Effects of High-Energy Electron Beam on Low-Density Polyethylene Materials Containing EVA

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ABSTRACT: A series of low-density polyethylene (LDPE) blends with different amounts of ethylene–vinyl–acetate (EVA) was prepared and irradiated with 10 MeV electron beam in the range of 0–250 kGy at room temperature in air. EVA was used as a compatibilizer and softener in four different amounts: 5, 10, 20, and 30 wt %, based on polyethylene (PE). The crosslinking of the samples was studied on the basis of gel-content measurements as well as some thermal and mechanical properties of the specimens. The results indicated that the LDPE and LDPE–EVA blends could be crosslinked by a high-energy electron beam, of which their thermal and mechanical properties changed effectively, however, because of EVA content of the polymer; the blends were more sensitive to lower doses of radiation. These studies were carried out to obtain a suitable compound for heat-shrinkable tubes. © 2004 Wiley Periodicals, Inc. J Appl Polym Sci 92: 1049–1052, 2004

Key words: crosslinking; electron beam irradiation; polyethylene (PE); gel content; LDPE–EVA compounds

INTRODUCTION

The radiation crosslinking of polymer blends can produce many materials with superior properties (e.g., the mixture of EVA and semicrystalline polymers such as polyethylene gives composite materials which possess high thermoshrinkage and high stability when subjected to aging, weathering, aggressive media, etc.).[1–5] The treatment of such compositions with high-energy radiation leads to additional and unpredictable changes in their structure and properties. This is so because the irradiation itself or the usage of different additives may have a significant and ambiguous effect upon the same polymer.[2] Until now, many investigations reported on the effect of medium- and low-energy irradiation on polyethylene (PE)[1] and blended PE[6]; nevertheless, studying the effects of 10 MeV electron beam on PE, and blended PE materials, has not been investigated as much as in the range of low and medium energy. In this article, the process of gel forming over different doses of radiation together with some physical properties of crosslinked PE by using 10 MeV is reported.

EXPERIMENTAL

The investigated samples were low-density polyethylene (LDPE) and blends of LDPE with 5, 10, 20, and 30% ethylene–vinyl–acetate (EVA) sheets. Codes L, L5, L10, L20, and L30 indicate pure PE with 5, 10, 20, and 30% EVA, respectively. The sheets were compression molded between aluminum at 180°C for 5 min under 200 bar pressure in an electrically heated press to prepare sheets of 2 ± 0.05 mm thickness. The LDPE was grade (LH-0075) from Bandar Emam Petrochemical Plant (Iran) and EVA component contained 18% EVA (supplied by BP Co., UK). The samples were then irradiated at room temperature in air by using high-energy electron accelerator (TT200 Rhodotron, Yazd/Iran Radiation Processing Center). The specifications of the Rhodotron are given in Table I.

The gel fraction (W_g) was determined by extracting the soluble component in boiling xylene for a total of 21 h at $T = 140$°C and by drying the residue for about 4 h at $T = 150$°C. To avoid oxidation, 1% Irganox 1010 was added to xylene according to ASTM D2765-95. The weight of the treated samples was marked as m_1, while the initial weight was registered as m_0. The gel fraction was defined as $W_g = m_1/m_0$. Three samples were used to determine the average gel content for each case.

The melting points of irradiated specimens were measured by using a differential scanning calorimeter (DSC-50, Shimadzu) under nitrogen atmosphere. The heating rate in the DSC measurement was 10°C/min, and the temperature range was from room temperature up to 300°C.

Elongation at break (EB) was measured on dumbbell specimens at a test speed of 50 mm/min at room temperature according to ASTM D3895 type V in an

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TABLE I
RHODOTRONTT200 Electron Beam Accelerator Parameters

Beam energy	≅ 10 MeV
Beam power at 10 MeV	≅ 70 kW
Energy dispersion at 10 Mev	≅ 300 KeV
Scanning range	30–100 cm
Total power consumption	<300 kW
R.F. frequency	107.5 MHz
R.F. power output	200 kW
Electron gun average current	0–10 mA
Resolution	±50 μA

Instron 4411. The average of three data was taken and the experimental error was 2.5%.

