Studies on Aging Behavior and Degradation Mechanism of Raw EPDM Rubber Under γ-Ray Irradiation

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Abstract: The aging behavior and mechanism of EPDM rubber were investigated by conducting different doses of γ-rays irradiation in raw EPDM rubber. A series of characterization methods such as Mooney viscosity, molecular weight and its distribution, rubber processing analysis, infrared spectroscopy analysis *et al*. were carried out. Structure and modifications of the raw EPDM rubber were tested under different irradiation doses. The results showed that the Mooney viscosity and molecular weight of EPDM rubber showed a significant decrease with increasing irradiation dose. The gel content and glass transition temperature increased gradually. As strain rose, the energy storage modulus diminished. The loss factor decreased at first and then increased with increasing strain during irradiation aging.

Keywords: EPDM rubber, Aging mechanism, γ-rays, Degradation, Mooney viscosity.

1. INTRODUCTION

Polymer materials are widely used in many fields such as aviation, military, automobile, construction and so on because of their excellent light weight, high strength and corrosion resistance and other characteristics beyond the performance of traditional materials. Furthermore, the application of polymer materials is becoming more and more extensive with the continuous development of modern industrial technology. As one of the polymer materials, rubber is subject to a comprehensive impact of various environmental factors such as light, heat, oxygen, water, chemical reagents and biological corrosion during its processing, transportation, storage and usage [1-6]. These lead to aging of the rubber material and lead to a gradual decline in its various properties. Moreover, the service life of the rubber was seriously affected.

EPDM rubber, as a new type of synthetic rubber will inevitably experience a certain degree of aging process during the use. Therefore, the anti-aging research on EPDM rubber is very necessary. In particular, the irradiation effect that rubber products are subjected to will lead to cross-linking and breakage of the molecular chains inside the rubber, so that the service life of rubber products will be reduced. This prolonged irradiation may cause electrical failures and bring great safety hazards. Therefore, it is particularly important to study the aging mechanism of rubber irradiation and improve its irradiation resistance. Researches on the aging mechanism of EPDM rubber can be in-depth investigation of its aging process. So more advanced anti-aging technology can be studied to improve the anti-aging properties of ethylene propylene rubber. Then the use of ethylene propylene rubber temperature, rubber aging slower and service life will be extended. As EPDM rubber is a polymer, its molecular structure is very complex, because it contains a large number of polar groups. Therefore, in-depth investigation of the mechanism of rubber aging is of vital significance to achieve its anti-aging goals [7-10].

The purpose of this paper is to study the mechanism and influencing factors of raw EPDM rubber in the aging process under γ-ray irradiation, and to investigate the changes in the structure and processing properties of raw rubber after irradiation. Theory and practice are linked to provide a theoretical and experimental basis for the preparation of irradiation-resistant EPDM rubber. By irradiating EPDM raw rubber with different doses of γ-rays and testing the changes of structure and properties of EPDM raw rubber under irradiation conditions, some reliable experimental support for the design of irradiation-resistant EPDM rubber can be provided.

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2. EXPERIMENT

The mixer was used to evenly mold the EPDM raw rubber. The raw rubber was passed through the gaps between two rollers of the mixer for several times until the surface of the raw rubber got smoothly. Then the raw rubber was wrapped and put in a certain dosage γ-rays irradiation environment.

In this experiment, EPDM raw rubber with brand KEP-240 was selected for Co-60 γ-rays irradiation test. The cumulative irradiation doses were selected as 0, 50, 200, 400, 600 and 1000 kGy, respectively. The EPDM raw rubber was obtained under different irradiation doses. The aging mechanism of EPDM rubber under γ-rays irradiation was studied. The raw EPDM rubber was tested for GPC, Mooney viscosity, gel content, RPA, FTIR and DSC analysis methods. Based on the experimental results, the effects and reasons of different γ-rays irradiation doses on the structure and processing performance of EPDM raw rubber were analyzed in details.

3. RESULTS AND DISCUSSIONS

3.1. Effect of γ-Rays Irradiation on Mooney Viscosity of EPDM

Mooney viscosities of EPDM raw rubber under different irradiation doses of γ–rays were shown as in Figure **1**.

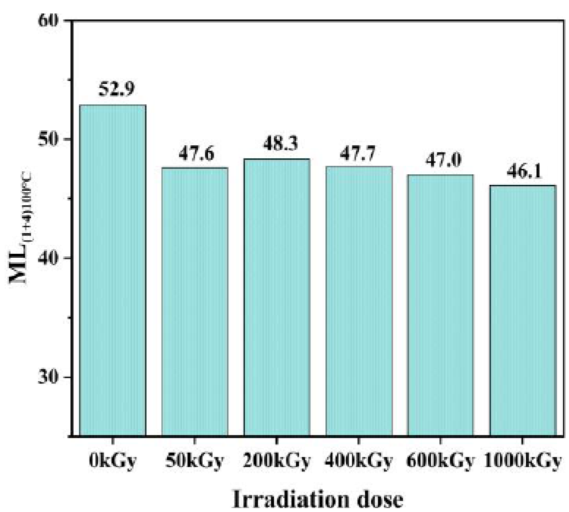

Figure 1: The effect of irradiation on Mooney viscosity of EPDM rubber.

