Indian J. Phys. **83** (**8**) 1117-1122 (2009)

AC electrical properties of proton irradiated EVA films

Anjum Qureshi[1], Sejal Shah[1], Dolly Singh[1], N L Singh[1]* and K P Singh[2]
[1]Department of Physics, M S University of Baroda, Vadodara-390 002, Gujarat, India
[2]Department of Physics, Panjab University, Chandigarh-160 014, Punjab, India

E-mail: singhnl_msu@yahoo.com

Abstract : MeV ions passing through polymeric films modify their electrical, optical and thermal properties and these changes are related to changes in the chemical structure of the polymers. Ethylene vinyl acetate (EVA) films were irradiated with 3 MeV proton beam at different fluences of 10^{13}, 10^{14} and 10^{15} ions/cm^2. AC electrical properties of pristine and irradiated samples were studied in the frequency range 100 Hz to 100 kHz by means of an LCR meter. There is an exponential increase in conductivity with log frequency and conductivity increases as fluence increases. The dielectric loss/constant is observed to change with the fluence. FTIR spectra reveals significant change in intensities of functional groups at a fluence of 10^{15} ions/cm^2 due to scissioning of polymer chains.

Keywords : Dielectric properties, FTIR, EVA.

PACS Nos. : 61.80.jh, 61.82.Pv, 77.22.Ch

1. Introduction

The dielectric properties of a material are of great concern for telecommunication applications. Since the signal propagation delay time is proportional to the square root of the dielectric constant of the transmitting medium, a low dielectric constant is often desirable. For example, high speed integrated packages require materials with dielectric constant lower than 3.5, whereas satellite communication applications require materials with a constant of 2.5, in addition to a low loss tangent. Few materials have high temperature thermal stability with low dielectric constant. Ethylene vinyl acetate (EVA) is among them. It is a flexible polymer, compatible with many other polymers and additives. It has excellent quality of heat resistance and heat stability. EVA based blends and homopolymers are widely used in heat shrinkable materials for wire and cable insulation and foam materials [1]. The best dielectric materials are those which contain a minimum of charge carriers and potential charge carriers which may be formed by the splitting of covalent atomic or molecular bonds under the influence of the

*Corresponding Author

energetic ions. An increase in temperature will also increase mobility. The dielectric response of a material provides information about the orientation and translational adjustment of mobile charges present in the dielectric medium in response to an applied electric field. The most important property of the dielectric material is its ability to be polarized under the action of the field [2].

The modification of polymer properties under ionizing radiation is a subject of widespread importance due to the increasing use of polymers in hard radiation environments encountered in space crafts, nuclear power plants, sterilization irradiators, high energy particle accelerators *etc.* During irradiation, various physical and chemical processes take place in the polymer. Coulombic interactions between ions and electrons of host atoms, excessive bond stretching due to localized energy deposition, and atomic displacement by nuclear collision can release pendant atoms such as hydrogen and cause bond breakage or chain scission. Thus various gaseous molecular species are released during irradiation [3]. The effects of electron beam irradiation and EVA content on the radiation crosslinking in LDPE/EVA blends have been studied by Gordiickuck *et al* [4], Mateev and Karageogiev [5] and Jamaliah *et al* [6]. Gamma irradiation effects of both LDPE/EVA and high-density polyethylene (HDPE)/EVA were studied by Dalai *et al* [7,8] and found that the factors of good compatibility, higher amorphous region's content, and higher EVA content in the blends were favorable with regard to enhance the radiation crosslinking of polyethylene (PE)/EVA blends. The mechanical and thermomechanical properties of gamma irradiated PP/EVA blends were studied by Thomas *et al* [9] and Minkova and Nikolova [10]. Sharif *et al* [11] studied the effects of radiation on mechanical properties of LDPE/EVA. In this paper, we studied effect of 3 MeV proton irradiation on electrical properties of EVA films at different fluences.

