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Electron beam modification of thermoplastic elastomeric blends made from polyolefins

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Thermoplastic elastomeric films have been prepared from blends of ethylene vinylacetate copolymer (EVA) with low-density polyethylene (LDPE), high-density polyethylene (HDPE) and polypropylene (PP) by electron beam modification in presence of radiation sensitizer. X-ray diffraction (XRD) and differential scanning calorimetry (DSC) have been used to evaluate the structural changes of these blends. Scanning electron microscopy (SEM) indicates the morphological features of the blends. Significant improvements of mechanical, dynamic mechanical and set properties have been obtained by electron beam modification of the blends of EVA with LDPE and HDPE. However, in the case of EVA/PP, degradation of the PP phase has been observed. The effects of ditrimethylol propane tetraacrylate (DTMPTA) as radiation sensitizer have also been evaluated from the mechanical and dynamic mechanical properties. © *2001 Kluwer Academic Publishers*

1. Introduction

Judicious blending of two or more polymers is a novel method for producing new materials having unique set of properties. Plastic and rubber blends have been commercialized as rubber toughened plastics or thermoplastic elastomers (TPE) [1, 2]. In general, when a relatively large proportion of hard plastic is used in the blend the composition is known as impact resistant plastic: if a relatively large amount of rubber is used, the blend will be soft and have at least some of the properties of an elastomer [3, 4]. The mechanical and rheological properties of the blends depend not only on those of constituent polymers but also on the morphology of the blend [5]. It has been found that addition of small quantities of crosslinking agents during the mixing operation gives rise to a stable morphology and final properties of such blends can be improved without affecting much on processing characteristics. This type of crosslinking is known as dynamic vulcanization [4–8]. Dynamically vulcanized thermoplastic elastomers have been prepared from this laboratory [9, 10]. It is also observed that an optimum quantity of crosslinking agent is required for the best compromise of properties.

Considerable work has been done in the past evaluating polymer property changes due to electromagnetic radiation [11–13]. Electron beam modification of polymers has certain advantages over conventional grafting and crosslinking process such as absence of catalyst residue, a solvent free system and uniform crosslinking with accurate reproducibility. Different authors have reviewed the treatment of polymers by electron beam irradiation. It has been observed that some polymers like, natural rubber, polyethylene, ethylene propylene rubber can be crosslinked with radiation, whereas some other polymers like polypropylene and polyvinyl chloride have a tendency to degrade [14–21]. As a result, in many of these references, a radiation sensitizer, which will plasticize the polymer and reduce the radiation dose level so as to minimize degradation, has been used.

The radiation compatibilization of binary systems has rather rarely been investigated [22–25]. Radiation modification, in particular for TPEs here offers an opportunity to considerably improve the range of material properties, retaining all the advantages of thermoplastic processibility.

In our earlier communications, we have demonstrated the effects of irradiation dose and sensitizer levels upon the modification of thermoplastic elastomeric blends of low-density polyethylene (LDPE) and ethylene vinyl acetate copolymers (EVA). Modification of the phases of the blends was explained from the changes in mechanical, dynamic mechanical and electrical properties. The advent of electron beam technology as an alternative to dynamic vulcanization process was also indicated [26–28].

The objective of the present work is to evaluate the effect of the variation of plastic components on the electron beam modification of thermoplastic elastomeric films with reference to its mechanical, dynamic mechanical properties, morphology and reprocessibility. The elastomeric part of the blends is EVA containing 45% vinyl acetate. The thermoplastic components have been varied, e.g., LDPE, HDPE and PP. In order to get a comparative view, blend ratio, irradiation dose and

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TABLE I Materials details

Polymer	Grade	Density (kg/m^3) (ASTM D792)	MFI (g/10 min) (ASTM D1238)	Source
EVA 45	LEVAPRENE 450	975	–	Bayer, Germany
LDPE-1	INDOTHENE MA 400	918	30	IPCL, India
LDPE-2	INDOTHENE 24F S040	922	4.0	IPCL, India
HDPE	PILENE PM 4577F	950	0	NOCIL, India
PP	Profax 8523	901	4.0	Himant USA Inc., USA

sensitizer level have been fixed, based on our earlier studies [28].

2. Experimental

2.1. Materials

The polymers used for the study were EVA (45% vinyl acetate), LDPE, HDPE and PP respectively. The details of these materials are given in Table I. Ditrimethylol propane tetraacrylate (DTMPTA, Ebecryl 140) as radiation sensitizer, was obtained from UCB Chemicals, Belgium.

2.2. Preparation of samples

In our earlier studies with LDPE-1 and EVA 45, it was indicated that a 60 : 40 (wt/wt) blend ratio of rubber to plastic gave the best thermoplastic elastomer [26–28]. So, keeping these in mind, all the blends were prepared on a ratio of 60 : 40 rubber to plastic. DTMPTA at a level of 1 wt% (optimized in earlier studies) was used.

