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Gamma irradiation induced effects of butyl rubber based damping material

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ABSTRACT

The effects of gamma irradiation on the butyl rubber based damping material (BRP) at various doses in nitrogen were investigated in this study. The results show that irradiation leads to radiolysis of BRP, with extractives increasing from 14.9 ± 0.8% of control to 37.2 ± 1.2% of sample irradiated at 350 kGy, while the swelling ratio increasing from 294 ± 3% to 766 ± 4%. The further investigation of the extractives with FTIR shows that the newly generated extractives are organic compounds containing C-H and C=C bonds, with molecular weight ranging from 26,500 to 46,300. SEM characterization shows smoother surface with holes disappearing with increasing absorbed doses, consistent with "softer" material because of radiolysis. Dynamic mechanical study of BRP show that tan δ first slightly then obviously increases with increasing absorbed dose, while storage modulus slightly decreases. The tensile testing shows that the tensile strength decreases while the elongation at break increases with increasing dose. The positron annihilation lifetime spectroscopy show no obvious relations between free volume parameters and the damping properties, indicating the complicated influencing factors of damping properties.

1. Introduction

Butyl rubber (BR) is an isobutylene/isoprene copolymer with good properties, such as good thermal stability, good moisture and chemical resistance, and low gas permeability (Brydson, 1988; Karaağaç et al., 2007). It has been widely used as tyre inner tubes, tyre inner liners, tyre curing bladders and stretchable electronics (Binglin et al., 1993; Vohra et al., 2016).

In this study, a melt-blended copolymer (BRP) of BR and phenolic resin (PR) was introduced (Qu et al., 2007; Dutta and Tripathy, 1990). Due to the excellent damping property, BRP was utilized as damping material to absorb shock in equipment. Thus there is a possibility, BRP may undergo chemical changes when the equipment exposed to gamma rays, say, in aerospace and nuclear power plant applications.

The previous study suggested a major chain scission reaction accompanied with significant reduction in molecular weight of BR when exposed to ionizing radiation (Telnov et al., 2002; Hill et al., 1992; Scagliusi et al., 2012; Chandra et al., 1982; Scagliusi et al., 2017). Thus high energy radiation was used to recycle scrap BR for its economical and ecological advantages compared with the conventional chemical, thermal and mechanical methods (Binglin et al., 1993; Zaharescu et al., 2001; Burillo et al., 2002). Veli Deniz reported the recycling of butyl rubber-containing inner tubes with gamma irradiation of 120 kGy (Karaağaç et al., 2007).

Since BR would undergo chemical scission when exposed to gamma rays, leading to the decrease of molecular weight and mechanical properties, BRP probably also undergo radiolysis reaction. The effect of absorbed dose on the chemical structure and properties of BRP is investigated in this study. ^{60}Co was used as the high-energy irradiation source to evaluate radiation effects of BRP. The structure, morphology and mechanical properties were studied.

2. Experimental section

2.1. Materials

The damping material used in this study was kindly supplied by Aerospace Research Institute of Materials and Processing Technology, China (Brand name: ZN-1). It was fabricated by melt-blending of BR and PR, where PR is vulcanizing agent (Qu et al., 2007; Chinese, 2001).

2.2. Methods

The dog-bone shaped and rectangular samples were sealed in glass bottle filled with nitrogen when irradiated. Irradiation of the samples was carried out at ambient temperature using a ^{60}Co source at Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics, China. The samples were irradiated at dose rate about 110 Gy/

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min, with total doses ranging from 10 kGy to 350 kGy. The dose rate was determined by $Ag_2Cr_2O_7$ standard dosimeter.

2.3. Characterization

Soxhlet extraction (in cyclohexane) was used to determine the decomposition products during irradiation. About 1 g BRPs were cut into thin slices, extracted in cyclohexane for 72 h at room temperature, and then vacuum-dried overnight at 70 °C.

To measure the swelling ratio (SR) of BRPs, the dried sample was immersed in cyclohexane for 3 days with replacing the solvent every 24 h to ensure complete equilibration. The swelling ratio was calculated according to the following equation:

$$SR = (W_t - W_0)/W_0 \quad (1)$$

where W_t and W_0 are the weights of the swollen gels and dried samples, respectively. All the SR experiments were repeated three times for reproducibility.

The FTIR spectra of the extracted samples were carried out with a Nicolet 6700 FTIR spectrometer.

The measurements of molecular weights of the extracted samples were carried out with PL-GPC-220 high temperature chromatograph. Trichlorobenzene was used as solvent at 150 °C, with concentration of about 0.2 wt%.

The observations of the morphological structures of the samples were carried out using ZEISS EVO 18 special edition scanning electron microscope at acceleration voltage of 10 kV. The samples with various doses were treated with acetone for 72 h to remove uncrosslinked PR and the radiolysis products. Then the surface was coated with gold.