2. Experimental details

Three pieces of EVA [$(C_6H_{10}O_2)_{n;}$ density 0.92 g/cm^3] each of thickness 260 μm and of size 1.2 cm × 1.2 cm were cut from commercially available sheet (Nocil Plastics, Mumbai, India). These samples were irradiated with 3 MeV protons using low energy Cyclotron accelerator at Physics Department, Chandigarh, India. The beam current density was of the order of 30 nA/cm^2 and irradiation was carried out at three fluences *viz*. 10^{13}, 10^{14} and 10^{15} ions/cm^2. The proton beam was made incident perpendicular to the target of 6 mm diameter. The irradiation was performed under vacuum at a pressure of 10^{-6} Torr. The AC electrical properties of all samples were measured in the frequency range 0.1–100 kHz using an LCR meter (General Radio, USA, and Model 1689). The dielectric constant was measured by applying an air drying type of silver paste. The AC conductivity was calculated using the relation $\sigma = t/RA$ $(\Omega.\text{cm})^{-1}$, where t, R and A are thickness of the film, resistance measured and cross-sectional area of the electrode respectively. The dielectric constant was calculated using the relation

$\varepsilon = Cp/C_0$, where Cp is capacitance measured and $C_0 = \varepsilon_0 A/t$, where ε_0 is the permittivity of vacuum. The tan δ was measured directly using LCR meter [12].

3. Result and discussion

The projected range of 3 MeV proton beam in EVA was calculated to be 155 μm using SRIM-2003 code [13]. The electronic stopping power $(dE/dx)_e$ and nuclear stopping power $(dE/dx)_n$ were found to be 1.137 eV/Å and 7.183×10^{-4} eV/Å, respectively.

3.1. FTIR spectroscopy :

Figure 1 shows the FTIR spectra of pristine and irradiated films. The absorption bands as obtained from the pristine spectrum are identified as (A) 615 cm^{-1} alkyne C-H bending vibration; (B) 1450 cm^{-1} alkane C-H bending vibration; (C) 1730 cm^{-1} C=O stretching vibration; (D) 2894 cm^{-1} C-H stretching vibration; (E) 3400-3700 cm^{-1} OH stretching vibration for the analysis.

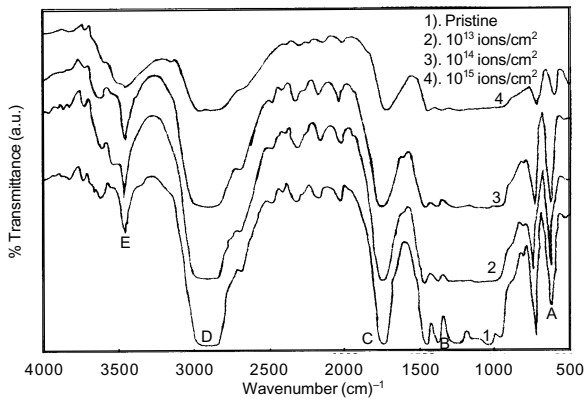

Figure 1. FTIR spectra of pristine and irradiated samples.

It is observed that there is no change in overall structure of the polymer but a minor change in intensities have been observed up to the fluence of 10^{14} ions/cm^2.The spectrum corresponding to 10^{15} ions/cm^2 indicates a significant change in intensities due to irradiation.

3.2. AC electrical frequency response :

AC conductivity measurement was performed for pristine and irradiated EVA films as shown in Figure 2. There is a sharp increase in conductivity at around 20 kHz frequencies in all cases and conductivity increases as fluence increases.

The increase in conductivity due to irradiation may be attributed to scissioning of polymer chains, resulting in an increase of free radicals, unsaturation *etc*. An AC field of sufficiently high frequency applied to a metal polymer metal structure may cause a net polarization, which is out of phase with the field. This result in AC

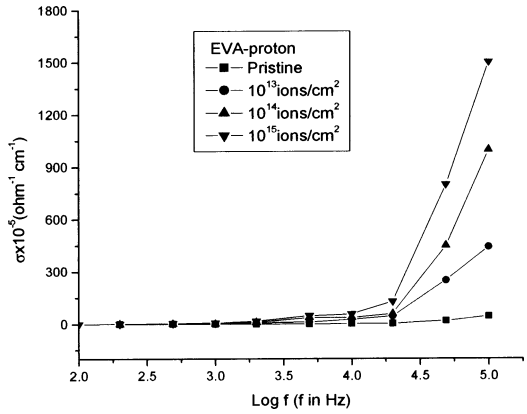

Figure 2. Conductivity *vs.* log *f* for pristine and irradiated samples.

conductivity, it appears at frequency greater than that at which traps are filled or emptied [14,15]. Figure 3 shows the plot of dielectric constant *versus* log frequency for pristine and irradiated samples. As evident from figure, the dielectric constant remains almost constant up to 100 kHz. At these frequencies, the mobility of free charge carriers is constant and so dielectric constant is constant.

