Original Article

JOURNAL OF biomaterials applications

Journal of Biomaterials Applications
0(0) 1–14

DOI: 10.1177/0885328217750476
journals.sagepub.com/home/jba
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E-beam sterilizable thermoplastics elastomers for healthcare devices: Mechanical, morphology, and in vivo studies

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Abstract

The effect of electron beam radiation on ethylene–propylene diene terpolymer/polypropylene blends is studied as an attempt to develop radiation sterilizable polypropylene/ethylene–propylene diene terpolymer blends suitable for medical devices. The polypropylene/ethylene–propylene diene terpolymer blends with mixing ratios of 80/20, 50/50, 20/80 were prepared in an internal mixer at 165°C and a rotor speed of 50 rpm/min followed by compression molding. The blends and the individual components were radiated using 3.0 MeV electron beam accelerator at doses ranging from 0 to 100 kGy in air and room temperature. All the samples were tested for tensile strength, elongation at break, hardness, impact strength, and morphological properties. After exposing to 25 and 100 kGy radiation doses, 50% PP blend was selected for in vivo studies. Results revealed that radiation-induced crosslinking is dominating in EPDM dominant blends, while radiation-induced degradation is prevailing in PP dominant blends. The 20% PP blend was found to be most compatible for 20–60 kGy radiation sterilization. The retention in impact strength with enhanced tensile strength of 20% PP blend at 20–60 kGy believed to be associated with increased compatibility between PP and EPDM along with the radiation-induced crosslinking. The scanning electron micrographs of the fracture surfaces of the PP/EPDM blends showed evidences consistent with the above contentation. The in vivo studies provide an instinct that the radiated blends are safe to be used for healthcare devices.

Keywords

Electron beam sterilization, mechanical properties, polypropylene/ethylene–propylene diene terpolymer blend, in vivo studies, biocompatible

Introduction

The medical care devices such drainage tubes, catheters, forceps, sealing (o rings), connectors, blood bags, dialysis fittings, syringes are mostly made up of plasticized polyvinyl chloride (PVC) in which di (2-ethylhexyl) phthalate (DEPH) is used as plasticizer. However, leaching of DEPH has been proved to bring toxic effects in liver, reproductive tract, kidneys, lungs, fetus, and heart.[1] In parallel, the demands for such healthcare devices are increasing due to the escalation of world demography. In 2014, the global market of plastics for medical device was USD 4644.46 million. The reports predict the compound annual growth rate (CAGR) for such medical device plastics at 4.8 to 7.12% till 2020.[2,3] The demand has driven the researchers to engineer various PVC free compounds such as thermoplastics elastomer (TPE) to provide an alternate safe material which are suitable for medical device

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industries. TPE based on ethylene–propylene diene terpolymer (EPDM)/polypropylene (PP) blend is proposed as alternative for medical applications.[4,5] PP and EPDM individually have proved the biocompatibility. However, although base materials are biocompatible, many final polymer compounds fail in in vivo sensitization due to incorporation of additives and contamination during processing, sterilization, and packaging stages.[6]

Thus, our work focuses on in vivo studies of melt-blended PP/EPDM, sterilized using electron beam (E-beam) radiation. Sterilization is a necessary requirement to produce medical devices free from viable microorganisms. E-beam sterilization has recently gained commercial importance due to environment sustainability, bulk processing, and fast processing capabilities. The minimum radiation dose required for effective sterilization is 25 kGy. However, upon radiation free radicals are generated in the PP/EPDM blend which undergoes simultaneous scission and crosslinking mechanisms. These competing reactions lead to change in the mechanical properties of the blend depending upon the absorbed radiation dose. Although a number of studies are available in literature stating the change in mechanical properties of polymers upon radiation,[7,8] a definitive data on the effect of radiation on the PP/EPDM blends at the radiation doses ranging between 20 and 100 kGy is scarce. The objective of present study is to analyze the effect of radiation on the tensile strength, hardness, and impact strength and biocompatibility of PP/EPDM blends. Such studies were made as mechanical properties and in vivo studies play a key role in engineering the materials to cater the manufacturing of medical devices

Materials and methods

Materials

Isostatic polypropylene (Grade: Globalene 8661) was purchased from LCY Chemical Corp., USA. The melt flow index, density, and melting point of the material are 18 g/10 min, 0.9 g/cm^3, and >120°C, respectively. The EPDM of grade 3092PM having 65% ethylene and 6.4% diene monomer of Mooney Viscosity-61 (at 125°C) was purchased from Mitsui Chemicals, Shanghai.

Preparation of PP/EPDM blend

Figure 1 shows the schematic of the sample preparation. The PP/EPDM blends with mixing ratios of 80/20, 50/50/20/80 and the individual components were prepared by melt blending in an internal melt mixer (Haake Rheomix Polydrive R600/610 of volumetric capacity of 44 cm^3) at 165°C. The rotor speed was set to 50 r/min. The EPDM and PP were loaded simultaneously and was allowed to blend for a total time of 10 min. The samples were collected and pressed into

