NUKLEONIKA 2007;52(2):77–81

ORIGINAL PAPER

# Electrical properties of EVA/LDPE blends irradiated by high energy electron beam

**Mahmood Borhani, Ghazanfar Mirjalili, Farhood Ziaie, Mohammad A. Bolorizadeh**

**Abstract.** A series of low-density polyethylene (LDPE) blends with different amounts of poly(ethylene-vinyl acetate) (EVA) was prepared and irradiated with 10 MeV electron beam in the range of 50–380 kGy at room temperature in air. Some electrical properties of the blends such as surface and volume resistance, breakdown voltage, permittivity and loss factor were studied as a function of the EVA content and the radiation dose. It was revealed that the surface resistance and volume resistivity of the blends reach a maximum at a 170 kGy dose of radiation and 30 wt% of EVA. There is no considerable change at breakdown voltage permittivity and loss factor of the irradiated samples; however, the permittivity and the loss factor of the blends increase significantly with increasing amount of EVA contents.

**Key words:** EVA/LDPE blends • high energy electron beam • electrical properties • surface resistances • permittivity (dielectric constant) • loss factor

M. Borhani, M. A. Bolorizadeh
Physics Department,
Shahid Bahonar University,
P. O. Box 76169-133, Kerman, Iran

G. Mirjalili✉
Physics Department,
Yazd University,
P. O. Box 89195-741, Yazd, Iran
and Yazd Radiation Processing Center,
Yazd, Iran,
Tel.: +98 351 8216545, Fax: +98 352 6234840,
E-mail: Gmirjalili@yazduni.ac.ir

F. Ziaie
Yazd Radiation Processing Center,
Yazd, Iran
and Material and Ion Beam Application Division,
Nuclear Research Center for Agriculture and Medicine,
P. O. Box 31485-498, Karaj, Iran

Received: 21 November 2006
Accepted: 2 April 2007

## Introduction

Polymers have found extensive applications in cables and capacitors as dielectric materials [24, 27, 29, 39]. Mixing of poly(ethylene-co-vinyl acetate) EVA and semi-crystalline polymers such as low-density polyethylene (LDPE) gives composite materials having better flexibility, toughness and high resistance to environmental stress cracking due to the increase of the adhesive strength at the matrix-rubber particle interface [1, 14, 28, 30].

The treatment of such composition with high energy radiation leads to various changes in their structure and properties [3, 7]. Electron beam (EB) processing has been demonstrated, to be a very effective means of improving and use properties of various polymers [9, 23]. Until now, many investigations have been reported on the effects of low, medium and high energy radiation on mechanical, electrical and thermal properties of EVA/LDPE blends [5, 16, 20, 22]. Nevertheless, studies of the effect of high energy electron beam on electrical properties of the blended EVA/LDPE samples have received less attention.

In a previous work [31], we reported the effect of high energy electron beam on mechanical and thermal properties of EVA/LDPE blends. In this work, the effect of 10 MeV electron beam on some electrical properties such as: surface resistance, volume resistivity, permittivity (dielectric constant), loss factor and voltage breakdown of these blends is reported.

## Experimental

LDPE (LH-0075) with a density of 0.921 g·cm$^{-3}$ was obtained from Bandar Emam Petrochemical Company (Iran). EVA containing 18% vinyl acetate was purchased from BP Co. UK. The EVA/LDPE blends in ratios of 0.0, 10, 20, 30 and 100 wt% of EVA were melt-mixed and then molded to flat sheets (2 ± 0.05 mm thickness) using hot pressing at a temperature of 180°C for 5.0 min under 20 MPa pressure and the sample were then cooled to room temperature. Radiation of the sample sheets was carried out at room temperature in air by a high energy electron accelerator (TT200, Yazd/Iran Radiation Processing Center) under various radiation doses ranging from 50 to 380 kGy.

Surface and volume resistance were measured at room temperature by a teraohmmeter, made by CEAST Company.

A dielectric loss measurement system, model TRS-10T (ANDO Electric Co., Japan) was used for determining the dielectric constant and the dielectric loss tangent of the samples. Frequency was tuned at 1 MHz in the experiments.

The voltage breakdown of the samples was measured by a Dielectric Rigidity system P/N 6135.053, CEAST Company, made in Italy.