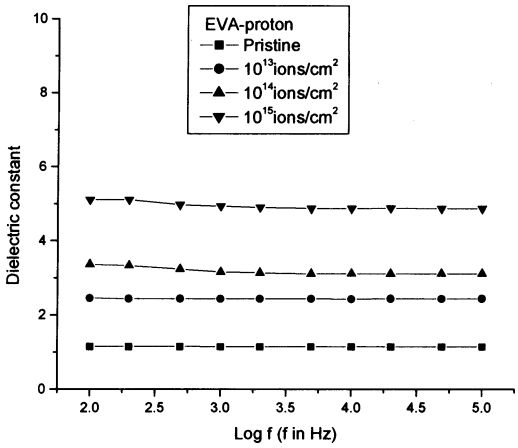

Figure 3. Dielectric constant *vs.* log *f* for pristine and irradiated samples.

It is also observed that dielectric constant increases as fluence increases. The increase in dielectric may be attributed to the chain scission and as a result increase of free radicals, *etc*. Figure 4 shows the plot of tan δ *versus* log frequency for pristine and irradiated films.

It reveals that tan δ drops sharply as frequency increases and becomes constant beyond a frequency of 1 kHz. This indicates that loss factor depends on frequency below 1 kHz and becomes constant beyond this frequency, suggesting that EVA films can be used as dielectric in capacitors being used above 1 kHz frequency. It is also

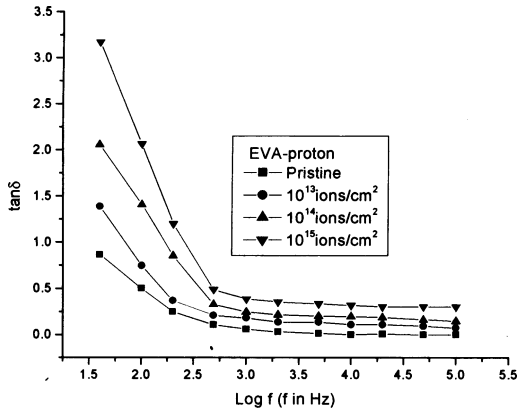

Figure 4. Tan δ *vs.* log *f* for pristine and irradiated samples.

observed that loss factor increases as fluence increases, tan δ has positive values indicating the dominance of inductive behaviour [15].

4. Conclusion

The FTIR spectra indicate that EVA gets chemically degraded at the highest fluence used. There is an exponential increase in conductivity with log frequency and the effect is significant at higher fluences. The loss factor and dielectric constant are observed to change significantly with the fluence. It is observed that EVA films can be used as dielectric in capacitors above a frequency of 1 kHz.

Acknowledgment

Authors are thankful to Cyclotron personal, Physics Department, Punjab University, Chandigarh for providing irradiation facilities.

References

[1] T Venkatesan, L Calcagno, B S Elman and G Foti *Ion beam modification of insulators* (eds) P Mazzoldi and G Arnold (Amsterdam : Elsevier) p301 (1987)

[2] D A Seanor *Electrical Properties of Polymers* (New York : Academic Press) **68** (1982)

[3] J Davenas, I Stevenson, N Celette, S Cambon, J L Gardette, A Rivaton and L Vignoud *Nucl. Instr. Meth.* **B191** 653 (2002)

[4] T N Gordiichuk and V P Gordineko *Kompoz. Polim. Mater.* **38** 33 (1988)

[5] M Mateev and S Karageogiev *Radial Phys. Chem.* **51** 205 (1998)

[6] S Jamaliah, H S S A A Sharifah and H Kamaruddin *Radial Phys. Chem.* **58** 191 (2000)

[7] S Dalai and C Wenxiu *J. Appl. Polym. Sci.* **86** 1296 (2002)

[8] S Dalai and C Wenxiu *J. Appl. Polym. Sci.* **86** 553 (2002)

[9] S Thomas, B R Gupta and S K De *Polym. Degrad. Stab.* **18** 189 (1987)

[10] L Minkova and M Nikolova *Polym. Degrad. Stab.* **23** 217 (1989)

[11] J Sharif, S H S A Aziz and K Hashim *Rad. Phys. Chem.* **58** 191 (2000)

[12] N L Singh, N Shah, C F Desai, K P Singh and S K Arora *Rod. Eff. Def. in Solids* **159** 475 (2004)

[13] Zeigler *SRIM-2003 The stopping range of ions in matter* (New York : IBM Research) (2003)

[14] A K Jonscher *Nature* **267** 673 (1977)

[15] N P Bogoroditsky, V V Pasynkov, B M Tareev *Electrical Engineering Materials* (Moscow : Mir Publisher) (1974)