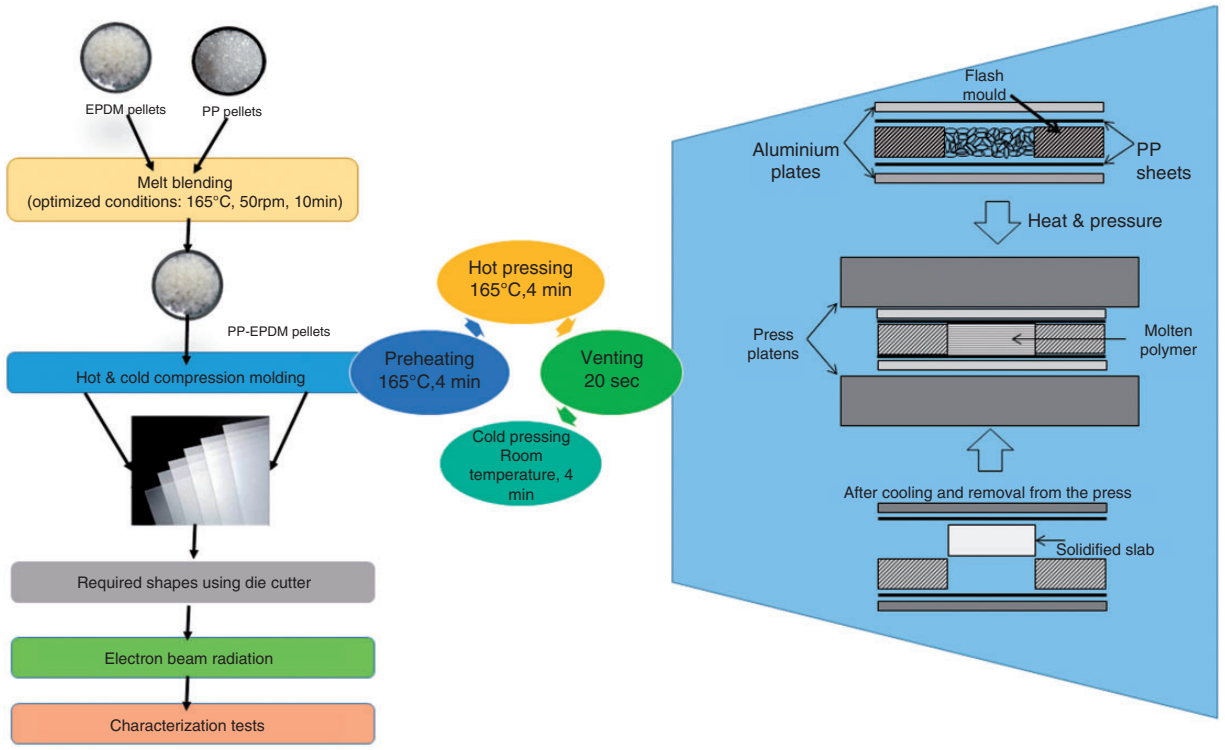


Figure 1. Schematics of sample preparation.

thin sheets of 1 mm thickness by hot and cold compression molding machine. The materials were placed in between aluminum sheets and were subjected to a fixed molding cycle. This process involved 4 min of preheating and 4 min of complete pressing in hot press at 165°C, followed by cooling at room temperature for 4 min under pressure equipped with chiller facilities. The schematics of sample preparation are illustrated in Figure 1.

E-beam radiation

The compression molded sheets were radiated under 3 MeV E-beam accelerator (model NHV-EPS-3000) under forced air cooling. The acceleration energy was set to 2 MeV, while the beam current and dose rate were fixed at 10 mA and 20 kGy per pass, respectively. The samples were radiated at various dose rates between 20 and 100 kGy. A 0.94 m/min conveyor speed was used with the distance of the sample from scan horn was set to 20 cm.

Gel content analysis

Gel content analyses were measured according to ASTM D2765. The samples were extracted for 24 h in boiling xylene using a Soxhlet extraction set. The resulting samples were then dried in oven at 70°C until a constant weight is obtained. Gel content was determined as per equation (1)

$$\text{Gel content } (\%) = \frac{w_1}{w_0} \times 100 \quad (1)$$

where w_0 and w_1 are the dried weights of sample before extraction and after extraction, respectively.

Tensile properties

The samples for tensile test were cut into dumbbell shape with dimensions (gauge length of 10 mm, thickness of 1 mm, and width of narrow section as 3.18 mm) in accordance with ASTM D638, specimen type V standard. The tensile properties were tested using a computerized tensile tester (Toyoseiki) with a load cell of 1 kN. The crosshead speed was set at 50 mm/min for all samples. The tensile strength and elongation at break were recorded for at least five specimens for each set of samples. The average results were reported as the resultant value. Standard deviation of the results was less than 10%.

Impact strength

All impact samples were cut into rectangular specimens and notched. The Izod impact tests were conducted according to ASTM D256 using CEAST (Model CE UM-636) Impact Pendulum Tester, with a 4 J hammer. The specimen dimensions were 6 mm wide and 3 mm thick with a notch of 2.59 mm. The samples of 3 mm thickness were dipped in liquid nitrogen for 1 min and impact energy was measured for cryo-fractured samples. Seven specimens were tested and at least five replicate specimens were presented as an average of tested specimens.

Hardness test

The Shore D hardness test was carried out according to ASTM D2240 using the Zwick 7206 Hardness Tester. Disc-shaped specimen with 5 mm thickness was used. The measured value of hardness was taken after 15 s of contact in Shore D indenter obtained at three different points distributed over the test piece. Three test pieces were used and their average value was determined. A minimum of seven hardness readings was recorded for each sample and average results were taken as the resultant value. Standard deviation of the results was less than 10%.

Scanning electron microscopy

Examination of the cryo-fractured surfaces was performed using field emission scanning electron microscope (FESEM, FEI Quanta 400). The surface of the fractured samples was sputter coated with gold before examination to avoid electrostatic charging and poor image resolution.

Dermal maximization test

The assay was performed to evaluate the potential of the radiated PP/EPDM blend to cause a delayed hypersensitivity reaction (Type 4) following exposure of the skin of guinea pigs. The review was performed on sound Dunkin Hartley albino guinea pig. The body weights of all the sound animals were noted preceding treatment. Twenty-four hours before treatment, the shoulder region of each guinea pig was clipped free of hair exposing 4 cm × 6 cm area. It was guaranteed that skin of all the guinea pigs were free from irregularities, skin injuries, and sick wellbeing. For induction phase: intradermal injection – three injection sites A, B, and C were designated and used to administer the test material and control items on three groups of animals comprising test, negative and positive control groups. The test material was prepared by extracting 3 cm^2 of test material in 1 ml of cottonseed oil at 37°C for 72 h in shaking water bath. The test material was administered by intradermal injection on three injection sites at intra-scapular region of the guinea pigs as shown in Table 1.