RESULTS AND DISCUSSIONS

The gel contents of different blends of pure PE and EVA as a function of irradiation dose are shown in Figure 1. For all cases, it is observed that gel contents increase rapidly with increasing irradiation dose and reach about 80% at 250 kGy. It also reveals that the materials are crosslinked properly. However, the sensitivity of the blends is higher at low-radiation doses in comparison with pure LDPE and the most sensitive blend is L30. It is also shown that the addition of EVA to LDPE facilitates the crosslinking process. In all radiation doses used, the amount of gel content of the blends is higher than that of pure PE. Furthermore, irradiation with doses greater than about 100 kGy leads to a slow increase of the gel content because the effectiveness of the gel forming process is lowered. This means that although the crosslinking processes continue, radiation-induced macromolecular chemical bonds occur predominately between already crosslinked macromolecular chains. This leads only to increasing the crosslinking density in the obtained spatial macromolecular lattices.[6]

In Figure 2, the variation of elongation at break versus absorbed dose has been plotted. This figure shows that the elongation at break decreases with the increase of absorbed radiation dose, which is due to a crosslinking process. This is similar to behavior of crosslinked PE in low-energy electron beam.[7] As shown in Figure 2, the elongation at break for L30 is less than other samples, which confirms the gel-content measurements in Figure 1. In other words, EVA content of blends enhances the sensitivity of the samples.

Variation of the melting point of the compounds against the radiation doses has been plotted in Figure 3. These points have been achieved by extensive DSC measurements of the samples. It is seen that the melting point gradually decreases with the augmentation of the radiation dose. This denotes that size of the crystallite becomes small, and crystallites with small size are dispersed in the crosslinked polymer matrices.[8]

In Figure 4, the tensile strength has been plotted against radiation dose. The tensile strength increases marginally with the radiation dose, which is in confirmation with Figures 1 and 2. The variation of heat of fusion ΔH versus irradiation dose has been plotted in Figure 5. It is seen that as the EVA content of the PE increases, the value of ΔH decreases, which corresponds to the reduction in the crystallinity of the compound. It is also seen that ΔH decreases margin-

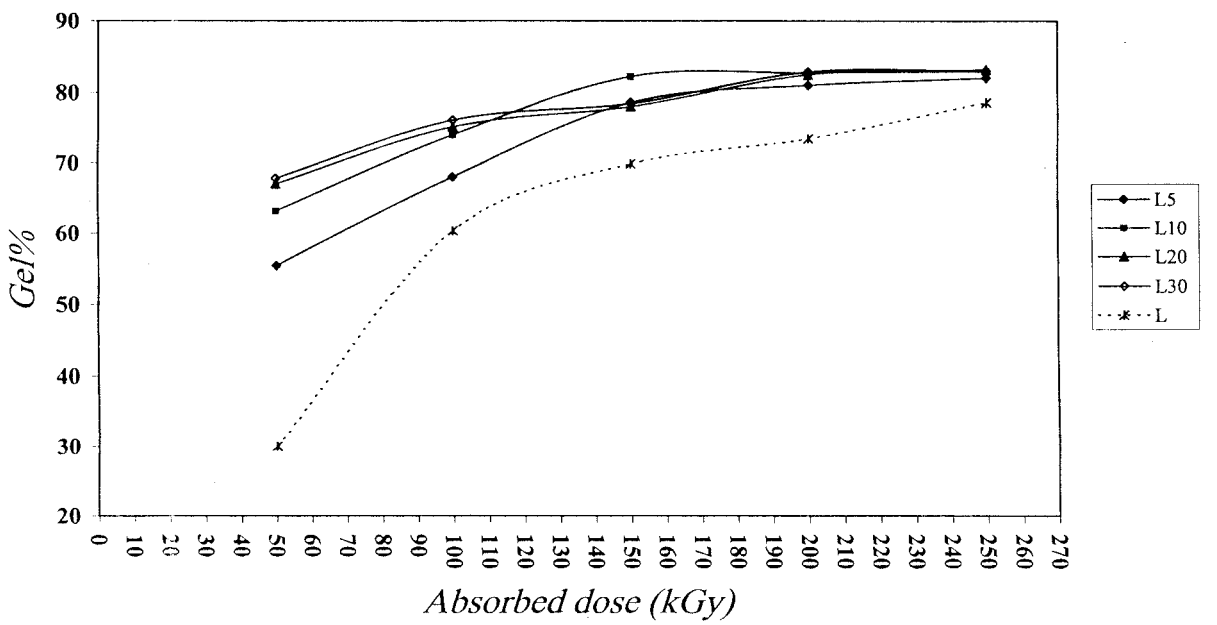

Figure 1 Gel-content variations versus absorbed dose for LDPE and LDPE–EVA blends.

