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The impact of electron beam irradiation on Low density polyethylene and Ethylene vinyl acetate

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Abstract. Improvement of measured gel content, hardness, tensile strength and elongation at break of Ethylene vinyl acetate (EVA) have confirmed positive effect of electron beam irradiation on EVA.Results obtained from both gel content tests show that degree of cross-linking in amorphous regions is dependent on dose. A significant improvement in tensile strength of neat EVA samples is obtained upon electron-beam radiation up to 210 kGy. Similarly, hardness properties of Low-density polyethylene (LDPE) improvewith increasing electron beam irradiation. This article deals with the impacts of electron beam (EB) irradiation on the properties of LDPE and Ethylene-Vinyl Acetate (EVA) as the two common based formulations for wire and cable applications.

1. Introduction

Ethylene-Vinyl Acetate (EVA) copolymers are used extensively in wire and cable industry for making heat shrinkable insulation, semi-conductive insulation jackets, and flame retardant insulation. The ability to accept high filler loadings without embrittlement or loss of mechanical integrity, and ease of cross-linking made it be used extensively forpolymer products [1, 2]. However, unmodified EVA has some shortcomings which can be overcome by formation of intermolecular covalent bonds between polyethylene chains [3, 4]. In this study It was our intention to obtain comprehensive data from effects of absorbed dose on behavior of gel content, tensile strength, elongation at break, hot set, density, melting point, heat of melting, DSC thermograms, thermogravimetry analysis, dielectric constant, surface resistance, volume resistivity, break down voltage, dielectric loss factor, hardness (Rockwell) and heat-deformation percent for ethylene-vinyl acetate (EVA).The original contribution of this paper compared to other papers is that how physical, mechanical, electrical and thermal properties of EVA with 18% vinyl acetate content change when it is exposed against different absorbing dose rate by electron beam which these data are very suitable for insulation of all wire and cable research tests. Likewise, Low-density polyethylene (LDPE) has the largest annual production in world today. Low cost of LDPE, along with its excellent mechanical, processing, and electrical properties, makes it a good choice for many applications [5-8]. However, LDPE has also some shortcomings, such as low softening temperature, cold flow, and a tendency for stress cracking on contact with chemical reagents. These shortcomings can be overcome by formation of intermolecular covalent bonds between polyethylene chains [9-13]. Formation of a network structure changes thermoplastics into materials that no longer melt and will resist temperature-induced deformation or flow. Electron beam (EB) irradiation of polyethylene improves maximum permissible conductor temperature, maximum

allowable temperature in case of ‘‘over-current’’, soldering and application even at higher its softening point for a long period of time [14-16]. These approaches will be useful to use LDPE for different applications such as wire insulation, shrinkable tubes and packaging films [17].

2. Results and Discussion:

The gel content of irradiated EVA and LDPE were determined to evaluate content of crosslinking produced by irradiation.Figure 1 shows variation of gel content as a function of absorbed dose for electron-beam exposed of EVA and LDPE. Unirradiated EVA and LDPE were found to be completely soluble in hot xylene; however, solubility reduced significantly due to formation of three-dimensional networks in irradiated samples. The gel content of irradiated EVA and LDPEdue to their amorphous structures increased. In general, irradiation below melting point allows for cross-linking reactions to occur within amorphous part of polymer, whereas more radicals are likely to be trapped in crystalline phases [18]. The radical species formed within amorphous regions can be utilized readily in cross-linking reactions, whereas those radicals trapped in crystalline regions have a much lower mobility and a longer lifetime; such radicals can migrate slowly to amorphous region, where they subsequently react [19-25].