Table 1. Test materials used at various sites for dermal maximization test.

Site	Test group	Negative control group	Positive control group
A	50:50 vol% Freund's Complete Adjuvant and cottonseed oil	50:50 vol% Freund's Complete Adjuvant and cottonseed oil	50:50% Freund's Complete Adjuvant and 80% ethyl alcohol
B	Test material extract	Cottonseed oil	0.08% DNCB in 80% ethyl alcohol
C	Test material mixed in equal ratio of Freund's Complete Adjuvant and cottonseed oil	Cotton oil mixed in an equal ratio of Freund's Complete Adjuvant and cottonseed oil	0.08% DNCB in 80% ethyl alcohol mixed in an equal ratio of Freund's Complete Adjuvant and cottonseed oil

Seven days after completion of intradermal induction, test material, positive and negative controls were topically applied on the same injection sites using filter paper and absorbent gauze. The patch area was pretreated with 10% sodium lauryl sulfate in petroleum jelly for 24 h before application of patch. Covering the filter paper with gauze and a non-reactive occlusive adhesive tape, the patches were covered with elastic bandage to secure test material for 48 h. Fourteen days after completion of topical induction phase, the test material was patched onto the untreated guinea pigs for 24 h. Patches were then removed and the skin was examined for allergic reactions. The intensity of reactions was scored at 24 h and 48 h after patch removal. Negative control animals were prepared and subjected to similar treatment using cottonseed oil as blank. Similarly, positive control group animals were subjected to same treatment with 0.08% dinitrochlorobenzene (DNCB) in ethyl alcohol. The tests for irritation and skin sensitization were performed according to ISO10993-Part 10:2010(E).

Acute systemic toxicity test

Assessment of changes occurring after administration of a single, multiple, or continuous dose of test material within 24 h in ICR mice was done. The test materials were extracted in normal saline solution with a ratio of 3 cm^2/ml at 37°C for 72 h. Five solid mice were infused intraperitoneally with 50 ml/kg of test material concentrate. Extra five mice utilized as control gathering got 50 ml/kg body weight of typical saline in a comparative way. The treated and control groups were observed for morbidity and mortality once daily for 14 days.

Results and discussion

Gel fraction

Polymers when exposed to E-beam radiation undergo simultaneous crosslinking and scission mechanisms. The polymeric material absorbs E-beam, which carries enough energy leading to hydrogen abstraction from polymer backbone. This generates initial reactive species through homolytic or heterolytic bond scission. The initial species also termed as free radicals can undergo various secondary mechanisms which may lead to chain propagation, scission, crosslinking, degradation, and/or grafting.[9] These mechanisms lead to change in mechanical properties of the polymer and the later depend on which of the two is predominant at a certain time. PP is one of the most popular polymers in the manufacture of medical disposables due to its good transparency, nontoxic, high mechanical properties, and low cost. However, PP undergoes degradation during radiation and during storage after degradation. On the other hand, elastomer such as EPDM or EVA due to high amorphous nature favors the formation of crosslinking. Hence, addition of EPDM to PP may deter the degradation caused by PP in the blend, which is supported by gel content analysis in this study. The schematics of possible crosslinking or scission mechanism possible in the PP–EPDM blend is depicted in Figures 2(a) to (c) and 3. Since the absorption of energy by the polymers occurs spatially at random on a molecular scale, both intra (self-crosslinking within PP or EPDM) and inter (interfacial crosslinking between PP and EPDM) are possible (as depicted in Figures 2(a) to (c) and 3). The occurrence of crosslinking and scission mechanisms is hard to be quantified individually and define the matrix of occurrence specifically. However, the cumulative crosslinking of the blends can be studied using gel fraction analysis. The gel fraction is indicative of three-dimensional network formation as a result of crosslinking. Table 2 depicts the gel fraction of PP/EPDM blends at various radiation doses. It is apparent from Table 2 that the gel fraction increases with radiation dose for EPDM dominant blends, including 50% PP blend. In contrast, for 80 and 100% PP blend, the gel fraction is found to drop with radiation dose. Such trend in gel fraction clearly implies the radiation-induced crosslinking is dominating in EPDM dominant blends, while radiation-induced

