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TECHNICAL NOTE

Dose rate effect on LDPE cross-linking induced by electron beam irradiation

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Abstract In this project, the radiation induced cross-linking in low-density polyethylene (LDPE) samples irradiated with the different dose rates of 5 MeV electron beam in the dose range of 20 up to 200 kGy were investigated and compared. The dose rate values were ranged from 13 to 300 kGy/min. The cross-linking of the samples were studied on the basis of gel content measurement and a hot set test. The yield of gel content showed slight increases with decreasing dose rate values. The same results were more clearly observed using the hot set test. It was concluded that, to irradiate the product based on LDPE, any variation in electron beam current causes different dose rates which, in turn, affect the absorbed dose value in materials.

Key words dose rate • electron beam • low-density polyethylene • cross-linking • chain scission

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Introduction

Many investigations about irradiation of polymers and its effects on chemical structure and its physical properties have been made and published. When polymers are exposed to ionizing radiation, even at low doses, often undergo structural changes accompanied by molecular cross-linking, grafting and chain scission reaction. Radiation can extract hydrogen atoms from the polymer chain and thus generate hydrocarbon radicals. Two of these radicals might combine to form an extended polymer molecule. As this process repeats, a polymer network is formed [6]. The radiation cross-linking of low-density polyethylene (LDPE) irradiated under the 5 and 10 MeV electron beams in the wide range of dose was previously investigated [3, 8, 9]. The effects of high energy electron beam on mechanical and thermal properties of LDPE have been reported as well [4].

The industrial application of cross-linking by irradiation with fast electrons has been extensively used for the production of heat shrinkable polyethylene films and tubes. Also, cross-linked polyethylene has been used in hot water piping installations and wire and cable industries [3, 4, 9]. In this work, an attempt has been made to investigate and to compare the induced cross-linking in LDPE samples, when irradiated with different dose rates of 5 MeV electron beam. For each individual dose rate, the range of radiation dose of 20 to 200 kGy was applied. To evaluate the cross-linking induced by irradiation the gel content and the hot set of irradiated samples were measure.

Experimental

Materials

Two grades of low-density polyethylene (LDPE) supplied by Bandar Imam Petrochemical Company LDPE 0030 and LDPE 0200 with a melt flow index (MFI) of 0.3 and 2 g/10 min, and a density of 0.92 g/cm^3 were used in this investigation. These materials were chosen on the basis of minimum and maximum available MFI of LDPE.

Sample preparation and irradiation

Several sets of the samples were prepared in the sheet form of 2 ± 0.1 mm thickness using a warm press system. The samples in each set were irradiated with doses varying from 20 to 200 kGy with a constant dose rate. The value of dose rate controlled by the electron beam current was changed from 13 to 300 kGy/min. The irradiation was performed using the Rhodotron type electron accelerator machine, TT200 model, using 5 MeV electron beam with a maximum of 16 mA beam current (Fig. 1). As it is shown in Fig. 2, the samples were irradiated in stationary mode on the region of uniform dose, which has been measured and reported before [10].

Measurements

Gel content

The gel content of the irradiated samples was determined by extracting the sample in boiling xylene for 16 hrs [2]. After the samples were dried, the insoluble residues were weighed. The average gel content was calculated as $100 \times (m_2/m_1)$, where m_1 is the sample initial mass and m_2 is the residue mass after chemical processing. For any given irradiation conditions, three samples were analyzed to determine the average gel content.

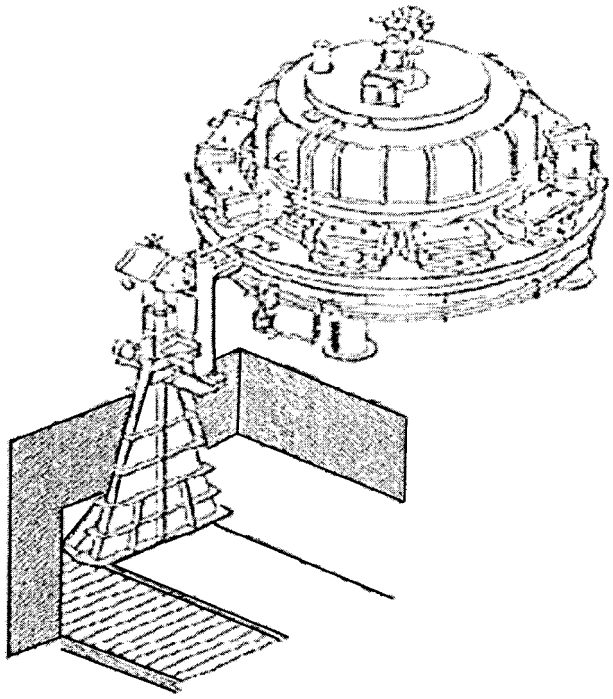

Fig. 1. Scheme of Rhodotron accelerator and irradiation system.