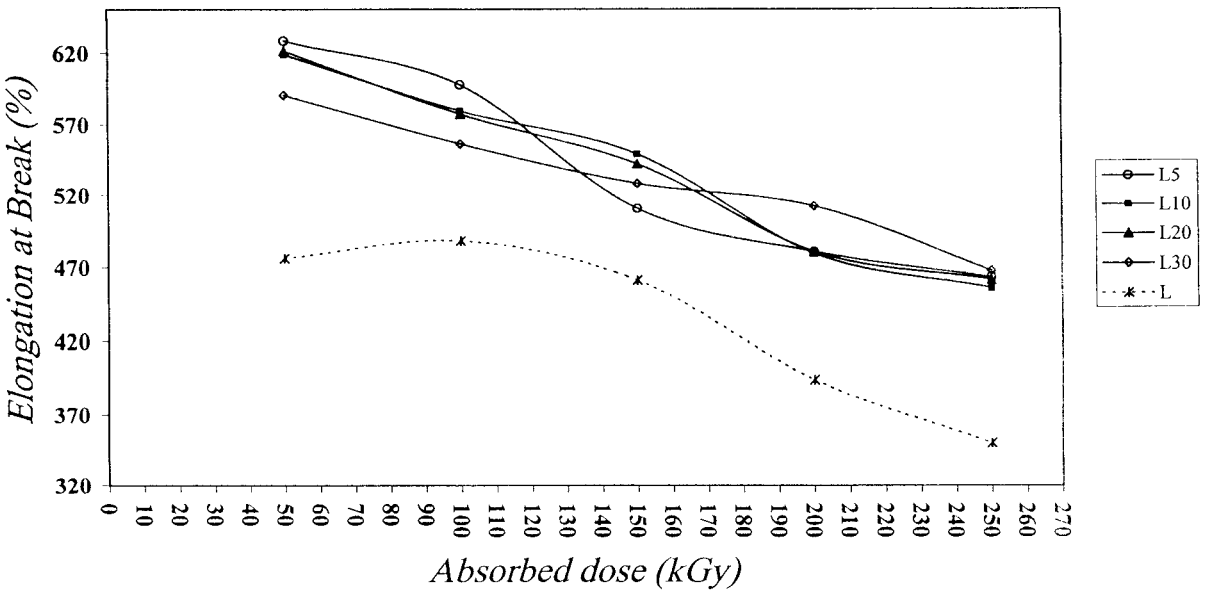

Figure 2 Variations of elongation at break as function of absorbed dose.