## Results and discussion

Figure 1 compares the surface resistance of EVA/LDPE blended samples containing 10, 20 and 30 wt% of EVA with unblended LDPE and EVA as a function of irradiation dose. It is clear from Fig. 1 that the surface resistance of the unblended EVA samples is substantially lower than that of LDPE and also EVA/LDPE blends. In addition, it can be seen that the more content of EVA is used in the blend, the lower surface resistance is achieved. All samples (except unblended LDPE) showed an increase of surface resistance when irradiated by high energy electron beam up to 170 kGy of irradiation dose. Around the 170 kGy radiation dose, which is an optimum dose for the best cross-linking of the EVA/LDPE blends [31]; there is a noticeable increasing of surface resistance for all blends. For the radiation doses higher than 170 kGy, the surface resistance of the samples decreased abruptly (with the exception of LDPE), and then converged to each other.

With increasing EVA content in the composition up to 30 wt%, the maximum of the trace observed at 170 kGy is increased more rapidly than the blends with the smaller content of EVA component. In addition, the surface resistance of the unblended EVA sample is substantially smaller than that of the blended samples at high irradiation doses. With increasing radiation dose, however, the surface resistance of the LDPE remained unchanged. Similar trend is revealed in Fig. 2 in which alike the surface resistance curves, the volume resistivity of the blend containing 30 wt% of EVA is higher than the other samples around the 170 kGy radiation dose. Nevertheless, the volume resistivity at higher doses for all cases are identical. The noticeable peaks in the surface and volume resistivity curves around 170 kGy may be related to higher cross-linking density at this dose of radiation which causes to increase the average molecular weight of polymer. Presence of much more cross-linking points can be considered as barrels to prevent the charge movement between polymer chains and thus increase the electrical resistance of cross-linked sample [4, 25]. This argument is in agreement with the Kolesov study on the PE in which he described the influence of molecular weight on morphological and electrical properties of PE, he also has shown that the increase of the molecular weight leads to an increase in the volume resistivity [17]. Also Marsacq *et al.* [21] have shown that the increase in molecular weight leads to an increase in charging ability of PE, i.e. the growth of a number of traps in material which prevent the charge movement inside the polymer and hence increase the electrical resistance. Therefore, it can be said that the accordance of electrical resistance peak of the blend with the 170 kGy irradiation dose is related to the increase of traps and molecular weight of polymer due to cross link formation.

At higher doses, the cross-linking process occurs along with chain scission reaction, probably this later is predominating [8, 35]. As a result, non homogeneities owing to molecular chain segmentation which are introduced into the polymer matrix and polar and ionic products due to irradiation can probably cause the resistance of samples to decrease [18, 26].

As can be observed from Figs. 1 and 2, the surface resistance and volume resistivity of the blends around

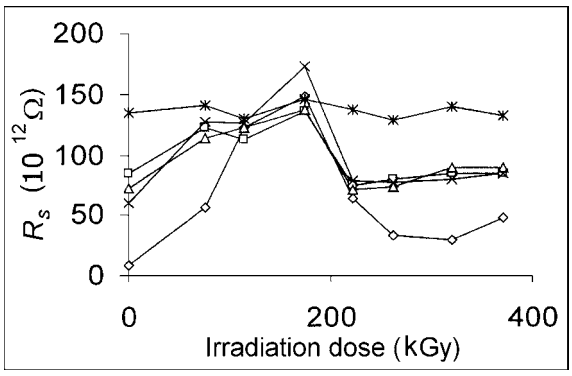


**Fig. 1.** Surface resistance vs. absorbed dose of 10 MeV electron beam for (◇) unblended EVA, (＊) LDPE and EVA/LDPE blends at (□) 10, (△) 20 and (×) 30 wt% of EVA.

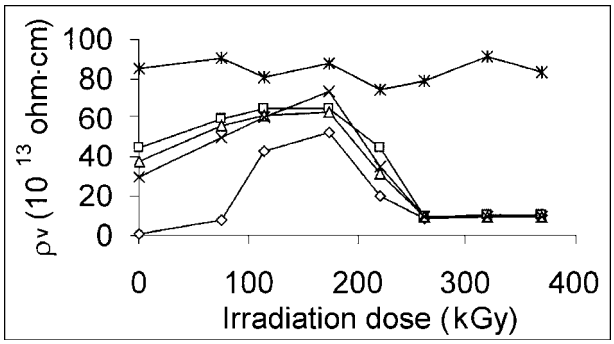


**Fig. 2.** Volume resistivity vs. absorbed dose of 10 MeV electron beam for (◇) unblended EVA, (＊) LDPE and EVA/LDPE blends at (□) 10, (△) 20 and (×) 30 wt% of EVA.