Dynamic mechanical properties were recorded on a DMA 242D (Netzsch, Germany) instrument in tension mode. Samples with uniform size of 14 × 4 × 2 mm^3 were used. The specimens were analyzed at a frequency of 5 Hz in a temperature ranging from − 100.0 °C to 80 °C at a heating rate of 3.0 °C min^{-1}.

Tensile properties of the samples were tested using a SANS CMT7000 testing machine, fitted with a 500 N load cell; at a crosshead of 250 mm/min. Five samples for each composition were tested for reproducibility.

Positron annihilation lifetime spectroscopy was carried out using a conventional fast-slow coincidence system which has a time resolution of 195 ps with energy window set for ^{22}Na. The positron annihilation lifetime was measured at room temperature.

3. Results and discussion

In the present study, BRP was prepared by the PR crosslinking of BR. The uncrosslinked polymer chains and the additives can be extracted to calculate the degree of crosslinking. The soxhlet extraction results show that the control has an extractives content of 14.9 ± 0.8%, which obviously increase with increasing absorbed dose to 37.2 ± 1.2% of sample irradiated at 350 kGy. The swelling ratio of the samples also increases from 294 ± 3% of the control to 766 ± 4% of sample irradiated at 350 kGy (Table 1). It can be concluded that BRP is sensitive to gamma irradiation, which would undergo chain scission to generate dissolvable components with increasing dose.

FT-IR was further used to examine the extractives of control and the sample irradiated at 350 kGy by solution casting (Fig. 1). The spectra show significantly increased organic compounds (characteristic peak: 2900 cm^{-1}, which corresponds to the C-H bond; 1470 cm^{-1}, which corresponds to C=C bond), confirming the decomposition of BRP under gamma irradiation. These degraded low-molecular weight compounds made the samples softer and sticky to some content with increasing absorbed dose. The molecular weights of the extractives were further characterized with a high temperature GPC.

The molecular weight and its distribution of extractives of the control and irradiated samples are shown in Fig. 2 and Table 1. The

Table 1
Extractives content and swelling ratio of BRP with various absorbed doses.

Sample	Control	10 kGy	100 kGy	200 kGy	350 kGy
Extractives content (%)	14.9 ± 0.8	15.3 ± 0.5	17.6 ± 0.9	24.1 ± 2.0	37.2 ± 1.2
Swelling ratio (%)	294 ± 3	305 ± 0	399 ± 2	528 ± 15	766 ± 4

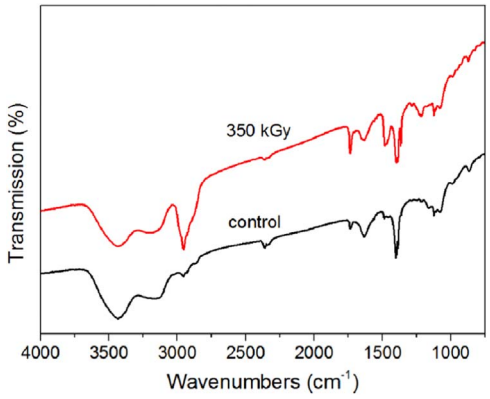

Fig. 1. FTIR spectra of the extractives of control and the sample irradiated at 350 kGy.

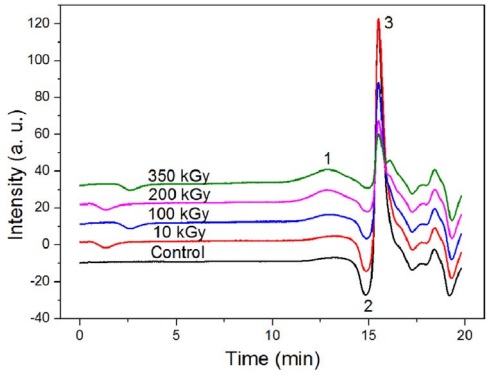

Fig. 2. GPC traces of the control and irradiated BRP.

extractives contain three components, which are attributed to the dissolvable BR, PR and additives used in the fabrication process of BRP, respectively. BR peak gradually shifts to longer retention time when increasing absorbed dose, with molecular weight increasing from 27,500 of control to 46,300 of sample irradiated at 500 kGy. Simultaneously, the intensity of BR peak slightly increases with increasing absorbed dose. This is probably attributed to the chain scission of the network structure of BR, generating dissolvable polymer chains. With increasing absorbed dose, larger amounts of chain segment with higher molecular weight generates because of higher possibility of chain scission, which are consistent with the soxhlet extraction and FTIR results. The molecular weights of the other two components, PR and the additive, are not sensitive to irradiation over the dose range investigated, probably due to the limited sensitivity of the GPC to the small molecular weight samples.

Fig. 3 shows the brittle fractured surface of BRP. It can be seen that uncrosslinked PR was dissolved in acetone, causing lots of holes in the materials. The control and sample irradiated with 10 kGy show dual-phase morphology with PR domains dispersed in the BR matrix. The dimensions of PR domains are several microns. With increasing absorbed dose, the material surface becomes smoother and the holes in the materials disappeared. It is proposed that, the irradiated samples