mm × 6.5 mm × 3 mm in dimensions according to ASTM D2863-77.

2.3.3 Scanning electron microscopy (SEM)

The SEM micrographs of samples were observed by JEOL JSM-5510 scanning electron microscope. The samples were chosen after the tensile test. The content of Mg(OH)$_2$ in the HDPE/EVA/Mg(OH)$_2$ composites was 40 wt.%. The surface of the irradiated and unirradiated samples was coated with a thin layer of gold to avoid electrostatic charging during examination.

3 Results and discussion

3.1 Mechanical properties

As seen from Fig. 1, with increasing the content of Mg(OH)$_2$ in the composites, the TS of the unirradiated samples decreased sharply from 15.19 to 8.93 MPa. For the irradiated ones, TS also decreased greatly from 19.34 to 9.9 MPa. Comparing the TS of the irradiated samples with that of the unirradiated ones, the improvement of

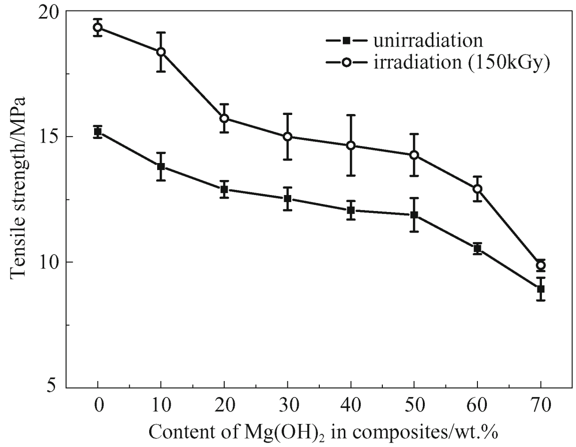

Fig. 1 Tensile strength versus the Mg(OH)$_2$ content in composites

the TS was around 22% for each sample and the maximum increase was 33% for the 10% content of Mg(OH)$_2$ in the composites.

The elongation at break shows the flexibility property of the test samples. From Fig. 2, the elongation at break (EB) of the unirradiated samples decreased greatly from 1472% to 10%, because of the increasing amount of Mg(OH)$_2$ in the composites. The tendency of the curve was the same with that of irradiated ones from 658% to 8%. It can also be seen from Fig. 2 that the EB of unirradiated samples were much higher than those of irradiation ones, when the content of Mg(OH)$_2$ in the composites was less than 30%.

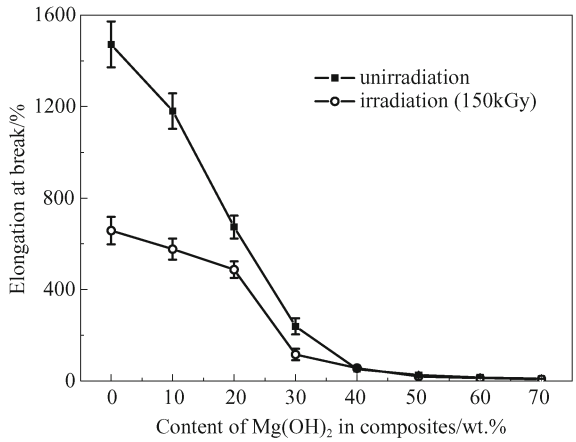

Fig. 2 Elongation at break versus the Mg(OH)$_2$ content in composites

From Fig. 3, with increasing the content of Mg(OH)$_2$ in the composites, the elastic modulus of the irradiated samples and unirradiated ones increases greatly. It can also be seen from Fig. 3 that elastic modulus of irradiated samples were much higher than those of unirradiated ones and the content of Mg(OH)$_2$ was 60%. Therefore, the TS and elastic modulus of HDPE/EVA/Mg(OH)$_2$ composites are

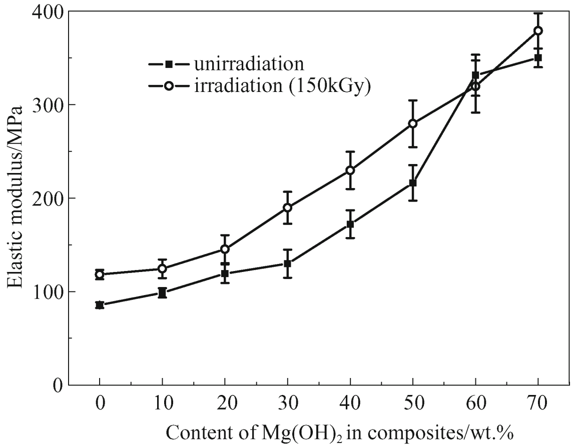

Fig. 3 Elastic modulus versus the $Mg(OH)_2$ content in composites

improved by the high-energy electron beam irradiation, which are attributed to the increase in cross-linking density.