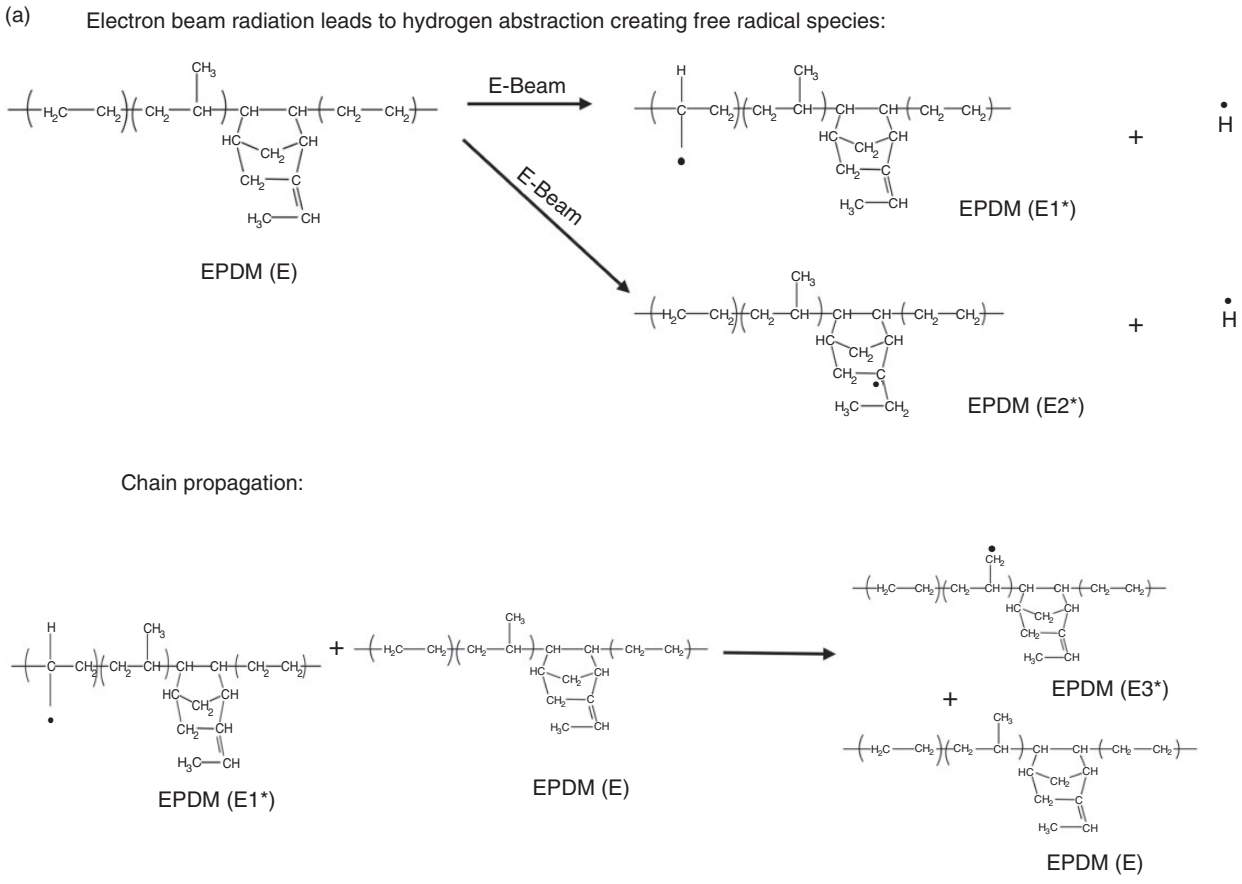


Figure 2. Schematics of free radical formation and chain propagation of (a) EPDM under E-beam and (b) PP under E-beam. (c) Schematics of possible crosslinking mechanisms.

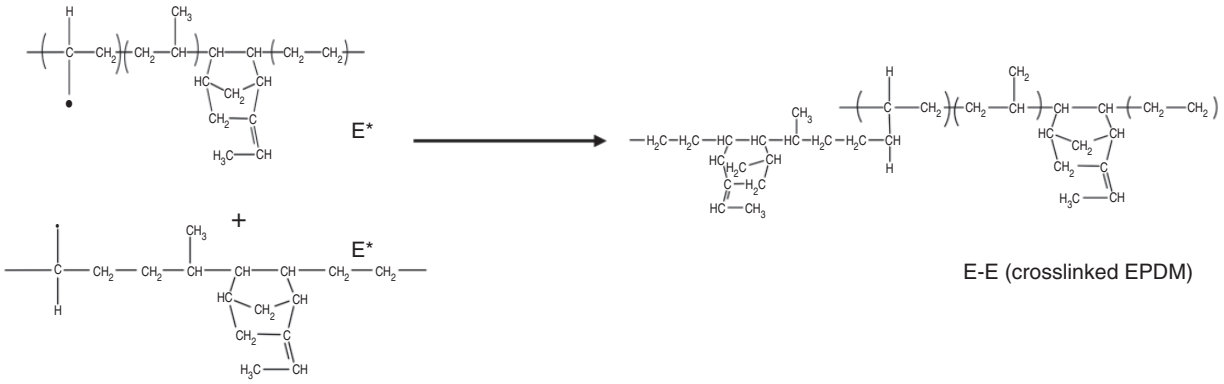
degradation is prevailing in PP dominant blends as reported by several researchers.[10]

Tensile strength

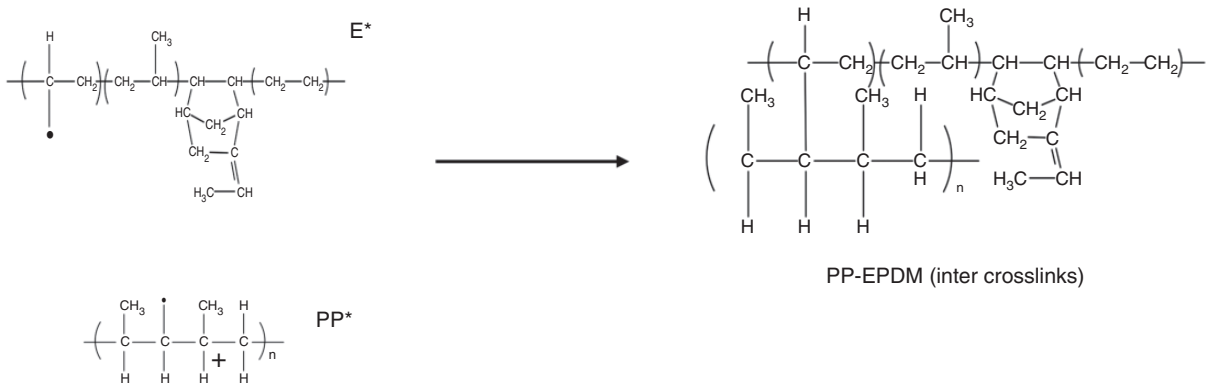
Figure 4 shows the tensile strength of PP/EPDM blend as a function of radiation dose ranging from 0 to 100 kGy. From the figure the tensile strength of 80% PP blend is 25.28 MPa, 50% PP blend is 19.59 MPa, and of 20% PP blend is 14.53 MPa, respectively. Tensile strengths of all the blends lie within 100% PP (31.59 MPa) and 100% EPDM (9.92 MPa) controls. It can be noticed that prior to radiation, the tensile strength of PP decreases with increasing EPDM content.

All the free radicals formed both EPDM and PP, they can either undergo self- crosslinking or inter-crosslinking

1) EPDM self-crosslinking



2) PP-EPDM inter-crosslinking



Similar to crosslinking mechanisms, all the free radicals can also lead to scission and further oxidation

Figure 2. (continued).