It can be seen that the Mooney viscosity value of EPDM raw rubber is the highest of 52.9 as no γ–rays irradiated the rubber. The Mooney viscosity values of the raw rubber showed a significant decrease after irradiation. All the values were below 50. The Mooney viscosity of EPDM raw rubber increased at first and then decreased with the increase of radiation dose. The EPDM raw rubber had the highest Mooney viscosity, good physical and mechanical properties. But it had general processing properties. The flowability of EPDM raw rubber was improved after irradiation because of the decrease in Mooney viscosity. Moreover, Mooney viscosity appeared to tend towards an asymptotic value for higher doses of radiations, this was because EPDM became similar linear small molecules with similar molecular weights at different high radiation doses, so the degrees of decrease in Mooney viscosity were similar and fluidity increased.

3.2. Effect of γ–Rays Irradiation on Molecular Weight and Distribution of EPDM Raw Rubber

The effect of γ–rays irradiation on molecular weight and distribution of EPDM raw rubber was shown as in Figure **2**.

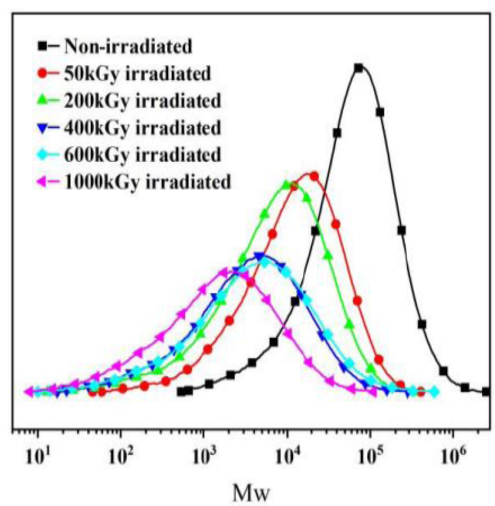

Figure 2: The effect of irradiation on molecular weight and distribution of EPDM rubber.

It showed that the EPDM raw rubber exhibited a single peak without γ-rays irradiation, and displayed a narrow molecular weight distribution. However, the molecular weight distribution of EPDM raw rubber was relatively wide when γ-rays irradiated. So the GPC spectrum revealed that γ-ray irradiation could effectively reduce the molecular weight of EPDM rubber.

3.3. The Effect of γ–Rays Irradiation on the Processing Performance of EPDM Raw Rubber

The tanδ and strain of EPDM raw rubber under different does of γ-rays radiation was shown as in Figure **3**. it can be clearly seen that there is a close relationship between the loss factor and strain of

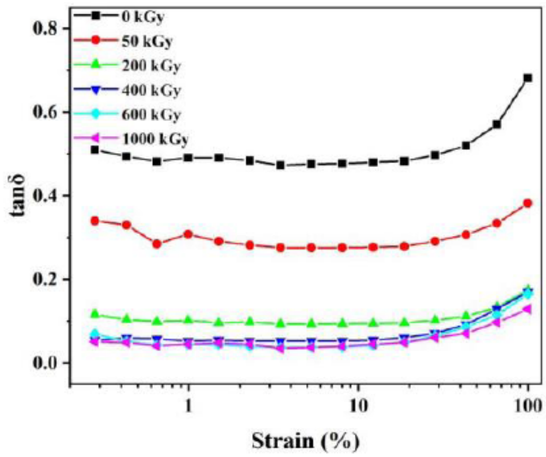

Figure 3: The relationship between tanδ and strain of EPDM raw rubber under different does of γ-rays radiation.

EPDM raw rubber, which can better reflect the processing performance of rubber. The results showed that the loss factor first decreased and then increased with the increase of strain. When the strain exceeded 10%, the loss factor rapidly increased. Because molecular chains of rubber have stronger elasticity under smaller strains, which can more effectively suppress internal friction and improve the processing performance of raw rubber. As the strain increased, the relative slip between molecular chains would lead to an increase in internal friction until a certain threshold reached. Compared with EPDM rubber without radiation, the loss factor tanδ of irradiated EPDM raw rubber was smaller, which indicated that γ-ray radiation reduced the processing performance of EPDM rubber.

