The Proceedings of **RAFSS 2007**
Regional Annual Fundamental Science Seminar 2007

Modified effects of LDPE/EVA blends by electron beam irradiation

Mazyiar Sabet, Azman Hassan, Mat Uzir Wahit

Faculty of Chemical and Natural Resources Engineering, University Technology Malaysia (UTM), 81310 Skudai, Johor Bahru, Malaysia

Abstract

The effect of electron beam irradiation on the properties of low density polyethylene (LDPE, LH0075) and ethylene vinyl acetate (EVA, with 18 %VA) blends were investigated. The improvement of the measured gel content, stress at ultimate, strain at Auto breaks and transition temperatures (Tg ,Tm) have confirmed the positive effects on blends but ineffective in density of blends.

Introduction

Blending is a well known method of modifying properties of polymer. It is an economically viable and versatile way to produce new materials and to over come deficiency in some material characteristics. There has been an impressive amount of research and development work carried out in radiation processing of polymer blends for electrical insulation purposes, Spenade et al. (1975) found that by blending 10-30 wt% rubber to polyethylene can produce a wire insulation that is more flexible and thus easier to install and splice. Low density polyethylene (LDPE, LH0075) can be radiation crosslinked without any addition of special additives or crosslinking agents. Radiation crosslinking makes this homopolymer harden and split easily at low temperature (Zhang Hui and Xu Jiufur 1993). On the other hand, ethylene vinyl acetate (EVA, with 18 %VA) has a rubbery property that is highly flexible. Ethylene vinyl acetate (EVA, with 18 %VA) can attain the same degree of crosslinking as LDPE but at lower dose (LDPE LH0075 at 210 KGY and EVA with 18%VA at 140 KGY). Therefore it is possible by blending certain amount of EVA with LDPE to improve the irradiation crosslinking performance of LDPE. The effect of electron beam irradiation and EVA content upon the gel-forming process in LDPE-EVA films have been studied by Mateev and Karageorgier (1998) and Jamalih, Sharifah together with Kamaruddin (1999). This paper will present some finding on the effect of electron beam irradiation on the properties of LDPE/EVA blends.

Experimental

i. Materials :

LDPE with density of 0.92 gr/cm^3 and melt flow index of 0.83 gr/ cm^3 and EVA with18% vinyl acetate content and melt flow index of 1.83 gr/10min were purchased from Iran(petrochemical complex of imam khomeiny) and China (attaché Tane elf company).

ii. Sample Preparation:

LDPE and EVA were blended together at ratios of 90/10, 80/20 and 70/30 by using Brabender Plasticorder Pl 2000 Model lab_ station TYPE 815801 at 50 ^{0}c and 130 RPM. The blended samples were then compression moulded into rectangular sheets (150x 200mm) of 2mm thickness under 1ton pressure.

iii. Irradiation:

Irradiation was carried out under air atmospheric by using a rhodotron 200TT, high voltage, 5-10MeV and 100x2 Kw power. Irradiation doses were selected in the range of 120-240 KGY and 30 KGY/pass.

iv. Gel Content:

Gel content was determined by refluxing the sample with xylene for 24 hr. The remaining insoluble sample was rinsed with methanol and dried in a vacuum oven at 70^{0}c to a constant weight. It is shown in Fig. 1 that gel content increased rapidly up to 200 KGY in all samples and then increase gradually with further increase in dose. Gel content increased by increasing EVA content in blends.

The highest gel content was for pure EVA .Burns (1979) proposed that part of the reason for superior crosslinking in EVA than the homopolymer. This finding was based on the continuing consensus (Lawton et., 1958; Patel and Keller, 1975) that at ambient conditions radiation crosslinking occurs mainly within the amorphous regions of a polyethylene. It is interesting to note that gel content of LDPE/EVA blend showed average value of gel content for pure LDPE and EVA.

v. Hot Set test:

Hot set test was done to measure the elongation of samples for static load at a fixed temperature. Measurements were carried out in an oven at 200^{0}c with normal air conditions. The sample loading was 20 N/cm^2 the elongation between two marks was determined after 15 min heating of the sample.

vi. Measurement of mechanical properties:

Tensile tests were carried out on dumbell shaped of specimens by using Instron tensile machine model 4411 with a crosshead speed of 50mm/min according to ASTM D-638.