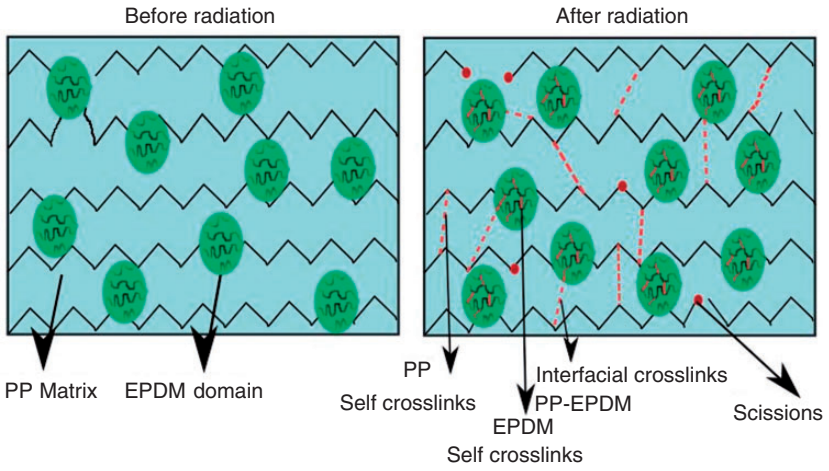


Figure 3. Schematics showing general crosslinking and scission formation after E-beam radiation.

The decrease in tensile properties is simply due to the addition of EPDM which has relatively lower tensile strength in comparison to PP. The lower tensile strength of EPDM can be attributed to its amorphous nature. PP/EPDM blends approximately followed the rule of mixture over the whole composition range.[11]

From Figure 4 it can be noticed that upon radiation the tensile strength of 100% PP shows a gradual decrease with increasing the radiation dose. Such a decline in tensile strength upon radiation is attributed to the radiation-induced scissions in PP dominant blends as observed from the gel fraction results in Table 2. Besides, upon radiation the free radicals formed from PP undergoes oxidative degradation also resulting in the decreased tensile strength.[12] The blends with 80% PP and 50% PP show good retention till 20 kGy and show a gradual decrease with increasing radiation up to 100 kGy. The decrease in tensile strength is again due to chain scission of PP matrix. However, the 80% PP and 50% PP can withstand radiation up to 20 kGy unlike 100% PP where the degradation was evident at 20 kGy itself. The presence of EPDM has enhanced the ability of the PP-rich blends to retain its tensile strength up to 20 kGy.

On the other hand, it can be noted that the tensile strength of 20% PP blend reaches an optimum in the range of 40–60 kGy radiation doses before shows a drop at 80 kGy. Such observation implies that exposing 20% PP blend to 40–60 kGy able to increase the radiation stability with concomitant enhancement in tensile strength of the blend. The increased tensile strength of 20% PP blend up to 23.06 MPa at 40 kGy could be attributed to the intense crosslinking induced by radiation. In fact, the tensile strength of 20% PP blend in this range gives strength close to 80% PP having tensile strength of 25 MPa. Similarly, 100% EPDM shows increase in tensile strength of 20 MPa at 20 kGy followed by a drop. The crosslinks induced by E-beam radiation reduces the segmental rotation and the mobility of the polymer chains which restricts its free movement, adding strength to the materials.[13] Hence, the presence of crosslinks increases the tensile strength. The drop in both 20% PP and 100% EPDM after optimum is associated with the embrittlement of EPDM caused by excessive formation of crosslinks, at above 60 kGy and 20 kGy for 20% PP and 100% EPDM, respectively. In the initial state of radiation, larger network system is framed by radiation actuated crosslinking bringing about an increase of tensile strength. However, later at higher radiation doses, additional crosslinks are formed between the promptly

Table 2. Gel fractions for PP/EPDM blends at various radiation doses.

Sample	Dose (kGy)	Gel fraction
100% PP	20 kGy	0.137
	60 kGy	0.162
	80 kGy	0.117
80% PP	20 kGy	0.153
	60 kGy	0.179
	80 kGy	0.148
50% PP	20 kGy	0.292
	60 kGy	0.527
	80 kGy	0.543
20% PP	20 kGy	0.440
	60 kGy	0.901
	80 kGy	0.974
100% EPDM	20 kGy	0.654
	60 kGy	0.976
	80 kGy	0.990

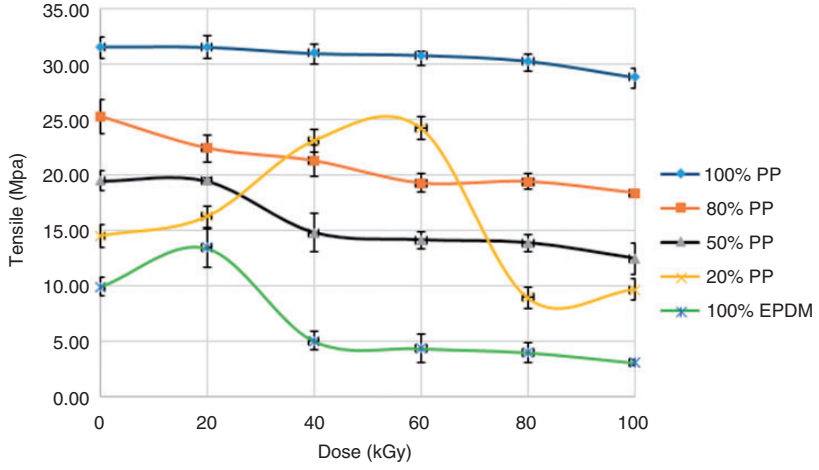


Figure 4. Tensile strength of PP/EPDM blends as a function of radiation dose.

crosslinked chains contravening the larger network structure into smaller networks. This results in the decrease of tensile strength. A similar trend has been observed by both Ratnam et al.[14] and Ramarad et al.[15] in radiation processing of poly (vinyl chloride), PVC/epoxidized natural rubber, and ethylene vinyl acetate (EVA)/waste rubber blends, respectively. It can also be inferred that, upon loading force, the energy is utilized to break the networks and only minimal energy is indulged in the polymer matrix.[16]

Elongation at break

Figure 5 shows the elongation at break of PP/EPDM blends with increasing radiation dose. Prior to radiation, the 80% PP blend exhibits 730% elongation, the 50% PP blend exhibits 800% elongation and the 20% PP blend shows up to 900% elongation. The elongation at break of all the blends lies within the limits of PP and EPDM blends. The 100% PP blend exhibits up to 600% elongation. On the other hand, 100% EPDM exhibits as much as 1000%. This implies that the elongation at break of PP blends increases upon addition of EPDM due to the amorphous nature of the later.