EVA 45 and Plastics (LDPE, HDPE and PP) were mixed in a Brabender Plasticorder PLE 330 at 130°C, 170°C and 190°C for LDPE, HDPE and PP based blends respectively at a rotor speed of 60 rpm. The plastic was first allowed to melt for 2 min., followed by EVA 45 and DTMPTA for a total mixing time of 4 min. The mixes so obtained were sheeted out under conditions through the open mill set at 2 mm nip gap. It was remixed in the Brabender Plasticorder for another 2 min. at the same temperature.

The sheets were compression molded between Teflon sheets for 2 min. at a temperature of 20°C above the mixing temperatures and at a pressure of 5 MPa in an electrically heated press to obtain films of thickness 2.5 ± 0.5 mm. The moldings were cooled under compression to maintain the overall dimensional stability.

2.3. Irradiation of samples

Our earlier studies indicated that with or without 1 wt% DTMPTA, 20 kGy irradiation dose gave the best thermoplastic elastomeric properties, e.g., permanent set and reprocessibility [26–28]. Hence, the molded films were irradiated in air at room temperature of 25 ± 2°C by an electron beam accelerator (Model ILU-6) under forced air cooling at the Bhabha Atomic Research Centre (BARC), Mumbai, India at a radiation dose of 20 kGy. The specification of the accelerator was given in earlier communications [18–19].

2.4. X-ray diffraction analysis

The X-ray diffraction patterns of the films were recorded with a Philips X-ray Diffractometer (type PW-1840) using crystal monochromated Co K_α radiation in the angular range 10–40° (2θ) and at an operating voltage of 40 kV and current of 20 mA. The degree of crystallinity (X_c) and crystallite size (P) were calculated as follows:

$$X_c = \frac{I_c}{I_a + I_c} \tag{1}$$

and

$$P = \frac{k \cdot \lambda}{\beta \cos\theta} \tag{2}$$

where I_a and I_c are the integrated intensity corresponding to amorphous and crystalline phases respectively, β is the half height width (in radian) of the most intense crystalline peak and λ is the wavelength of the X-ray radiation used (0.179 nm) and k is the Scherrer constant taken as 0.9 [29]. The results reported here were based on the average of three experiments.

2.5. Differential scanning calorimetry (DSC)

DSC studies were carried out using Du Pont 9100 thermal analyzer at a heating rate of 20°C/min. in nitrogen atmosphere in the temperature range of -150 to 200°C. The transitions were determined from the discontinuity of the thermogram [30].

2.6. Scanning electron microscopy (SEM)

Scanning electron micrographs of the blends were taken with a scanning electron microscope (JEOL, Model JSM 5800). The accelerating voltage was 15 kV. The elastomeric phases of the samples were etched out using methyl ethyl ketone (MEK) for uncrosslinked samples. The samples were gold coated prior to SEM studies [31].

2.7. Mechanical and dynamic mechanical properties and reprocessibility studies

Tensile and tension set specimens were punched out from the molded films using ASTM Die-C. The tests were carried out as per ASTM D412-98a method using a Universal Testing Machine (Zwick 1445) at a crosshead speed of 500 mm/min. at 25°C. The average of three tests is reported here.

Dynamic mechanical thermal properties were evaluated on DMTA-II, Rheometric Scientific Inc. under dual cantilever mode. The experiments were carried out at a frequency of 1 Hz. The measurements were taken from -60 to $+60$°C at a heating rate of 2°C/min. and a double strain amplitude of 64 μm. The storage modulus and the loss tangent (tan δ) were measured

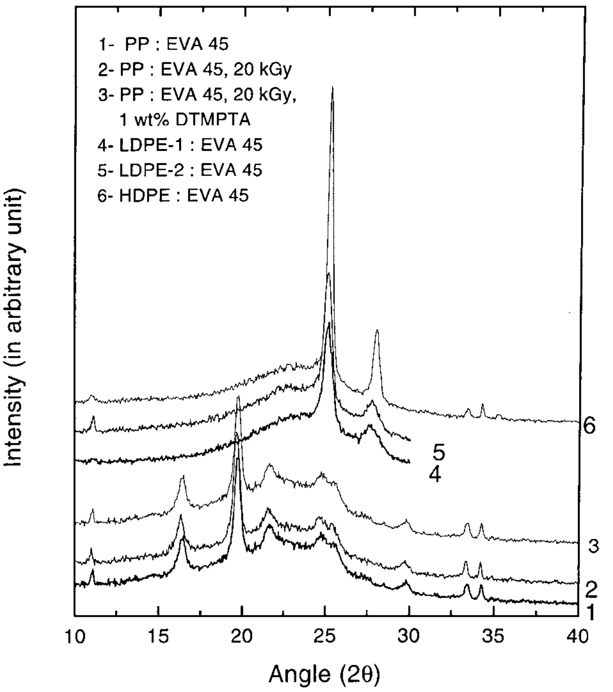

Figure 1 X-ray diffraction patterns of irradiated and unirradiated blends.

for all samples under identical conditions. The data were analyzed using ORIGIN 4.0 software.