Result and discussion

Gel content:

Fig.1 shows the relation ship between the gel content with irradiation for LDPE, EVA and LDPE/EVA blends. Unirradiated samples were found soluble in hot xylene, however upon irradiation they undergo crosslinking and as a result of network formation in the polymer, solubility of the samples was reduced significantly. It is shown in Fig.1 that gel content increases rapidly up to dose 200KGY in all samples and then increases gradually with further increase in dose. Increase in gel content in blends was enhanced by increasing EVA content. Gel content was highest for pure EVA. Burns (1979) propose that part of the reason for superior crosslinking in EVA than the homopolymer. This finding was based on the continuing consensus (Lawton et al., 1958; Patel and Keller, 1975) that at ambient conditions radiation crosslinking occurs mainly with in the amorphous regions of a polyethylene. It is interesting to note that gel content of LDPE/EVA blends showed average value of gel content for pure LDPE and EVA.

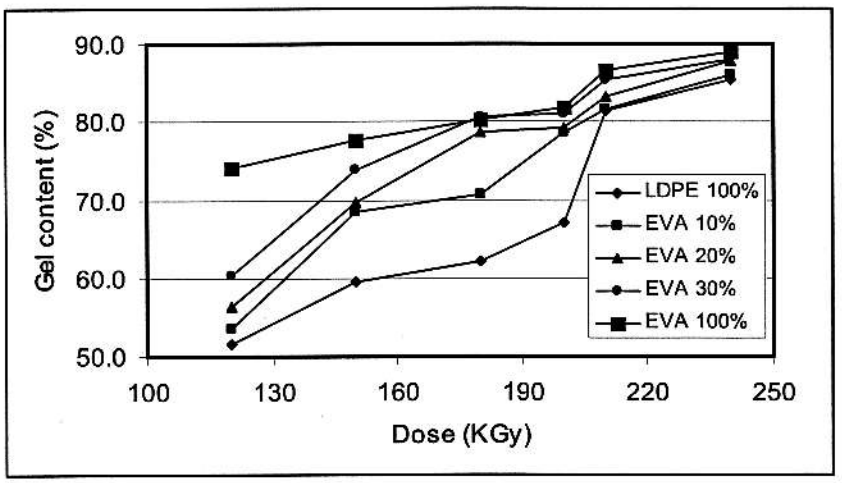

Fig 1. Effect of absorbed dose on gel content of LDPE, EVA and blends

i. Hot Set test:

Hot Set provides a quick check on crosslinking of samples and can be correlated with gel content. This test was carried out and the results are shown in table .1 it was found that Unirradiated samples failed the test immediately under elevated temperature 200^0c and load 20 N/cm^2 for 15 min. Irradiated samples up to 120 KGY still failed in this test but it

took more time. However, results of samples at 200 ^{0}c temperature improved as the dose increased. These achievements are due to formation more crosslinked polymer blend as evident by the gel content. It is obvious that EVA reaches a certain value of hot elongation at much lower dose than LDPE.

The performances of the blends rely on the amount of EVA in the blends. This behavior is believed to be due to the nature of EVA which more easily crosslinks when expose to electron beam. Wiesnner (1991) reported that different polymer structures require different specific numbers of crosslinking for certain reduction of deformation caused by the load in the hot set test. Formation of adequate radiation crosslinking net work in the polymer render the polymer better elongation at high temperature. It is shown that hot elongation value was noted when gel content value reached more than 75% for pure polymers as well as their blends.