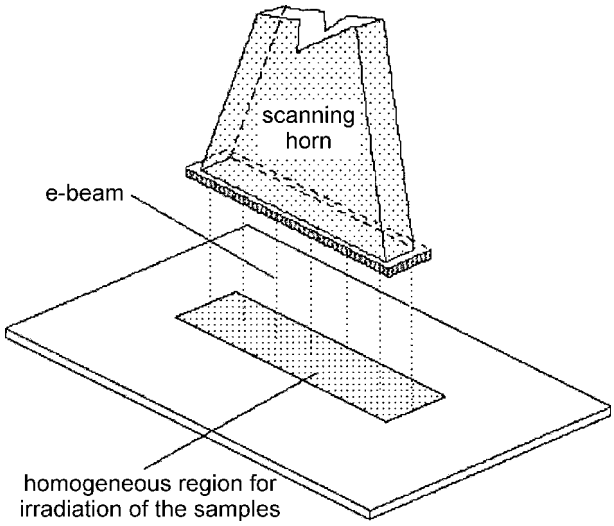

Fig. 2. Scheme of the scanning horn and irradiation geometry.

Hot set testing

The Heraeus UT 6050HS heating oven was used for determining thermal expansion of the samples [1].

The temperature of the oven was set at 200°C with measuring period of 15 minutes. This measuring system was characterized by the temperature control precision of ± 3°C and the measuring accuracy of integral electronic callipers ± 0.01 mm. Testing and measuring were made by the use of laser optics. The working formula for calculation of hot set value (thermal expansion) was $100 \times (L_1 - L_0)/L_0$ where L_0 and L_1 is the initial and final sample length, respectively. To determine the average thermal expansion, five samples were used for each case.

Results and discussion

Figure 3 shows the variations of gel content as a function of radiation dose for two grades of 0030 and 0200 LDPE samples. In all cases, it was observed that the gel content increases with increasing absorbed dose. On the other hand, these variations are slightly decreased by increasing the dose rate value. Also, Fig. 4 shows that the hot set values continuously decrease while absorbed dose values increase. Figure 4 also indicates that the variations increase with increasing dose rates. This is due to the fact that irradiation causes further cross-linking in amorphous regions, but extensive chain scission of taut tie molecules leads to increase of the crystallinity and crystal perfection [5, 7]. LDPE has a very low melt flow index and thus contains long chains. As a result of chain

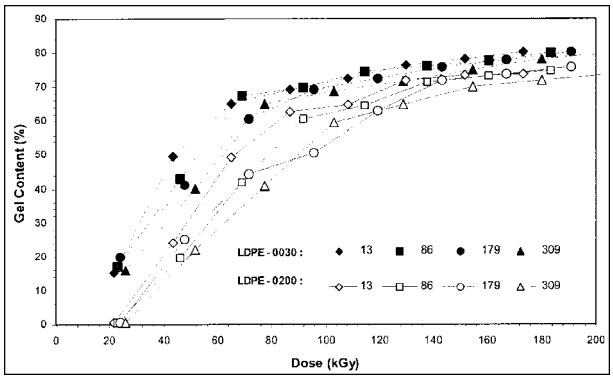

Fig. 3. Variations of gel content of the samples as a function of radiation dose. (Dose rate values are also indicated in the legend in kGy/min.)

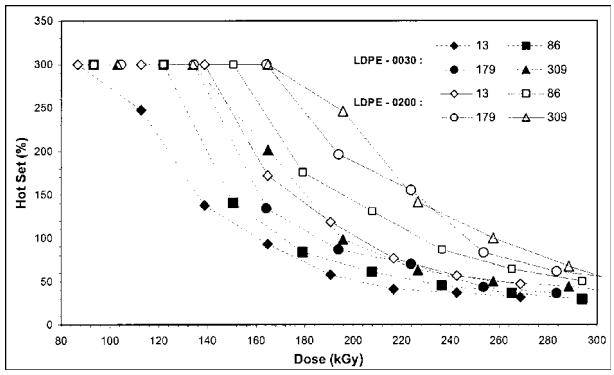

Fig. 4. Hot set variation *vs.* the dose. (Dose rate values are also indicated in the legend in kGy/min.)

scission the long chains of LDPE are replaced by shorter chains, which are more readily aligned in crystalline regions and show increase in degree of crystallinity.

Also irradiation with higher dose rate periodically induces different mechanisms with competing contributions to the formation and quantity of the crystalline phase which leads to the creation of new crystalline regions. Also, a further increase of the dose leads to decreasing the crystallinity. This is due to the radiation local overheating which leads to melt the crystalline phase [9]. The melting is a result of the slowed down heat exchange processes between the sublevels of structural units. In these results, it can be seen clearly that increasing the dose rate reduces the radiation induced cross-linking in the samples as the effect of dose rate.

Conclusion

To improve the properties of the LDPE using radiation, one of the important parameters which should be taken into consideration, is the dose rate. Therefore, this may make some limitations in using different radiation sources depending on the desired properties of the material. This is due to the fact that the dose rate values of gamma rays emitted by a gamma source are very low in comparison with the high energy electron beams. Also the dose rate, depending on the available electron beam current of electron accelerator machine, can be varied.

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