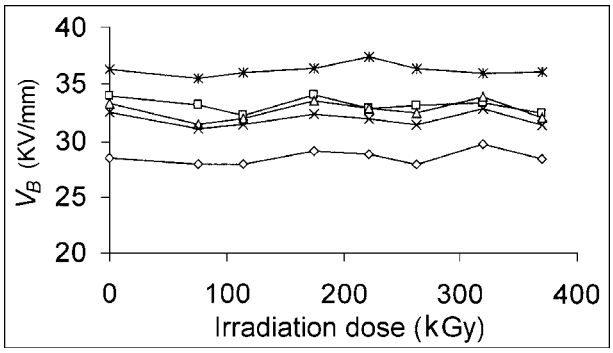


**Fig. 3.** Breakdown voltage vs. absorbed dose of 10 MeV electron beam for (◇) unblended EVA, (∗) LDPE and EVA/LDPE blends at (□) 10, (△) 20 and (×) 30 wt% of EVA.

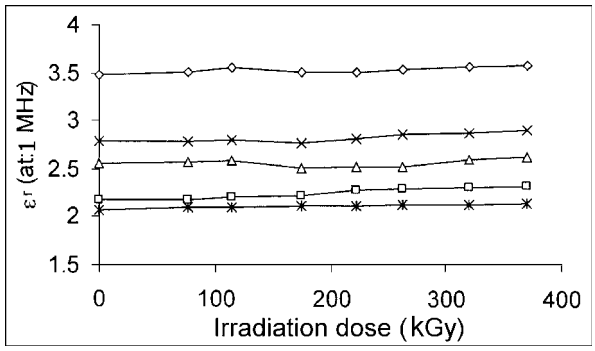


**Fig. 4.** Variation of the dielectric constant of (◇) unblended EVA, (∗) LDPE and EVA/LDPE blends at (□) 10, (△) 20 and (×) 30 wt% levels of the former, with absorbed dose of 10 MeV electron radiation.

the 170 kGy increase with the content of EVA. Several studies have indicated that the gel content and the cross-linking density of EVA/LDPE blends at an optimum radiation dose increase with increasing EVA content [10, 31, 32], and the highest gel content is observed when the amount of EVA in the blended samples reaches 30 weight percent [10, 11]. So, we can state that the highest surface resistance and volume resistivity of the blends at 170 kGy and 30 wt% of EVA may be related to the highest cross-linking density.

In Fig. 3, the breakdown voltage of the samples against the radiation dose has been plotted. EVA samples undergo dielectric breakdown at voltages lower than LDPE and EVA/LDPE blends. In all cases no considerable variation of breakdown voltage is revealed when the radiation dose is increased. This may be attributed to the fact that increasing of the radiation dose leads to an increase in cross-linking degree in amorphous area which may be barrels to prevent the electrical breakdown path inside the polymer matrix [19, 25, 40]. On the other hand, increasing of the radiation dose increases the chain scission of the PE and leads to increase the carbonyl groups and also shallow trap centers [6, 15], so, this causes to increase the mobility of the charge carrier and finally decrease the breakdown voltage. The mutual effect of radiation, i.e. the increase of breakdown voltage due to cross-linking on the one hand, and the decrease of breakdown voltage due to chain scission, on the other hand, causes the breakdown voltage of the polymer sample to remain without considerable variation. Our assumption is that the above argument shows an opposite behavior, so it may be concluded that the radiation dose does not make effect so much on the breakdown variation. From Fig. 3, it can also be seen that the blends and EVA break electrically in voltages lower than LDPE samples which may be attributed to the presence of polar groups on the EVA back bone [34, 38].

Figures 4 and 5 show the dependence of the permittivity (dielectric constant) and the loss factor of the samples on the radiation dose, respectively. Figure 4 shows that the permittivity of the blends is smaller than that of EVA, and with increasing radiation dose the permittivity increases very slowly. One may conclude that the electron beam radiation disintegrates molecules and increases the number of dipoles, so that the higher radiation dose produces a larger permittivity [13, 33].