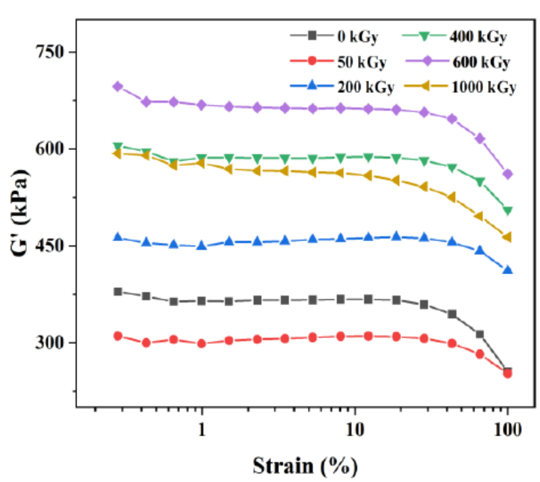

Figure 4: The relationship between G' and strain of EPDM raw rubber under different does of γ-rays radiation.

The relationship between G' and strain of EPDM raw rubber was shown as in Figure **4**.

The Payne effect is a phenomenon of change in the energy storage modulus (G') of a material, which manifests itself as a significant decrease in the energy storage modulus with increasing strain, and the strength of this change can be measured by the difference between the highest value of G' at low strain and the lowest value at high strain (ΔG'). Figure **4** demonstrated the relationship between energy storage modulus and strain for raw EPDM rubber, and it was clear that the energy storage modulus G' decreased significantly as the strain increases. The decrease in G' becomes faster as the strain exceeded 10%, while at smaller strains this change was slower. Because the increase in strain caused deformation or rupture of the three-dimensional network of the packing and resulting in a decrease in G' with an increase in strain. The decrease of G' was slower at small strains, while the decrease of G' accelerated when the strain was greater than 10%, which was due to the molecular chain from the initial orientation and slippage, became to break gradually, and the packing network was destroyed more than rebuilt.

On the other hand, Figure **4** showed the relationship between ΔG' and irradiation dose of raw rubber, and comparing the data, it can be seen that the ΔG' value of EPDM raw rubber with an irradiation dose of 600 kGy has the largest value, and the ΔG' value of EPDM raw rubber with an irradiation dose of 1,000 kGy has the second largest value, whereas ΔG' values at other irradiation doses were all reduced. ΔG' values were reduced and the smallest ΔG' value was found at an irradiation dose of 200 kGy.

3.4. The Effect of γ–Rays Irradiation on the Gel Content of EPDM Raw Rubber

The gel content of raw rubber was shown as in Figure **5**.

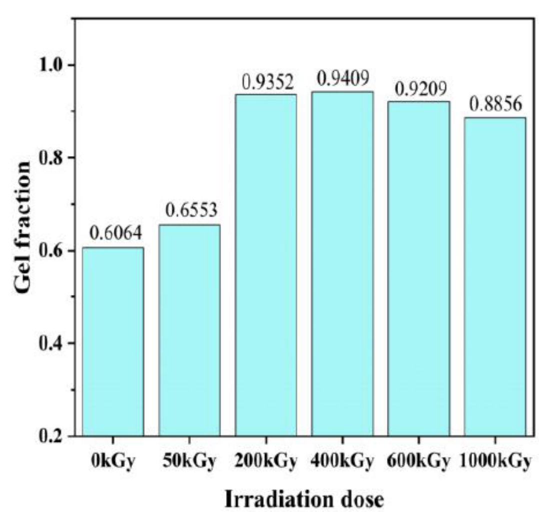

Figure 5: The gel content of raw rubber.

Gel is a special kind of rubber, which is formed due to branching or cross-linking and cannot be dissolved in organic solvents. The initial gel fraction of raw EPDM rubber without γ-rays irradiation was 0.6064, and the gel fraction gradually increased with the increase of irradiation dose the gel fraction gradually increases. When the irradiation dose of 400 kGy, the gel fraction reached the highest value, the gel fraction of 0.9409, and the irradiation dose then increases, the gel fraction instead of a decline. This is because due to the role of ray irradiation, when the molecular chain is subjected to external forces, their branching reaction will be accelerated, and ultimately form a kind of gel-like material that cannot be dissolved by toluene, if the dose exceeds the standard, it may damage the rubber material, so that it is subjected to degradation. If the dosage is exceeded, the rubber material may be damaged and subjected to degradation.

3.5. Infrared Spectral Analysis (FTIR)

Using the Fourier Transform technique, we can obtain the infrared spectra of EPDM raw rubber in Figure **6** to further delve into the structure of its internal molecules and observe their relative vibration and motion characteristics when receiving infrared light irradiation. In addition, this technique can also be used to evaluate the radiation resistance characteristics of rubber, helping us to understand more comprehensively the radiation damage it endures.