Further, upon irradiation, 100% PP and 80% PP decrease constantly up to 40 kGy. With further increase in radiation dose above 60 kGy, the blends do not show any elongation. This is due to the scission mechanism dominating at these doses, as discussed earlier. Similarly, the blends with 50% PP and 20% PP

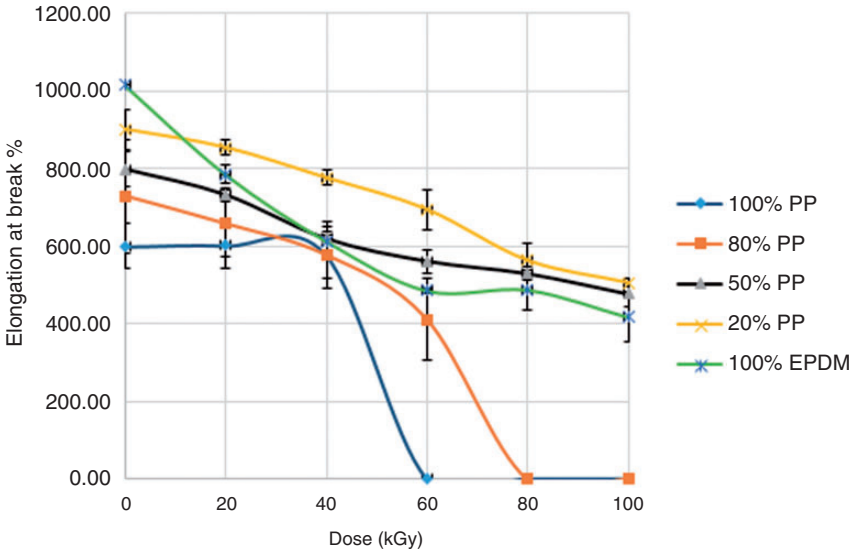


Figure 5. Elongation at break of PP/EPDM blends as a function of radiation dose.

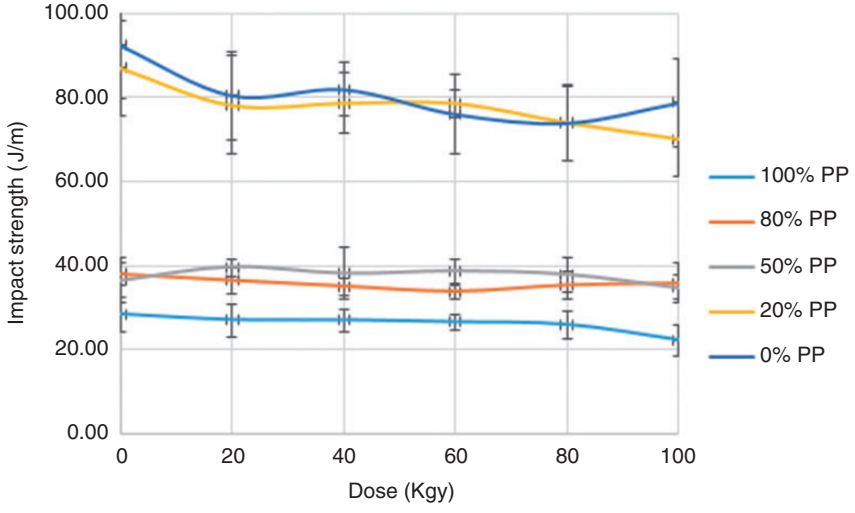


Figure 6. Impact strength of PP/EPDM blends as a function of radiation dose.

show a gradual decrease in elongation up to 100 kGy. The 100% EPDM blend also shows a progressive decrease in elongation at break with increase in the radiation dose. The radiation-induced crosslinks causes embrittlement of the EPDM, hindering the movement of the rubber chains.[17] This in turn reduces the ability of blend to plastically deform resulting in reduction of elongation at break with increased dose.

Impact strength

Figure 6 indicates the changes in the impact strength of PP/EPDM blends with the function of radiation. The impact strength of 80% PP increases up to 38.13 J/m (in comparison with 100% PP with 28.43 J/m impact strength) upon addition of EPDM. The addition of EPDM increases the impact strength of PP. This observation implies the toughening effect of EPDM which serves as a soft elastomer in rigid PP matrix. However, with further increase in the EPDM loading up to 50%, there is a slight decrease in impact strength. The impact strength of 50% PP is slightly decreased to 36.59 J/m. This is in accordance to other established works, which states that the optimum EPDM loading into PP for better impact strength is 20–40%. At lower EPDM loading, the EPDM is dispersed as rubber domains in continuous PP matrix.[18] The EPDM dispersed domains absorb energy and deforms. These deformed particles further initiate shear yielding and craze in the polymer matrix, which further absorbs impact energy.[19] In addition, a number of voids are found in 20% PP blend (Figure 7(a)). The voids suppress nucleation of cataclysmic cracks at the location of crazes or matrix deformation and improve the impact strength.[20–22] However, at 50% loading, the voids and the rubber as dispersed particles are lesser comparatively to the 20% EPDM blend and tends to form co-continuous phase, which is witnessed in Figure 7(a) and (b) leading to a slight decrease in impact property. Upon further addition of EPDM, impact strength increases significantly to 87.1 J/m (for 20% PP blend). This is again attributed to the toughening mechanism of EPDM, as major part of this blend is in amorphous form. However, all the impact strength of all the blends lie with the range of PP having the least impact strength of 28.3 J/m and EPDM having the highest of 92.35 J/m.