Reprocessibility studies were performed on the samples by repeated mixing and molding of the samples and measuring their tensile properties. The experiments were carried out for three such mixing and molding cycles for each sample.

3. Results and discussion

3.1. X-ray diffraction

Fig. 1 shows the X-ray diffraction pattern of 60 : 40 blend of EVA 45 and LDPE (two different grades), or HDPE or PP respectively. In the case of the blends containing EVA 45 with LDPE and HDPE, there are mainly two peaks in the angular range of 12.4–12.6° and another between 13.8–14.0° respectively corresponding to the (110) and (200) planes of the orthorhomic unit cell. In the case of the PP blends, the X-ray diffraction gives rise to a number of peaks corresponding to the planes of the monoclinic unit cell, e.g., peaks in the angular ranges 8.2–8.3°, 9.8–9.9°, 10.8–10.9° and 12.4–12.5° corresponding to (110), (130), (111) and (131) planes respectively [29]. On the other hand, EVA 45 contributes only to the amorphous portion in the blend. Percent crystallinity and crystallite size (*P*) were calculated with respect to all the peaks in the case of PP based blends and for PE based blends the two high intensity peaks were considered and are reported in Table II. Percent crystallinities in all the cases are more or less the same as compared to the crystallinities calculated from the additivity rule indicating that these blends have a high degree of incompatibility, so that the plastic phase retains its identity in the blend.

The crystallite size also remains same upon blending, and also supports the same fact [30]. Upon irradiation, (20 kGy) or incorporation of DTMPTA (1 wt%), the percent crystallinity or crystallite size does not change to a significant extent in the case of the blends of EVA with LDPE and HDPE which indicates that the irradiation effect at low irradiation does (20 kGy) is limited to the amorphous portion of the concerned blends. A similar effect has been reported in an earlier communication [27].

However, in the case of PP and EVA blend, both crystallinity and crystallite size decreased upon irradiation. No significant change in crystallinity and crystallite size are observed in the case of the blends containing 1 wt% DTMPTA and irradiated at 20 kGy compared to the control blend. Polypropylene degrades upon electron beam irradiation [23]. However, compared to the pure PP, the decrease in crystallinity due to irradiation (not

TABLE II X-ray diffraction results

	Peak centre (θ)		Percent crystallinity (%)		Crystallite size (nm)	
Blend/Polymer	Low angle	High angle	Experimental	Theoretical	Low angle	High angle
LDPE-1 : EVA 45						
0 kGy	12.5	13.9	18	18	19.0	8.4
20 kGy	12.6	14.0	18	–	18.8	8.5
20 kGy, 1 wt% DTMPTA	12.5	13.8	18	–	19.0	8.5
LDPE-2 : EVA 45						
0 kGy	12.6	14.0	19	19	21.0	8.6
20 kGy	12.6	14.0	19	–	21.2	8.6
20 kGy, 1 wt% DTMPTA	12.5	13.9	19	–	21.0	8.7
HDPE : EVA 45						
0 kGy	12.6	14.0	24	24	27.0	9.1
20 kGy	12.6	14.0	24	–	27.0	9.1
20 kGy, 1 wt% DTMPTA	12.6	14.0	24	–	26.5	9.1
PP : EVA 45						
0 kGy	8.3	9.9	16	17	5.7	7.2
20 kGy	8.2	9.8	14	–	4.6	6.9
20 kGy, 1 wt% DTMPTA	8.3	9.9	16	–	5.6	7.3
LDPE-1	12.4	13.8	44	–	19.0	8.5
LDPE-2	12.5	13.8	47	–	21.0	9.0
HDPE	12.6	14.0	60	–	27.6	10.0
PP	8.3	9.9	42	–	6.0	7.8

shown) in the case of PP/EVA blends is less, which proves the fact that degradation of PP in the crystalline phase is suppressed to a certain extent in the presence of EVA. Sensitizers like DTMPTA decrease the probability of chain scission and degradation. Hence, both the percent crystallinity and crystallite size does not change upon irradiation in the case of blends containing DTMPTA.