The permittivity of the polymer is representative of the various polarization phenomena that come into play when the polymer is subjected to an electric field. The overall polarization of a polymer, like the PE, is the sum of four terms: electronic, atomic, orientation and space-charge polarization, among them the first two are intrinsic in nature and for non-polar polymer are important [2]. For polar polymers, both the atomic and electronic polarization are often negligible compared to orientation and space charge polarization [2]. It has been shown that there is a relationship between injecting depth and energy of electron radiation. For example, as the radiation energy of electrons is 15 MeV or 30 keV, the injecting depth can be approximately deduced as 4 mm and 50 μm, respectively [36, 37]. This means that 10 MeV electron beam can penetrate totally inside the 2 mm polymer sheet, so there will be very little space charges left in the bulk of the sample. So, it is the orientation polarization that has a major contribution to the permittivity of a polar polymer like the EVA. This arises owing to the presence of permanent dipoles on the back bone of the macromolecular chain. Since, LDPE is a non-polar polymer with smaller permittivity than EVA; its blends would show permittivity which is held between the permittivity of the LDPE and EVA depending on the content of EVA component in the blends. In other words, the more content of EVA in the blends, the larger amount of permittivity will be achieved. It is desirable to keep the capacitance of the

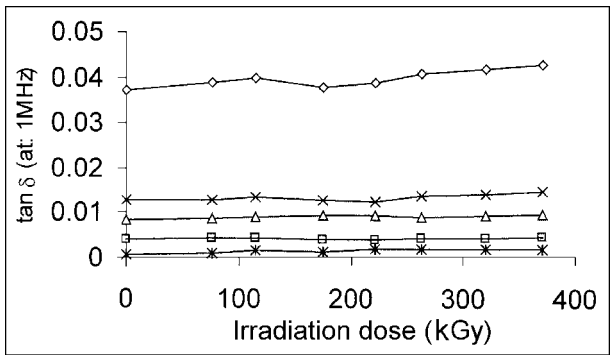


**Fig. 5.** Variation of the dielectric loss factor of (◇) unblended EVA, (∗) LDPE and EVA/LDPE blends at (□) 10, (△) 20 and (×) 30 wt% levels of the former, with absorbed dose of 10 MeV electron radiation.

insulating material as minimal as possible when they are used as cables in industries. Furthermore, from Fig. 4 it can be seen that although the permittivity of the blends is smaller than that of EVA and larger than that of LDPE, however, the content of EVA in the blends (up to 30 wt%) and the radiation dose (up to 380 kGy) have negligible influence on the permittivity of the LDPE samples.

Figure 5 reveals that the loss factor of the LDPE polymer is smaller than that of EVA/LDPE blends and EVA which can be related to the presence of polar groups and heterogeneity nature of the blend samples. For non-polar polymers, the loss factor may, however, increase sharply due to the presence of additives, impurity concentrations and physical heterogeneity [12]. However, the loss factor of all samples remains without significant change with increasing radiation dose. This means that mixing of a polar polymer at low amount level can affect the electrical properties of the non-polar polymer, whereas the radiation dose cannot create considerable effect on the electrical properties of such polymers.

## Conclusions

Effects of adding the EVA polymer and of high energy electron radiation on the electrical properties of low-density polyethylene have been investigated. The surface and volume resistance and also breakdown voltage of the blends prepared by mixing of various amounts of EVA with LDPE decrease compared to the unblended LDPE, while the permittivity and loss factor increase. The larger increase of the amount of EVA in the blends, the greater change of the cited electrical properties occur in the polymer sample. On the other hand, the breakdown voltage, dielectric constant and loss factor of all the blends remain relatively constant and the surface and volume resistance of the samples were affected significantly by increasing the radiation dose. So, the resistance of all blends increases when the radiation dose increases up to 170 kGy. The increase of the resistance of the blend containing 30% of EVA is larger than that of the other blend samples. Increasing the radiation dose over 170 kGy considerably reduces the surface and volume resistance of all samples.