The effect of radiation upon impact properties of all the blends with respect to the neat polymers can be studied from Figure 6. The blends up to 50% EPDM concentration show minor or no changes in impact strength upon increasing radiation dose. However, the EPDM-rich blend with 20% PP and 100% EPDM shows a steady decrease in its impact strength with increasing dose. There is a decrease of up to 25% and 37% in impact strength of 20% PP and pure EPDM blends at 100 kGy, respectively. This can also be attributed to the extensive crosslinking of the EPDM elastomer as mentioned earlier.[23] A similar trend has been reported by Van Gisbergen et al.[24] in 70%:30% high molecular weight PP/EPDM blend. It can also be inferred that the larger networks formed upon radiation deter the energy to be dissipated into the polymer matrix, thus reducing the impact strength.

Hardness

The 80% and 50% PP show relatively closer values of hardness around 95 shore D. The addition of EPDM up to 50% did not show any significant change in the hardness value. The hardness of these blends is close to the hardness of the 100% PP. However, the EPDM-rich blend of 20% PP shows a significant decrease up to 65.25 shore D. This is due to the low hardness value of EPDM, where 100% EPDM shows only 57.9 shore D, due to its amorphous nature.

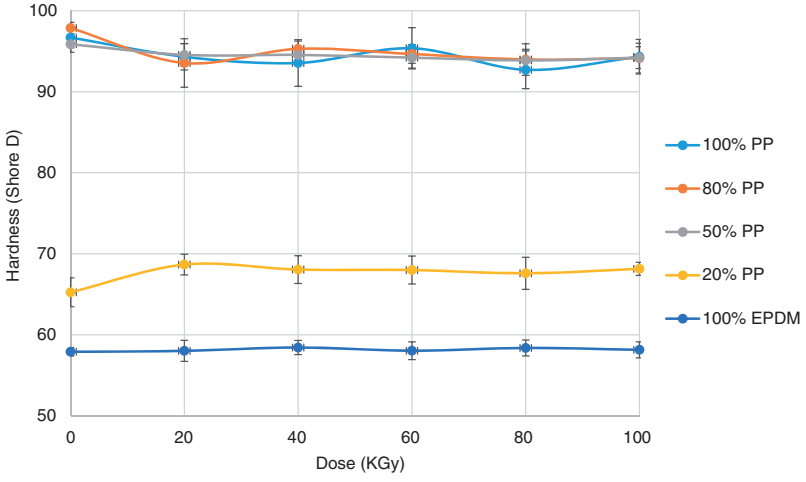


Figure 7. Hardness of PP/EPDM blends as a function of radiation dose.

From Figure 7 it can be noted that 100–50% PP blends showed a downward trend in hardness upon radiation from 20 to 100 kGy. Such a decline is due to the reduction in resistance to deformation of the blends as a result of radiation-induced chain scissioning of PP. However, the hardness of 20% PP increased by 5% upon a radiation dose of 20 kGy, due to crosslinking induced by radiation. Upon further radiation, no increment in hardness was observed. Similarly, the crosslinking due to E-beam radiation did not cause a significant effect to the hardness of 100% EPDM, even at high dose (up to 100 kGy), implying the degree of crosslinks achieved at 20–100 kGy is inadequate to cause enhancement in the hardness of the 100% EPDM and 20% PP blends.

Morphology studies

Figure 8(a) and (b) shows the SEM micrographs of 100% PP before and after radiation at 40 kGy, respectively. From Figure 8 it can be noted that before radiation, 100% PP showed a semi-crystalline morphology due to the amorphous content (poly-ethylene) in the co-polymer. However, upon radiation at 40 kGy, a few cracks in the matrix were witnessed. These cracks could be due to degradation of the polymer matrix due to radiation-induced chain scissoring as well as oxidation, making the polymer vulnerable to failure upon loading of energy.

Figure 9(a) and (b) represents the morphology of 80% PP and 50% PP blends prior to radiation. It can be witnessed that more voids are formed in 80% PP. The voids formed could be due to the delamination of EPDM during deforming process and blending process. The presence of voids signifies the incompatibility between the blends. Also, the elastomer is found to be dispersed as droplets in the PP matrix, due to the thermodynamic immiscibility.[25] Although droplets of elastomer in matrix can be seen in 50% PP blend, they are fewer in comparison to 80% PP. The presence of more

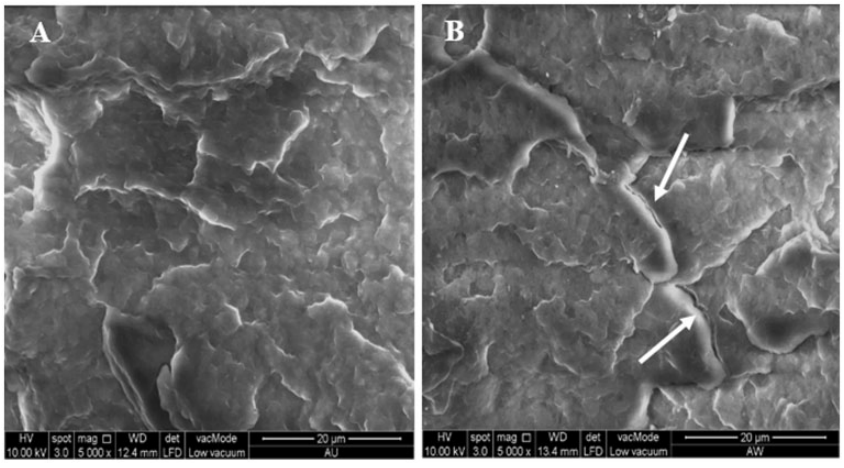


Figure 8. SEM image for 100% PP: (a) before radiation and (b) after radiation at 40 kGy.