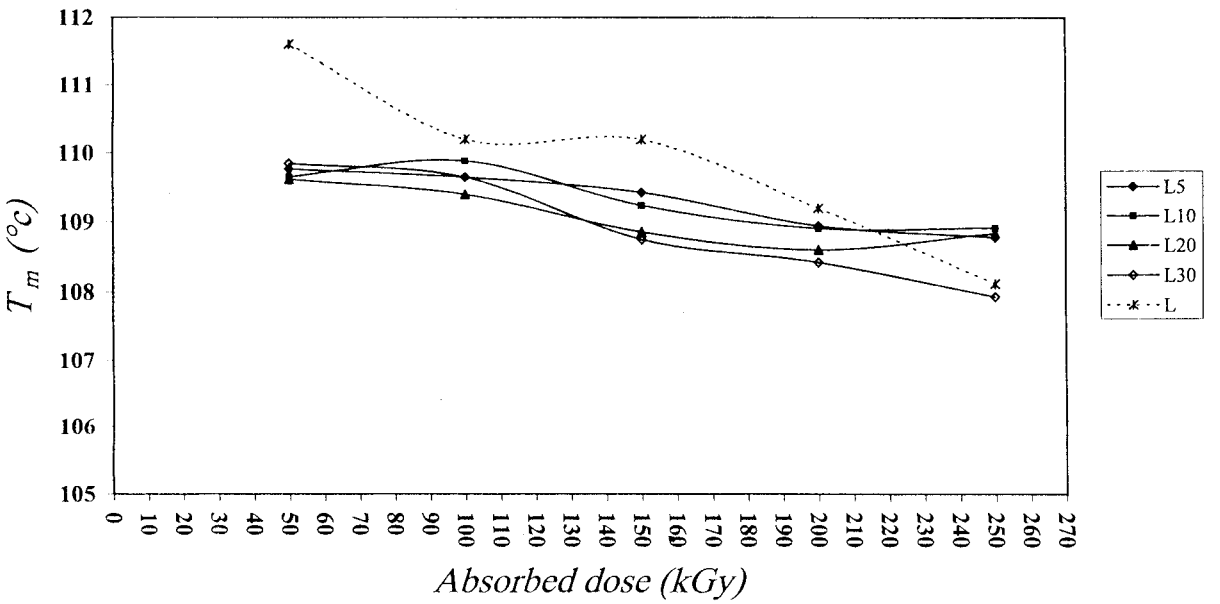

Figure 3 Variations of melting point versus absorbed dose.

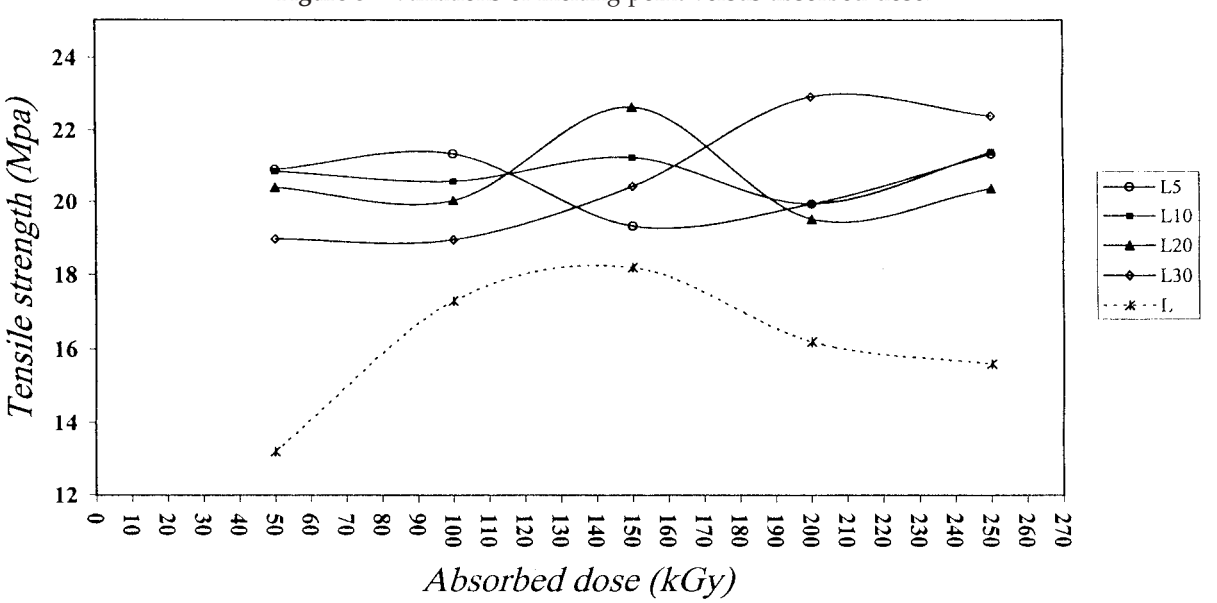

Figure 4 Variation of tensile strength versus absorbed dose.