Table 1. Hot Set test results a 200 ^{0}c under 20 N/cm^2 load for 15min

Irradiation Dose (KGY)	100% LDPE	90% LDPE-10% EVA	80% LDPE-20% EVA	70% LDPE-30% EVA	100% EVA
0	Fails immediately	Fails immediately	Fails immediately	Fails immediately	Fails immediately
30	Fails immediately	Fails immediately	Fails immediately	Fails immediately	Fails (15 sec)
60	Fails (0.5 min)	Fails (40 sec)	Fails (1 min)	Fails (1.5 min)	Fails (2 min)
90	Fails (0.75 min)	Fails (1 min)	Fails (1.5 min)	Fails (2 min)	Fails (3 min)
120	Fails (2 min)	Fails (135 sec)	Fails (148 sec)	Fails (247 sec)	Fails (6.5 min)
150	Fails (4 min)	Fails (5.5 min)	Fails (6.5 min)	Fails (7.5 min)	160%
180	Fails (4.5 min)	Fails (7.3 min)	208%	197.3%	78%
210	185%	183%	166.7%	146.3%	64.5%
240	147%	118.4%	117.95%	109%	45%

ii. Mechanical Properties:

Fig .2 illustrates the tensile strength, T_S as a function of irradiation dose. It was shown that upon irradiation T_S of LDPE increased rapidly up to dose 200 KGY and remained

constant with further increase of radiation dose to 240 KGY. It was reported by Burns (1979) that in order to get optimum mechanical properties from EVA the irradiation dose should not exceed 200 KGY. Tensile strength of the blends also increased with the dose and decreased after 210 KGY. The reduction is most likely to be caused by the chain scission at high dose. It is obvious that tensile strength of the blends was ascertained by the amount of EVA in the blends. Blend with high content of EVA showed high value of tensile strength.

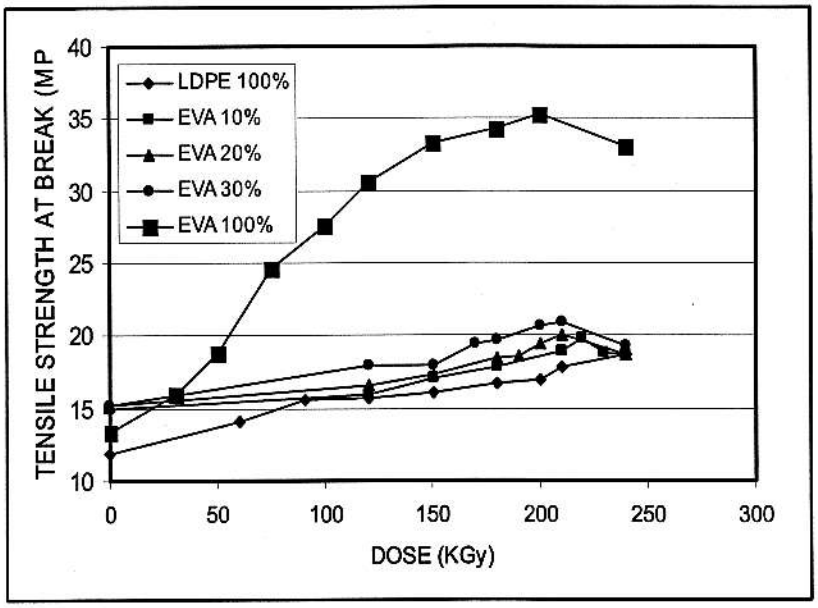

Fig 2. Effect of absorbed dose on tensile strength of LDPE, EVA and blends

Fig .3 shows the relationship between elongation at break and irradiation dose of the samples. Generally increasing irradiation dose results in reduction in elongation at break of the LDPE, EVA and blends. As the dose increases more crosslinks are produced in the sample matrix which prevents the structural reorganization during drawing (De Bore and Penning, 1983). Therefore by increasing three- dimensional and gel- like structures internal chain mobility and elongation decreases. . It is obvious from Fig.3 that elongation at break of the blends were ascertained by the amount of EVA in the blends. Blend with high content of EVA showed high value of elongation at break. As a

consequence elongation at break of blends has in agreement with the amount of EVA in blends.