3.2. DSC studies

DSC experiments of the blends have been done in the temperature range −150 to 200°C. There are mainly three transitions observed for all the blends, e.g., β-transition at −25 to −18°C (due to movement of the branch points), γ-transition at −130 to −120°C (Crankshaft motion) and melting endotherms (106 to 160°C). On incorporation of the blend component or irradiation, the γ-transition does not change to a significant extent. Hence, the change of β-transitions and melting endotherms are shown in Fig. 2 for all the blends.

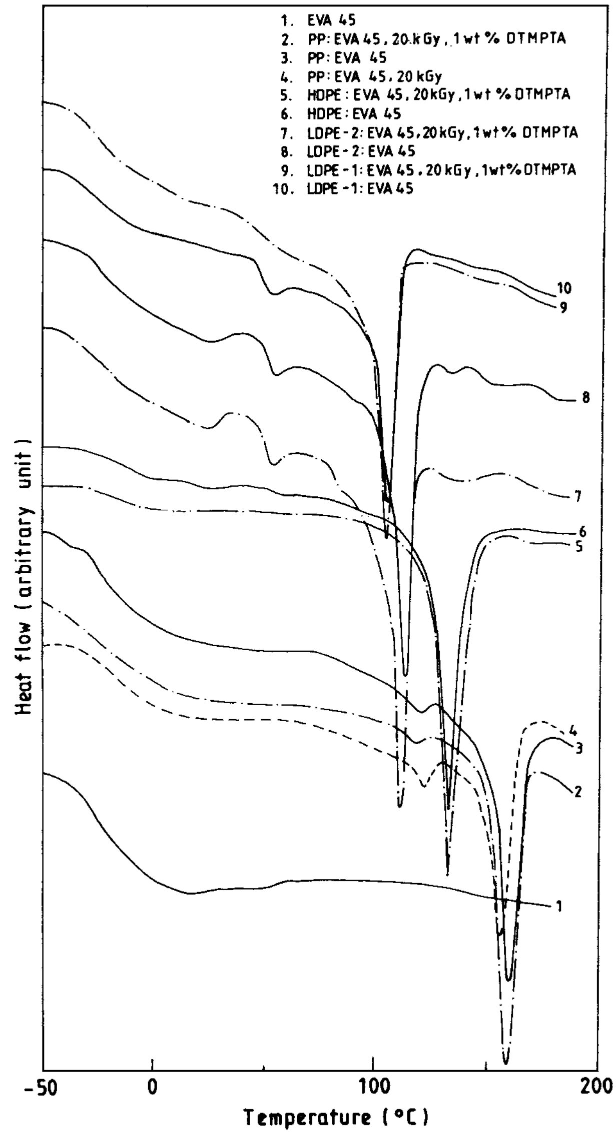

Figure 2 DSC thermogram of EVA 45 and its irradiated and unirradiated blends.

EVA 45 shows a sharp β-transition due to the movement of the vinyl acetate branch points at −24°C. LDPE or PP on the other hand, does not show a clear β-relaxation (not shown in the figure). A broad β-relaxation is observed in all the blends at −18 to −20°C, which may be treated as the glass transition temperature of the blend. In the case of the blends containing LDPE and PP, a broad hump appears at an intermediate temperature before the melting endotherm signifies the polydispersion of crystal size [29]. In the blends, the melting temperature (T_m) increases expectedly from 106 to 160°C from LDPE-1 to PP. Hence, their temperature range of application increases upon changing the plastic component of the blend.

Upon irradiation to 20 kGy, no significant change has been observed in the case of the blends containing LDPE and HDPE. At an irradiation dose of 20 kGy, only small amounts of crosslinks are formed in the amorphous portion of the blends [26–28]. Hence, the melting endotherm and the β-transitions do not change upon irradiation.

However, in the case of the blend containing PP, the melting temperature decreases from 160 to 157°C due to irradiation, which signifies a small amount of degradation in the crystalline portion.

In all the blends containing 1 wt% of DTMPTA and irradiated at 20 kGy, no change in melting endotherms has been observed compared to the control blends. The β-relaxation increases slightly. Sensitizers like DTMPTA increases the efficiency of crosslinking of electron beam radiation at lower irradiation doses thereby eliminating the probability of degradation [26–27]. This is obvious in the case of blends of LDPE and PP with EVA.

3.3. SEM studies

Fig. 3 shows the morphology of the blends of EVA 45 with LDPE-1, LDPE-2, HDPE and PP respectively. The rubber portion of the blends is etched out with MEK. In all the cases, the blends display hetero-phase morphology. In the cases of the blends containing LDPE and PP, thermoplastic portions of the blend form the continuous phase, due to high MFI and low melt viscosity at mixing temperatures compared to EVA 45, although the blends contain more rubber. However, in the case of HDPE/EVA blend, a coarser and co-continuous morphology is resulted due to the lower MFI and inferior flow characteristics of HDPE compared to the other thermoplastics used.