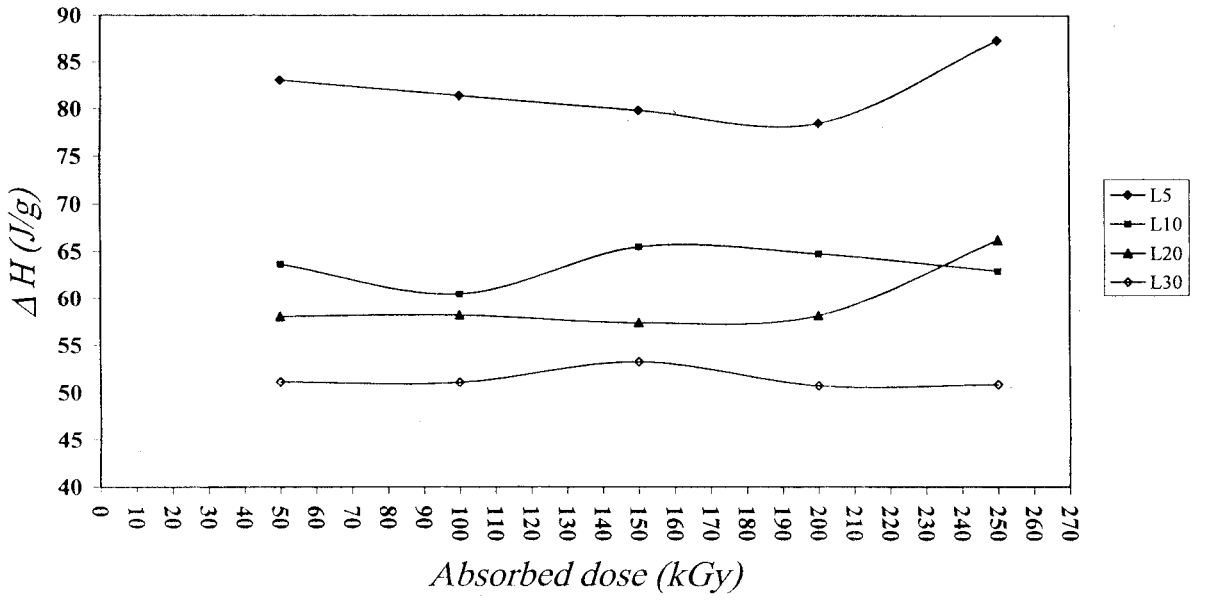

Figure 5 Changes of heat of fusion (ΔH) versus irradiation dose.

ally with irradiation dose.[9–11] The changes of ΔH are well related to the mechanical properties in Figures 2–4; that is, the crystallinity decreases, enhancing the mechanical properties. Finally, we found that the LDPE–EVA blends at certain conditions can be used as a good material for shrinkable tubes.

CONCLUSION

Gel-content measurement shows that crosslinking could be induced by high-energy electron beam (10 MeV) in LDPE and LDPE–EVA blends; crosslinking increases with irradiation dose up to about 200 kGy. However, EVA content of blends enhances the sensitivity of the samples at lower radiation doses.

Mechanical properties of the samples are improved by irradiation, which is in confirmation with gel-content measurements. The heat of fusion ΔH also decreases marginally with irradiation dose. The changes of ΔH are well related to the mechanical properties; that is, the crystallinity decreases, enhancing the mechanical properties. The melting point (T_m) also reduces gradually with the increase of radiation, which supports the variations of crystallinity.

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References

1. Qu, B. J.; Ranby, B. Polym Eng Sci 1995, 35(14), 1161.
2. Charlesby, A. Atomic Radiation and Polymers; Pergamon Press: Oxford, 1960.
3. Crompton, T. R. Practical Polymer Analysis; Plenum Press: New York, 1993.
4. Hoyle, C. F.; Kinstle, J.F. Radiation Curing of Polymeric Materials; American Chemical Society: Washington, DC, 1960.
5. Mark, J. Physical Properties of Polymer, 2nd ed.; American Chemical Society: Washington, DC, 1993.
6. Mateev, M.; Karageorgiev, S. Radiat Phys Chem 1998, 51(20), 205.
7. Datta, S. K.; Chaki, T. K.; Khastgir, D.; Bhowmick, A. K. Polym Polym Composites 1996, 4(6), 419.
8. Oshima, A.; Ikeda, S.; Seguchi, T.; Tabata, Y. Radiat Phys Chem 1997, 49(2), 279.
9. Tikku, V. K.; Biswas, G.; Despande, R. S.; Majali, A. B.; Chaki, T. K.; Bhowmick, A. K. Radiat Phys Chem 1995, 45, 829.
10. Chaki, T. K.; Despande, R. S.; Roy, S.; Majali, A. B.; Tikku, V. K.; Bhowmick, A. K. Appl Polym Sci 1995, 53, 141.
11. Seguchi, T.; Arakawa, K.; Ito, M.; Hayakawa, N.; Machi, S. Radiat Phys Chem 1983, 21, 495.