## References


1. Arsac A, Carrot C, Guillet J (1999) Rheological characterization of ethylene vinyl acetate copolymers. J Appl Polym Sci 74:2625–2630
2. Banik I, Chaki TK, Tikku VK, Bhowmick AK (1998) Effect of electron beam irradiation on the dielectric properties of fluorocarbon rubber. Angew Makromol Chemie 263:5–13
3. Bhattacharya A (2000) Radiation and industrial polymer. Prog Polym Sci 25:371–401
4. Boudou L, Guastavino J (2002) Influence of temperature on low-density polyethylene films through conduction measurement. J Phys D: Appl Phys 35:1555–1561
5. Chattopadhyay S, Chaki TK, Bhowmick AK (2001) Development of new thermoplastic elastomers from blends of polyethylene and ethylene-vinyl acetate copolymer by electron-beam technology. J Appl Polym Sci 79:1877–1889
6. Chen G, Banford HM, Davies AE (1995) Effect of gamma-irradiation on electrical breakdown stress of LDPE. In: Proc of the 1995 IEEE 5th Int Conf on Conduction and Breakdown in Solid Dielectrics, 10–13 July 1995, Leicester, UK, pp 556–560
7. Chen G, Banford HM, Davies AE (1996) The influence of gamma radiation on space charge formation in low-density polyethylene. In: IEEE Annual Report of the Conf on Electrical Insulation and Dielectric Phenomena, 20–23 October 1996, San Francisco, USA, pp 821–824
8. Chen G, Zhong LS, Xie HK, Banford HM, Davies AE (2002) The influence of oxidation on space charge formation in gamma-irradiated low-density polyethylene. Radiat Phys Chem 66:247–255
9. Dadbin S, Frounchi M, Haji Saeid M, Gangi F (2002) Molecular structure and physical properties of e-beam cross-linked low-density polyethylene for wire and cable insulation applications. J Appl Polym Sci 86:1959–1969
10. Dadbin S, Frounchi M, Sabet M (2005) Studies on the properties and structure of electron-beam cross-linked low-density polyethylene/poly [ethylene-co-(vinyl acetate)] blends. Polym Int 54:686–691
11. Dalai S, Wenxiu C (1996) Investigation of radiation cross-linked foam of LDPE/EVA blends. J Appl Polym Sci 62:75–80
12. Das-Gupta DK (1994) Polyethylene: structure, morphology, molecular motion and dielectric behavior. IEEE Electrical Insulation Magazine 10:5–15
13. Datta SK, Chaki TK, Khastgir D, Bhowmick AK (1996) Electron beam initiated grafting and cross-linking of ethylene vinyl acetate copolymer. Part II. Mechanical and electrical properties. Polym Polym Compos 4:419–428
14. Hui Z, Jiufu X (1993) The modification of the flexibility of radiation cross-linked PE by blending PE with EVA and CPE. Radiat Phys Chem 42:117–119
15. Kacarevic-Popovic Z, Kostaski D, Novakovic L, Miljevic N, Secerov B (2004) Influence of the irradiation conditions on the effect of radiation on polyethylene. J Cerb Chem Soc 69:1029–1041
16. Kim JI (2001) Study on the cross linking characteristics of LDPE/EVA blends by γ-ray irradiation. J Korean Ind Eng Chem 12:800–805
17. Kolesov SN (1980) The influence of morphology on the electric strength of polymer insulation. Dielectrics and Electrical Insulation, IEEE Transactions on Electrical Insulation 15:382–388
18. Kostoski D, Dojcilovic J, Novakovic L, Suljovrujic E (2005) Effects of charge trapping in gamma irradiated and accelerated aged low-density polyethylene. Polym Degrad Stab 91:2229–2232
19. Laghari JR, Hammoud AN (1990) A brief survey of radiation effects on polymer dielectrics. IEEE Trans Nucl Sci 37:1076–1083
20. Lee C, Kim K-Y, Ryu B-H (2006) Radiation effects on electrical and mechanical properties of LDPE/EVA blends. In: IEEE Conf Record of the Int Symp on Electrical Insulation, 11–14 June 2006, Toronto, Ontario, Canada, pp 261–264
21. Marsacq D, Hourquebie P, Olmedo L, Janah H (1995) Effects of structural parameters of polyethylene on space charge properties. Sci Tech Appl 275:432–438
22. Martinez-Pardo ME, Zuazua MP, Hernandez-Mendoza V, Cardodo J, Montiel R, Vazquez H (1995) Structure-properties relation ship of irradiated LDPE/EVA blend. Nucl Instrum Methods Phys Res B 105:258–261
23. Mateev M, Kargeorgiev S (1998) The effect of electron beam irradiation and content of EVA upon the gel-