3.4. Mechanical properties

Tensile stress vs. elongation plots of the blends without irradiation (inset) and with radiation doses are depicted in Fig. 4. Tensile strength, modulus, elongation at break and permanent set data are given in Table III. In the case of the blends containing LDPE and HDPE with EVA, the mechanical properties are improved upon irradiation and incorporation of DTMPTA in the blends. The improvements in mechanical properties due to

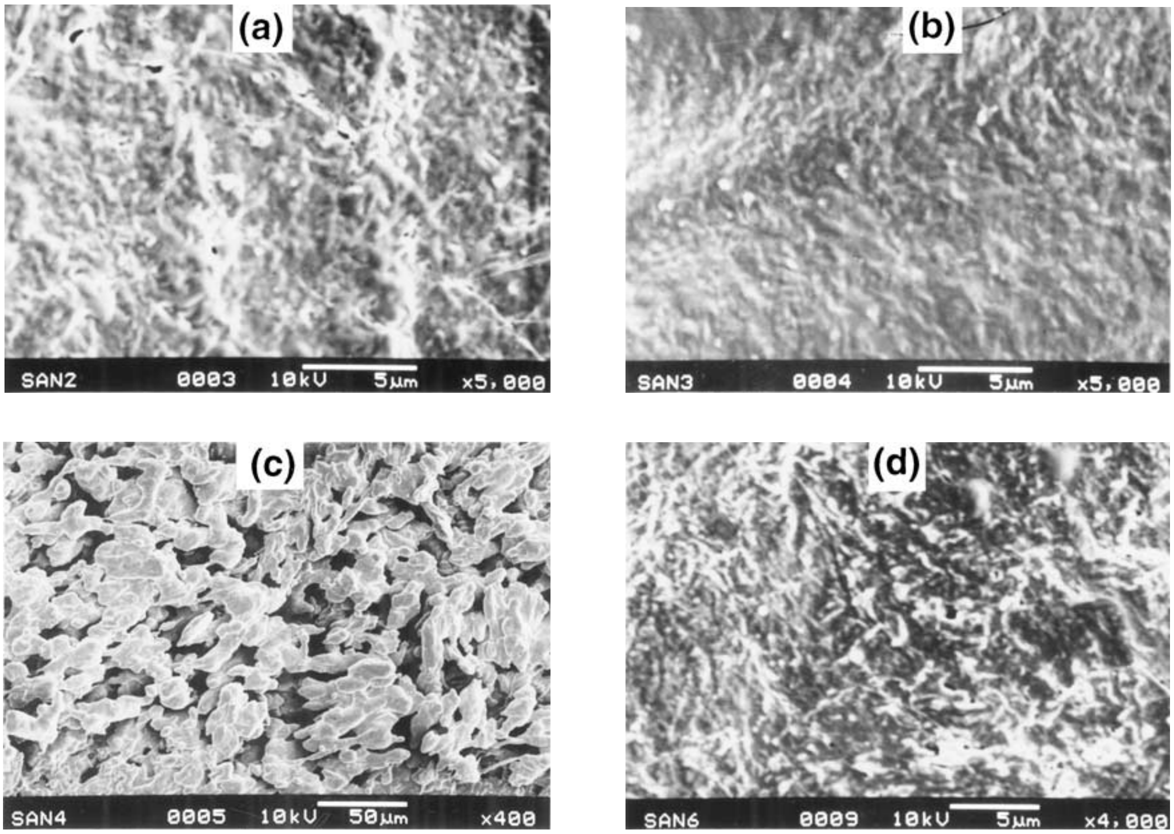

Figure 3 SEM micrographs of solvent etched. (a) LDPE-1 and EVA, (b) LDPE-2 and EVA, (c) HDPE and EVA, (d) PP and EVA.

irradiation (20 kGy), are not very significant for the blends containing relatively rigid plastics (HDPE). However, in presence of DTMPTA, mechanical properties are markedly increased upon irradiation. The permanent set of all these modified blends comes within the specification of a thermoplastic elastomer.

On the other hand, in the case of the blend containing PP and EVA, the mechanical properties decrease significantly upon irradiation (20 kGy). This may be due to the degradation, both in the crystalline and amorphous portion of the hard plastic phase of the blend [23]. However, the blend of PP and EVA containing 1 wt%