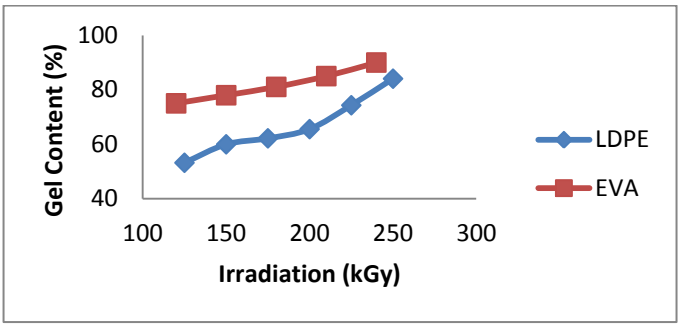

Figure 1 Gel Content of EVA and LDPE as a function of absorbed doses

Figure 2displays variation of density for LDPE and EVA at various absorbed doses. The density of unirradiated EVA and LDPE showed the highest value but increasing irradiation dose decreased density values. This reduction in density is attributed to reduction in crystallinity of LDPE and EVA upon irradiation [1-5].It was reported that increasing irradiation dose made crystalline size smaller and consequently extended amorphous region [6-10].The density of unirradiated EVA shows a slight reduction upon exposing to an electron beam at 120 kGy and then remains almost unchanged when increasing absorbed dose up to 240 kGy. This reduction in density is consistent with reduction in crystallinity of semicrystalline polymers [11-13]. The results obtained from DSC measurements demonstrate slight reductions in melting temperatures of irradiated EVAs. Such slight reductions with increasing dose suggest formation of crystalline regions with slightly reduced sizes. As pointed out earlier, crystalline regions may become slightly impaired due to branching and cross-linking in interfaces between amorphous and crystalline regions [26].

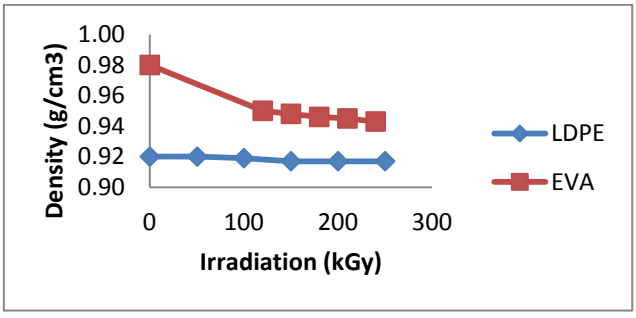

Figure 2 Density of EVA and LDPE as a function of absorbed doses

Figure 3 demonstrates variation of tensile strength of EVA and LDPEat various irradiation doses. The tensile strength of EVA shows a remarkable increase from 13 to about 35MPa. It was shown that upon irradiation tensile strength of EVA increased rapidly up to dose 200 kGy and remained then reduced to 33.5 MPa with further increase of radiation dose to 240 kGy. The reduction is most likely to be caused by chain scission at high dose. It can be understood that for achieving optimum mechanical properties from EVA irradiation dose should not exceed 210 kGy [11].The tensile strength of LDPE increased from 12 to 18.8MPa. Crosslinking and chain scission took place simultaneously over whole range of irradiation doses (0-250 kGy) and it seems that rate of cross-linking was much more dominated [4]. The tighter network in LDPE accounts for high tensile strengths of irradiated LDPE due to restricted movement of molecular chains [5]. Consequently, tensile strength values of LDPE increased after irradiation up to 250 kGy.

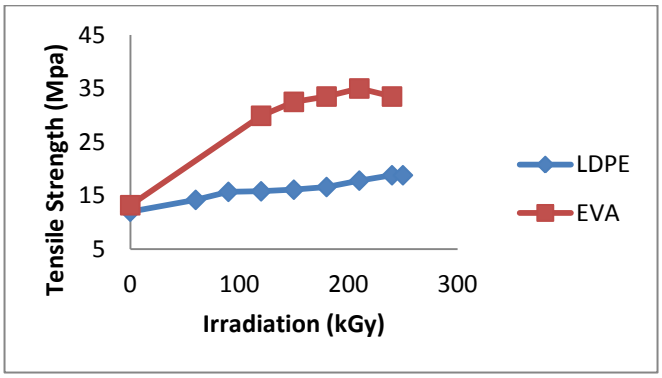

Figure 3 Effect of absorbed dose on tensile strength of EVA and LDPE

Fig. 4 shows relationship between elongation at break and irradiation dose of EVA and LDPE at different irradiation doses from 0 to 250 kGy.The tighter network in EVA samples and high tensile strengths of irradiated samples is due to restricted movement of molecular chains. This causes some losses in elongation of EVA, as shown in Figure 4. As dose increases more cross-links are produced in EVA matrix which prevents structural reorganization during drawing. Therefore by increasing three-dimensional and gel- like structures internal chain mobility and elongation decreases. It is obvious from Figure 4 that elongation at break of EVA was decreased by increasing of absorbed dose [10].Unirradiated LDPE shows the highest value of elongation at break (561%) whereas for all irradiated LDPE due to formation of three dimensional network structures and restricted movement of molecular chain elongation at break decreased sharply to 382% at 250 kGy.Increasing irradiation dose of EVA and LDPEcaused reduction of elongation at break and generated more crosslink bonds in polymeric matrix which prevents structural reorganization during drawing [8, 10-12].