3.2 LOI test

LOI is defined as the minimum fraction of oxygen in an oxygen-nitrogen mixture that is just sufficient to sustain combustion of the specimen after ignition. Figure 4 shows the LOI test results of HDPE/EVA/$Mg(OH)_2$ composites with increasing the content of $Mg(OH)_2$ in the composites. It can be seen that both the LOI value of irradiated samples and unirradiated ones increased along with the content of $Mg(OH)_2$ in the composites. It can also be seen from Fig. 4 that the LOI value of irradiated samples are higher than those of unirradiation ones. With increasing the content of the $Mg(OH)_2$ in the composites, the tendency of the irradiated samples are much pronounced. For example, the maximum LOI value was 42.5 for the 70% content of $Mg(OH)_2$ in irradiated composites.

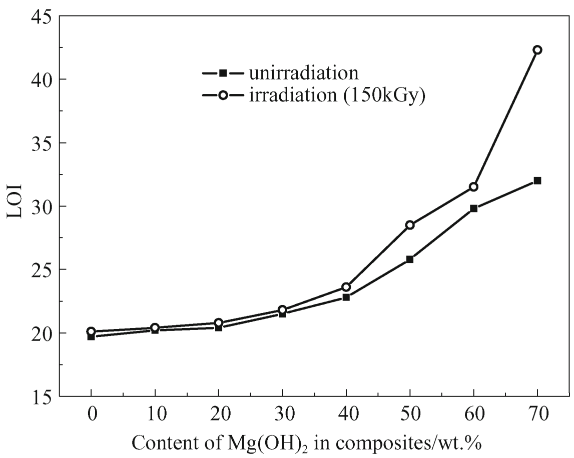

Fig. 4 LOI values versus the $Mg(OH)_2$ content in composites

3.3 Microstructures

Figures 5 and 6 show the SEM micrographs of the surfaces of irradiated and unirradiated HDPE/EVA/$Mg(OH)_2$ (40 wt.%) composites, respectively. From Fig. 6, it can be seen (shown by the white arrows) that there were many caves on the surface of the unirradiated HDPE/EVA/$Mg(OH)_2$ (40 wt.%), which could not be seen in Fig. 5. Most of the caves appeared beside the interface between the filler and the matrix. It is the structure of these caves that give poor mechanical properties of the unirradiated samples. Meanwhile, it can be proved that the high-energy beam irradiation increased the cross-linking degree of the composites, which modified the interface of the composites and enhanced the interaction of the filler.

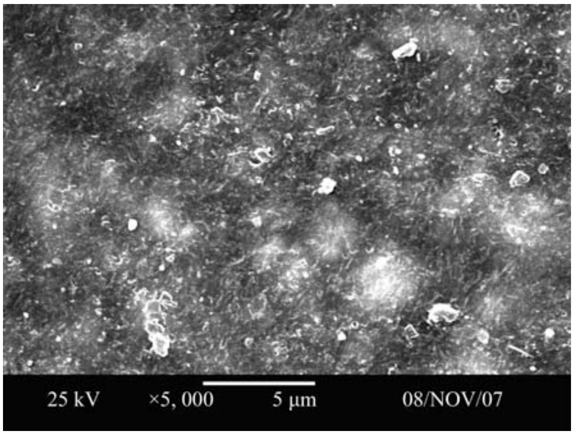

Fig. 5 SEM image of the surface of irradiated HDPE/EVA/$Mg(OH)_2$ composites (40 wt.%)

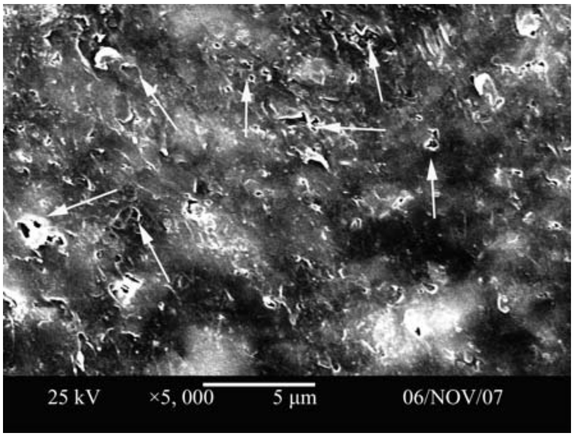

Fig. 6 SEM image of the surface of unirradiated HDPE/EVA/$Mg(OH)_2$ composites (40 wt.%)

4 Conclusions

(1) With increasing the content of $Mg(OH)_2$ in HDPE/EVA/$Mg(OH)_2$ composites, the tensile strength and the

elongation at break decease, whereas the elastic modulus increases greatly.

(2) The high-energy electron beam irradiation has much effect on the mechanical properties of the HDPE/EVA/$Mg(OH)_2$ composites. Both the tensile strength and the elastic modulus increase greatly than in the unirradiated samples.

(3) The LOI value of irradiated samples are higher than those of unirradiation ones. With increasing the content of $Mg(OH)_2$ in the composites, the tendency of the irradiated samples are much pronounced.

(4) The structure of the caves of the unirradiated HDPE/EVA/$Mg(OH)_2$ composites give poor mechanical properties. The high-energy beam irradiation increased the cross-linking degree of the composites, which modifies the interface of the composites and enhances the interaction of the filler.

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