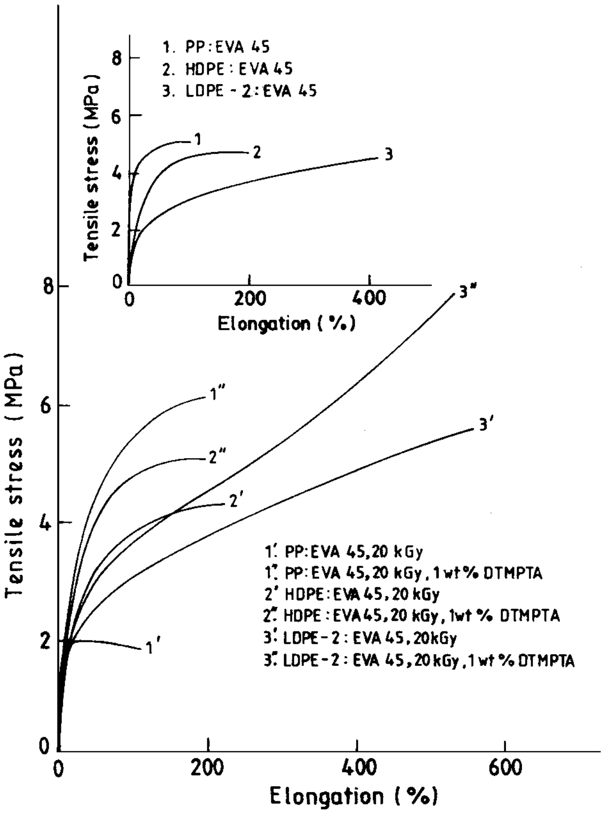

Figure 4 Stress vs. elongation plots of control and irradiated blends.

TABLE III Mechanical properties of the blends

Sample	Modulus (MPa) 100%	200%	300%	T.S. (%) (MPa)	E.B. (%)	P.S. (%)
LDPE-1: EVA 45						
0 kGy	2.92	3.41	3.85	4.1	421	28
20 kGy	3.21	3.57	3.95	5.1	560	23
20 kGy, 1 wt% DTMPTA	3.65	3.69	4.15	5.8	532	20
LDPE-2: EVA 45						
0 kGy	2.95	3.41	3.85	4.5	410	28
20 kGy	2.99	3.72	4.36	5.7	550	24
20 kGy, 1 wt% DTMPTA	3.36	4.46	5.37	7.7	530	16
HDPE: EVA 45						
O kGy	3.46	–	–	4.1	178	30
20 kGy	3.56	4.06	–	4.3	237	20
20 kGy, 1 wt% DTMPTA	4.92	–	–	5.4	196	18
PP: EVA 45						
O kGy	4.10	–	–	4.9	180	10
20 kGy	1.86	–	–	2.0	141	14
20 kGy, 1 wt% DTMPTA	5.50	5.80	–	6.2	202	8