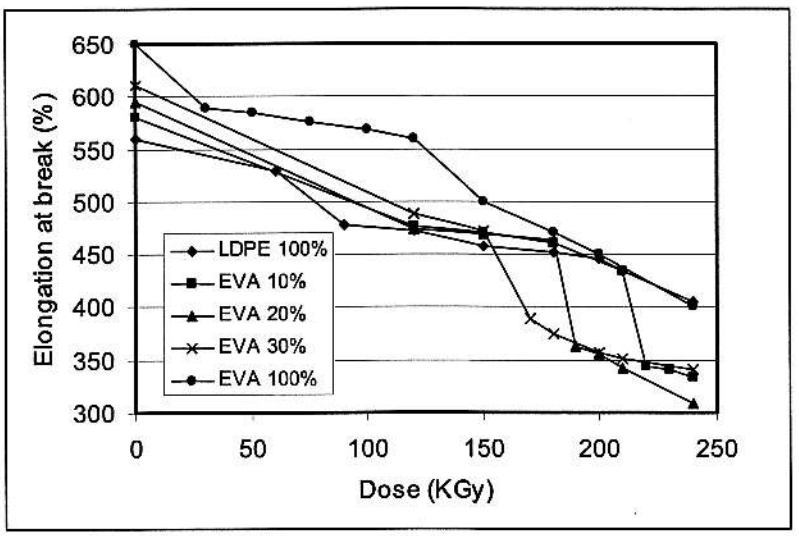

Fig 3.Effect of absorbed dose on elongation at break of LDPE, EVA and blends

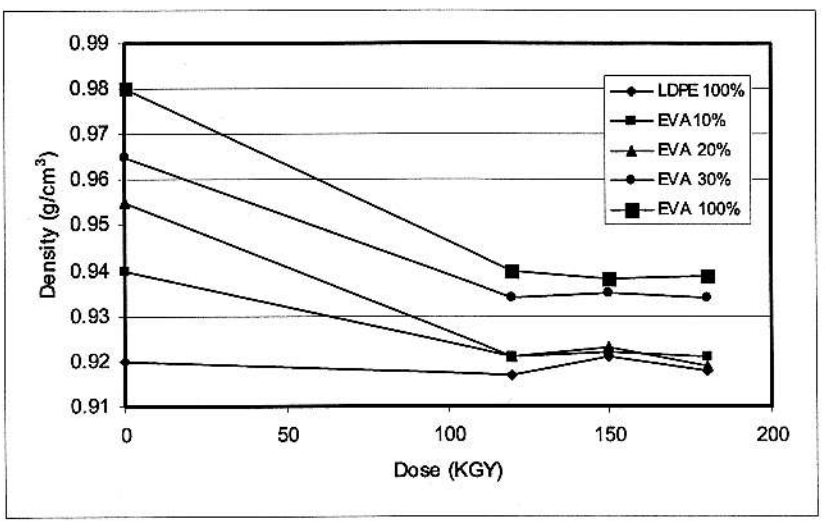

Fig 4.Effect of absorbed dose on density of LDPE, EVA and blends.

Fig .4 shows the plot of composition dependence of the density of the sample with increasing irradiation doses. It was found that at the beginning of irradiation the density of LDPE, EVA and blends gradually decreased (up to 120 KGY) but with further absorbed dose there is no significant changes. Fig.5 shows the plot of composition dependence of the hardness of the sample with increasing irradiation dose. In definition, hardness is generally referred to resistance of material to local deformation (Vishu Shah, 1984).

It was found that the hardness of LDPE increased gradually with increase irradiation dose up to 240 KGY. In agreement with the results of tensile strength the hardness of EVA increased up to 210 KGY and then decreased with further increased in radiation dose. Hardness of the blends depended on the polymer composition seemed to show slight increased in hardness. Thus it is evident that the enhancement in hardness of LDPE, EVA and blends is due to the formation of radiation crosslinks as confirmed by gel content.