forming processes in LDPE-EVA films. Radiat Phys Chem 51:205–206
24. Oda E (1981) Wire and cable. Radiat Phys Chem 18:241–252
25. Okashita M, Ymaguchi M, Fujita M, Shintate H, Wang S, Shiono T (1998) Relationship between impulse breakdown and morphology of polyethylene. In: IEEE Proc of Int Symp on Electrical Insulating Materials, 27–30 September 1998, Toyohashi, Japan, pp 549–552
26. Oproiu C, Martin D, Toma M, Marghitu S, Jianu A (2000) Transitory and permanent effects of electron beam irradiation on insulating materials. Nucl Instrum Methods Phys Res B 166/167:669–675
27. Ota S (1981) Current status of irradiated heat-shrinkable tubing in Japan. Radiat Phys Chem 18:81–87
28. Peon J, Vega JF, Aroca M, Martinez-Salazar J (2001) Rheological behavior of LDPE/EVA-c blends. 1. On the effect of vinyl acetate co monomer in EVA copolymers. Polymer 42:8093–8101
29. Rado R, Zelenak P (1992) Applications of cross-linked polyethylene. Int Polym Sci Tech 19:72–77
30. Rodriguerz-Perez MA, Duijsens A, De Saja JA (1998) Effect of addition of EVA on the technical properties of extruded foam profiles of low-density polyethylene/EVA blends. J Appl Polym Sci 68:1237–1244
31. Salehi SMA, Mirjalili G, Amrollahi J (2004) Effects of high-energy electron beam on low-density polyethylene materials containing EVA. J Appl Polym Sci 92:1049–1052
32. Sharif J, Hanisah S, Aziz SA, Hashim K (2000) Radiation effect on LDPE/EVA blends. Radiat Phys Chem 58:191–195
33. Shinyama K, Fujita S (2001) Dielectric and thermal properties of irradiated polyetheretherketone. IEEE Transactions on Dielectrics and Electrical Insulation 8:538–542
34. Suh KS, Park JH, Kim JY, Takada T (1994) Nature of interface and charge formation in PE/EVA laminates. In: IEEE Conf Record of the Int Symp on Electrical Insulation, 5–8 June 1994, Pittsburgh, USA, pp 166–169
35. Suljovrujic E, Kacarevic-Popovic Z, Kostoski D, Dojcilovic J (2000) Effect of ageing on the dielectric relaxation of oriented and gamma irradiated LDPE. Polym Degrad Stab 71:367–373
36. Vallayer B, Hourquebie P, Marsacq D, Janah H (1996) Measurement of the trapping and detrapping properties of polymers in relation with their microstructure. In: IEEE Conf Record of the Int Symp on Electrical Insulation, 16–19 June 1996, Montreal, Quebec, Canada, pp 674–677
37. Xu L, Weifang J, Demin T, Muratu K, Takada T (1997) Electrical properties of radiation cross-linked polyethylene. In: IEEE Proc of the 5th Int Conf on Properties and Applications of Dielectric Materials, 25–30 May 1997, Seoul, South Korea, pp 311–314
38. Zhimin D, Jie K, Demin T (2000) Relationships between space charge accumulation and electrical or water treeing in improved PE and XLPE by ethylene-acrylic acid copolymer. In: IEEE Proc of the 6th Int Conf on Properties and Applications of Dielectric Materials, 21–26 June 2000, Xi'an, China, pp 562–566
39. Zhiming Z, Zhenhao L, Renrui T *et al*. (1993) Construction of production line of radiation cross-linking wire and cable in Tianshui. Radiat Phys Chem 42:113–116
40. Ziaie F, Borhani M, Mirjalili G, Bolorizadeh MA (2006) Effect of crystallinity on electrical properties of electron beam irradiated LDPE & HDPE. In: Proc of the 6th Int Meeting on Radiation Processing (IMRP), 27 February – 3 March 2006, Kuala Lumpur, Malaysia