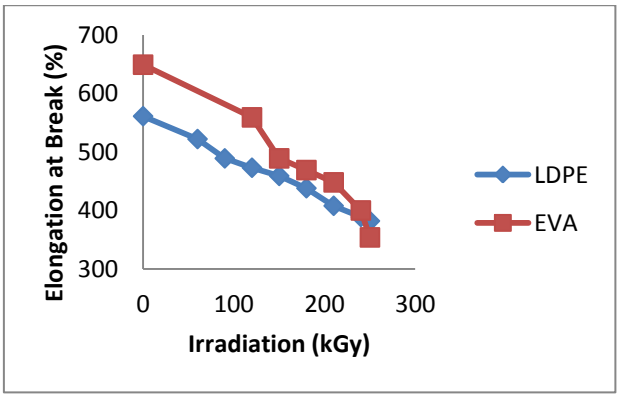

Figure 4 Effect of absorbed dose on Elongation at Break of EVA and LDPE

Figure 5 shows the dependence of hardness of LDPE and EVA with increasing irradiation dose. It was found that hardness of EVA increased gradually with increase irradiation dose up to 200 kGy which isin agreement with the resultshas been achieved for tensile strength test. Thus, it is evident that enhancement in hardness of EVA is due to formation of radiation cross-links as confirmed by gel content either.Figure 5 also displays that hardness of LDPE increased gradually with increase irradiation dose up to 250 kGy. The hardness value of unirradiated LDPE was measured 32 Rockwell L whereas increasing irradiation dose to 250 kGy increased hardness value to 37.8 Rockwell L. Thus, it confirms that enhancement in hardness of LDPE is due to formation of radiation induced crosslinks as confirmed by gel content and hot set tests. Generally, hardness is referred to resistance of material to local deformation [9], and results proved that irradiated LDPE and EVA were more resistance toward local deformation and increased hardness values.

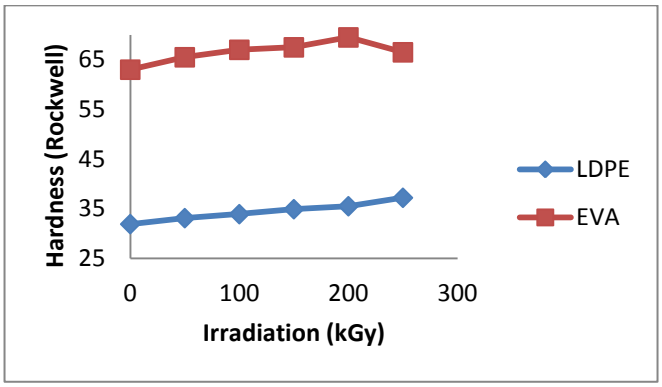

Figure 5 Effect of absorbed dose on Hardness of EVA and LDPE

3. Conclusion

This study investigated effects of electron-beam irradiation on different properties of EVA and LDPE. Cross-link density at a given irradiation dose depends on level of amorphous material in EVA and LDPE. Crystalline regions were only slightly affected by irradiation. Density of EVA decreased slightly with increasing absorbed dose, resulting from slight decreases in crystallinities of EVA. Tensile strength of EVA increased with increasing dose. Cross-linking of EVA increased as dose increased up to 210 kGy. Tensile strength of EVA increased when irradiated up to 210 kGy and decrease with further increase in dose. Hardness of irradiated EVA improved due to formation of radiation cross-links. Deformation of EVA at high temperature reduced with increase of irradiation dose.It has been proved that EVA samples are cross-linked and cross-linking increases with irradiation dose up to about 210 kGy. Mechanical and thermal properties of EVA samples are improved by irradiation and optimum irradiation dose is about 210 kGy. Dielectric constant and loss factor of EVA remain relatively constant and surface and volume resistance of samples were affected significantly by increasing radiation dose. Resistance of EVA increases when radiation dose increases up to 190 kGy. Increasing radiation dose over 190 kGy considerably reduces surface and volume resistance of EVA. Thermal degradation of EVA occurred through two steps as shown by thermogravimetric and derivative curves maximum rate 180 and 310 ℃. First step involves evolution of acetic acid and second degradation involves decomposition of polyene. Likely, reduction in densities, enthalpies of melting, enthalpies of crystallization, and melting points of LDPE with increase in irradiation dose clearly indicates that crystallinity of LDPE reduces upon irradiation. Thermal degradation of LDPE improves with increase in irradiationdose. A significant improvement in TS of neat LDPE samples obtains upon EB radiation up to 250 kGy with a concomitant decline in elongation at break. Surface resistance, VR, and dielectric strength reveal an approach of maximum value at 250 kGy irradiation dose. However, no considerable changes of dielectric constant and dielectric loss factor are observed with increasing irradiation dose. Enhancement in heat deformation, hardness, and thermal aging

properties of LDPE upon EB irradiation suggests that irradiated LDPE is more thermally and mechanically stable than virgin LDPE.

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