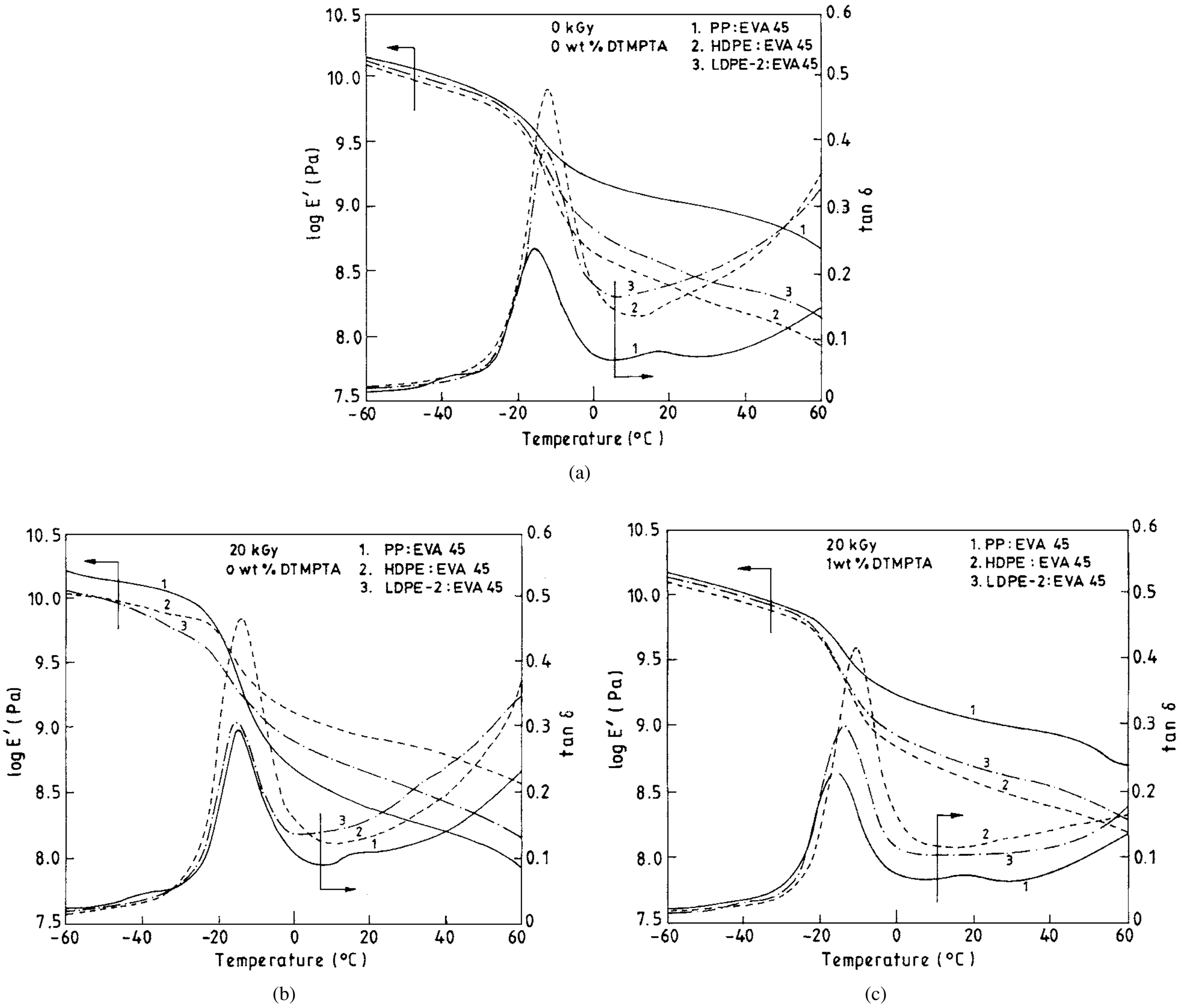

Figure 5 Log E' vs. temperature and tan δ vs temperature plots of blends: (a) unirradiated, (b) irradiated (20 kGy) without DTMPTA, (c) with 1 wt% DTMPTA and irradiated at 20 kGy.

DTMPTA registers an increase in mechanical properties and permanent set upon irradiation. This proves the radiation sensitizing effect of DTMPTA.

TABLE IV Dynamic mechanical properties of the blends

	Log (E') at			Tan δ at		
Blend	T_β^*	25°C	50°C	T_β^*	25°C	50°C
LDPE-1 : EVA 45						
0 kGy	9.411	8.794	8.634	0.350	0.160	0.250
20 kGy	9.557	8.814	8.500	0.309	0.158	0.204
20 kGy, 1 wt.% DTMPTA	9.500	8.800	8.671	0.241	0.087	0.098
LDPE-2 : EVA 45						
0 kGy	9.269	8.471	8.304	0.381	0.164	0.263
20 kGy	9.308	8.598	8.314	0.312	0.174	0.290
20 kGy, 1 wt.% DTMPTA	9.390	8.647	8.421	0.300	0.102	0.138
HDPE : EVA 45						
0 kGy	9.151	8.250	8.083	0.471	0.164	0.263
20 kGy	9.278	8.310	8.111	0.466	0.145	0.237
20 kGy, 1 wt.% DTMPTA	9.147	8.501	8.305	0.419	0.123	0.150
PP : EVA 45						
0 kGy	9.573	8.984	8.805	0.234	0.066	0.109
20 kGy	9.485	8.904	8.679	0.298	0.117	0.180
20 kGy, 1 wt.% DTMPTA	9.600	8.970	8.830	0.225	0.061	0.094

*T_β is β-transition temperature.

3.5. Dynamic mechanical properties

Dynamic mechanical thermal analysis (DMTA) of the control and irradiated thermoplastic elastomeric films are measured over a temperature range of −60 to 60°C at a frequency of 1 Hz. Fig. 5a–c show the log E' vs. temperature and tan δ vs. temperature plots of the control blends, irradiated blends at 20 kGy without and with DTMPTA respectively. The results are given in Table IV.

In the case of the blends of LDPE and HDPE with EVA, there is only one lower temperature transition corresponding to the β-relaxation [32]. At higher temperature, the low melting crystallites start melting giving rise to a decrease in storage modulus and increase in loss tangent. In the case of the blend of PP with EVA, apart from the β-relaxation of EVA (−18°C), a small transition peak appears at −39°C due to the movement of the $-CH_3$ group of PP. Another transition appears at around 17°C due to the movement in the crystalline features of PP. These signify the incompatible nature