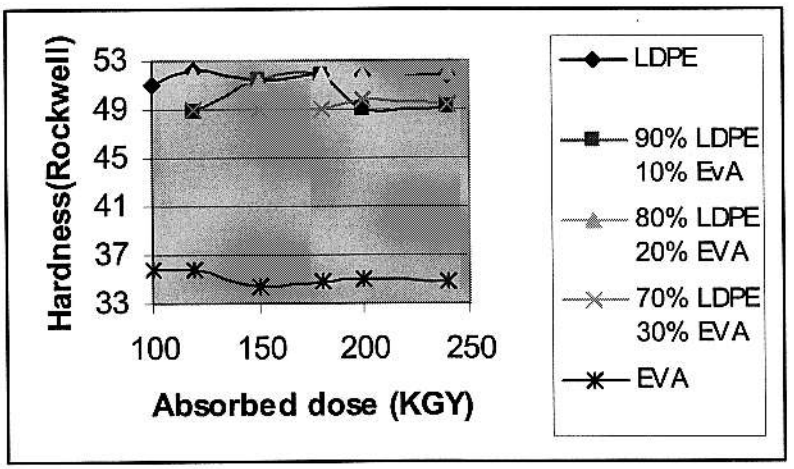

Fig 5. Effect of absorbed dose on hardness of LDPE, EVA and blends

Conclusion

This study has confirmed that electron beam radiation has positive effects on the properties of LDPE, EVA and blends. It was found that:

1. Crosslinking of the LDPE, EVA and blends increase as the dose increase up to 240 KGY. The amount of crosslinking in the blends increase as the composition of EVA increase.
2. The tensile strength of LDPE increased when irradiated up to 240 KGY. The tensile strength of EVA and the blends increased when irradiated up to 200 KGY and decreased with further increase in dose.
3. The density of LDPE, EVA and blends did not change significantly with increase irradiation dose up to 240 KGY.
4. The hardness of the irradiated samples improved due to the formation of radiation crosslinks. Hardness of EVA was found to be higher than that of LDPE whereas hardness of the blends depended on the polymer composition.

References

1. Burns.N.M."The radiation crosslinking of ethylene copolymers". Radiation physics and chemistry 14,797-808, 1979.
2. De Boer.J.Pennings.L.," The effect of pendant chains on the elasticity behavior of polyethylene networks". Colloid polymer science 261, 750, 1983.
3. Lawton. E.J.Balwit.J.S.Powell.R.S." Effect of physical state during the electron irradiation of hydrocarbon polymers. Part 1. The influence of physical state on reactions occurring in polyethylene during and following the irradiation". J.Polymer Science 32, 257, 1958.
4. Mateev.M.,Karageorgev.S." The effect of electron beam irradiation and content of EVA upon the gel forming process in LDPE-EVA film". Radiation physics and chemistry 51, 205, 1998.
5. Patel.G.N.Keller.A." Crystallinity and the effect of ionizing radiation in polyethylene. II crosslinking in chain folded single crystals". Gamma rays. J.Polym.Sci. Poly.Phys., Ed.13, 323, 1975.
6. Spenadel.L." Radiation crosslinking of polymer blends". Radiation physics and chemistry14, 683-697, 1979.
7. Spenadel. L.,Smith. W.S.,Murphy.,J.J" Crosslink able EPDM". Rubber Age 107,41,1975.
8. Vishu Shah"Hand book of plastic testing technology".Wiley,New York, p 79, 1984.
9. Wiesner.L." Effects of radiation on polyethylene and other polyolefins in the presence of oxygen" Radiation physics and chemistry 37, 77-81, 1991.
10. Zhang Hui. Xu Jiufu." The modification of the flexibility of radiation crosslinked PE by blending PE with EVA and CPE" Radiation physics and chemistry 42,117-119, 1993.
11. Jamaliah Sharif,Sharifah Hanisah Syed Abdul Abdul Aziz, Kamaruddin Hashim "Radiation effects on LDPE/EVA blends".