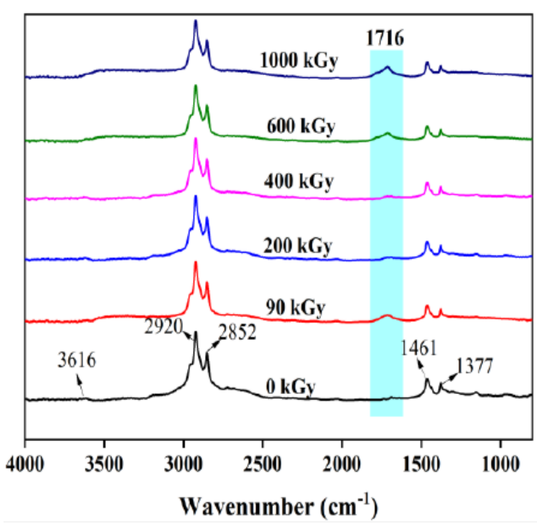

Figure 6: Infrared spectrogram of raw rubber.

From the infrared spectra, it can be seen that the peak at 3616cm^{-1} was the stretching vibration peak of the carbon-hydrogen bond on the double bond of the third monomer; the absorption peak at 2920cm^{-1} was the asymmetric stretching vibration peak of the methylene group, and the symmetric stretching vibration peak of the methylene group at 2852 cm^{-1}; the absorption peak at 1716cm^{-1} was the bending vibration peak of the extracyclic double bond out of the face of the third monomer; the bending vibration peaks of the methylene group and the asymmetric bending vibration peaks of the methylene group were at 1461 cm^{-1}; 1377cm^{-1} absorption peak was the deformation vibration of methyl group.

3.6. Differential Scanning Calorimetry (DSC) Analysis

The DSC curves of raw rubber were shown as in Figure **7**.

The thermal properties of rubber materials can be quickly and reliably analyzed by means of differential scanning calorimetry, the basic principle of which is to measure and record the power difference between a specimen and a standard reference at a constant temperature, thus deriving a differential scanning calorimetric curve for the material. In addition, the method can also be used to study the heat absorbed or released by melting or chemical reactions of crystallization within the rubber to better understand the thermal property characteristics of the material.

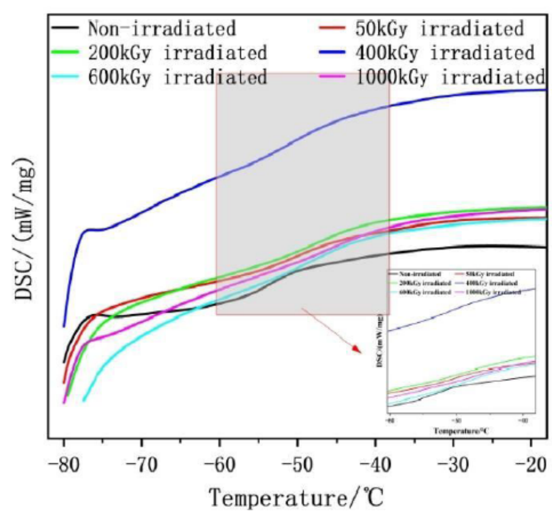

Figure 7: DSC curves of raw rubber.

Figure **7** showed the DSC curves of raw rubber under different γ-rays irradiation doses, and a comparison of the curves showed that the curves of raw EPDM rubber without γ-rays irradiation were more varied than those of raw rubber irradiated by γ-rays. This indicated that as the irradiation dose increased, the flexibility of the molecular chain decreases, which leads to an increase in the glass transition temperature of the EPDM rubber.

4. CONCLUSIONS

This paper investigates the EPDM raw rubber irradiated with different doses of γ-rays, and finds that the structure and properties of EPDM changed after γ-rays irradiation through the tests of Mooney viscosity, molecular weight and its distribution, gel content, rubber processing analysis, infrared spectroscopy and differential scanning calorimetry, etc., and that different irradiation doses have different influences on the comprehensive performance of the EPDM raw rubber.

In this paper, the structure and properties of EPDM were found to be changed after γ-rays irradiation through the testing of Mooney viscosity, molecular weight and its distribution, gel content, rubber processing analysis, infrared spectroscopy and differential scanning calorimetry analysis, etc., and different irradiation dosages will have different effects on the comprehensive performance of EPDM rubber. The following conclusions were drawn. The aging behavior of EPDM rubber was affected by γ-ray irradiation. The Mooney viscosity of EPDM raw rubber increased at first and then decreased with the increase of radiation dose. GPC spectrum revealed that γ-ray irradiation could effectively reduce the molecular weight of EPDM rubber. The loss factor tanδ of irradiated EPDM raw rubber decreased clearly.

ACKNOWLEDGMENTS

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