TABLE V Reprocessibility studies

	Initial properties			1st Reprocessing cycle			2nd Reprocessing cycle			3rd Reprocessing cycle		
Sample	100% Mod. (MPa)	T.S. (MPa)	E.B. (%)	100% Mod. (MPa)	T.S. (MPa)	E.B. (%)	100% Mod. (MPa)	T.S. (MPa)	E.B. (%)	100% Mod. (MPa)	T.S. (MPa)	E.B. (%)
LDPE-1 : EVA 45												
0 kGy	2.92	4.1	421	2.90	3.9	410	2.85	4.0	400	2.88	4.1	390
20 kGy	3.21	5.1	560	3.21	5.0	536	2.85	5.1	450	3.27	4.9	420
20 kGy, 1 wt.% DTMPTA	3.25	5.8	535	3.30	5.4	470	3.35	4.9	396	3.45	4.5	370
LDPE-2 : EVA 45												
0 kGy	2.95	4.5	410	2.98	4.6	400	3.00	4.4	405	3.02	4.2	380
20 kGy	2.99	5.7	550	2.99	5.5	522	3.05	5.4	400	3.10	5.2	380
20 kGy, 1 wt.% DTMPTA	3.36	7.7	530	3.48	7.7	450	3.50	7.3	380	3.55	7.1	350
HDPE : EVA 45												
0 kGy	3.46	4.1	178	3.34	3.7	150	3.30	3.5	130	3.28	3.4	100
20 kGy	3.56	4.3	237	5.26	5.5	120	–	5.9	55	–	6.0	45
20 kGy, 1 wt.% DTMPTA	4.92	5.4	196	–	6.0	83	–	7.8	47	–	8.0	38
PP : EVA 45												
0 kGy	4.10	4.9	180	4.20	4.7	165	4.25	4.8	140	4.20	4.7	130
20 kGy	1.86	2.0	141	2.09	2.2	105	–	2.0	90	–	1.9	70
20 kGy, 1 wt.% DTMPTA	5.5	6.2	202	5.60	5.7	157	5.70	5.9	138	5.75	5.8	120

of the blend. However, in the case of the LDPE/EVA blends, only one β-transition is prominent in both the cases and they appear almost at the same temperature. The major relaxation peak at around -18 to -12°C can be considered as the glass transition or β-transition temperature of the blend.

In the case of HDPE/EVA blends, the lowest storage modulus and high loss have been observed which can be explained from its coarse and co-continuous morphology. PP/EVA blends gives rise to highest storage modulus and low loss due to the rigidity of its hard phase (PP). Among two different LDPEs, the blends of LDPE-1 with EVA gives higher modulus and lower loss tangent due possibly to its finer microstructure.

In the case of the blend containing LDPE and HDPE with EVA an increase in storage modulus and a decrease in loss tangent have been observed due to irradiation. In the presence of DTMPTA, the extent of improvement of dynamic mechanical properties is very prominent in the case of HDPE/EVA blend.

For PP/EVA blend, a decrease in storage modulus and an increase in loss tangent are observed due to irradiation, which is possibly due to the radiation induced damage of the PP phase. However, in the case of the blend containing DTMPTA, improved dynamic mechanical properties are obtained for the PP/EVA blend.

3.6. Reprocessibility studies

Reprocessibility studies have been performed on the samples for three mixing and molding cycles. The results are given in Table V. It is observed that all the irradiated or unirradiated blends containing LDPE and EVA are reprocessible. The reprocessibility of the blend containing HDPE (MFI 0 g/10 min.) is inferior due to its coarse microstructure and inferior flow characteristics. Upon irradiation with or without DTMPTA, its reprocessibility becomes very poor. On the other hand, the blends containing PP (MFI 4 g/10 min.) and EVA show relatively fair reprocessing characteristics. Hence it can be concluded that the reprocessibility of the irradiated blends largely depend on their flow characteristics and morphology. In all the cases of the blends containing DTMPTA, the reprocessibility is comparatively poor possibly due to the formation of a larger number of networks.

4. Conclusions

Thermoplastic elastomeric blends of EVA with LDPE (two different grades), HDPE and PP have been prepared by electron beam modification and the following conclusions are drawn:

(1) XRD and DSC studies indicated that all the blends studied are incompatible in nature. Irradiation effects are limited to the amorphous portion of the blends of EVA with LDPE and HDPE, however small changes have been observed in the crystalline portion of blends containing EVA and PP.

(2) SEM study indicated a hetero-phase morphology containing plastic as a continuous matrix due to its higher MFI and lower melt viscosity at mixing temperatures. The blend of HDPE/EVA on the other hand forms a coarser and co-continuous morphology.

(3) Mechanical and dynamic mechanical properties and permanent set properties of the blends of EVA with LDPE and HDPE with or without DTMPTA are improved by electron beam irradiation. The blends obtained from PP and EVA on the other hand, probably degrade in the presence of irradiation giving rise to marked decrease in the above properties. However, in the presence of DTMPTA significant improvements of those properties are observed.

(4) Electron beam modified films obtained from the blends of EVA with LDPE and PP show good reprocessing characteristics and the reprocessibility

depends on the flow characteristics and morphology of the blends. Electron beam modified EVA/HDPE blends on the other hand, are not reprocessible and hence cannot be treated as thermoplastic elastomer.

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