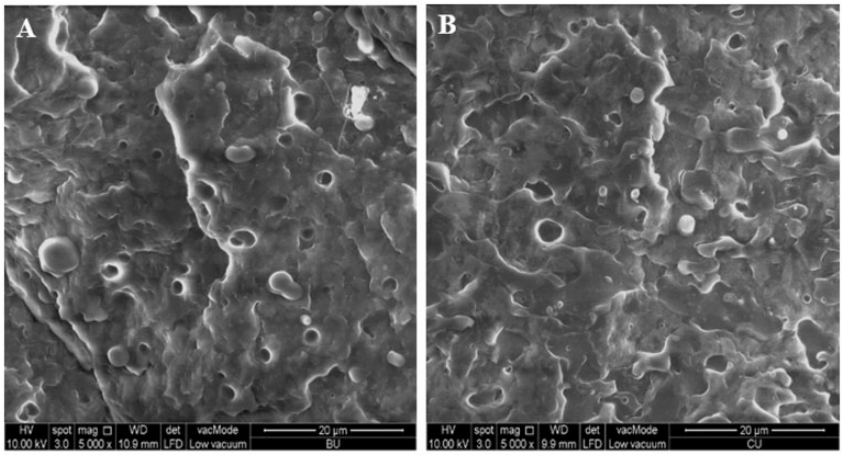


Figure 9. SEM images of blends before radiation: (a) 80% PP and (b) 50% PP.

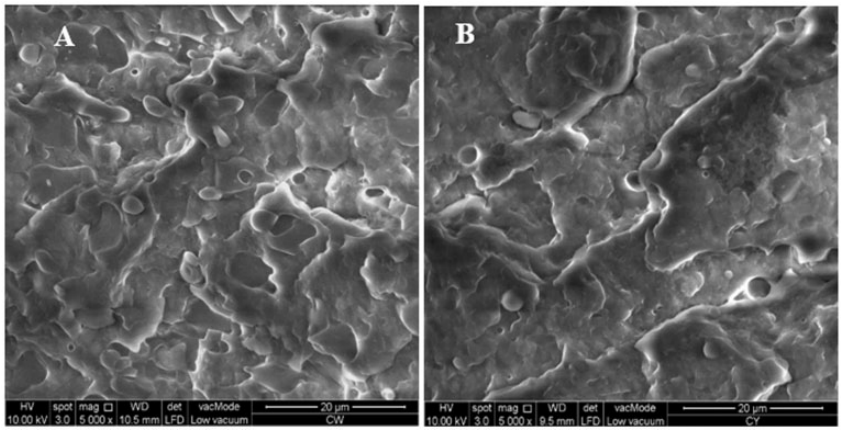


Figure 10. Morphology of 50% PP blend: (a) at a radiation dose of 40 kGY and (b) at a radiation dose of 80 kGy.

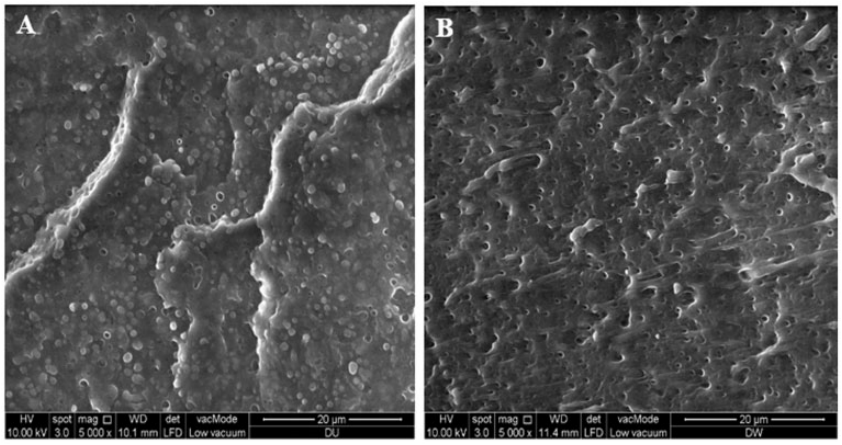


Figure 11. Morphology of 20% PP blend: (a) before radiation and (b) after radiation at 40 kGy.

voids and elastomer as droplets improves the impact strength of 80% PP blend, which is explained detailed in Impact strength section.

Further, Figure 10(a) and (b) shows the SEM micrograph of 50% PP blends before and after radiation. Apparently, from Figures 9(b), 10(a) and (b), no significant changes are observed for 50% PP blend upon radiation. However, in 20% a significant difference in the morphology upon radiation could be observed (Figure 11). Upon radiation the continuous matrix (EPDM phase) covers or overlapping of the dispersed particles (PP phase) is achieved. This is an indication of enhancement of interfacial adhesion, as the PP has grafted to EPDM due to radiation-induced crosslinking. A similar interference is reported in literature where crosslinking was achieved by various methods.[20,26] However, in this case the interfacial adhesion did not result in appreciable change in the impact properties of the blends. The continuous matrix overlaps or covers the dispersed phase, deterring them to act as stress concentrators. The continuous phase simultaneously overlaps the voids, thus reducing the impact strength.[20] The decrease in impact strength, elongation at break, and tensile strength beyond optimum in radiated EPDM-rich blends can also be related to the restriction of mobility of the macromolecules caused by the crosslinking. The macro-molecular immobility is well evident from the dynamic mechanical analysis (DMA) analysis, which will be detailed in a separate study. This in turn reduces the elasticity and increases the stiffness and brittleness, which is evident from Figure 12. Figure 10 shows the impact fractographs of 20% PP blend before and after radiation at 80 kGy. From Figure 12 it can be seen that energy is dissipated in the form of long fibrils, witnessing semi-ductile surface. However, upon radiation the morphology indicate semi-crystalline matrix, where brittleness is indicated by the increase in number of crazing formed and the roughness of the matrix.

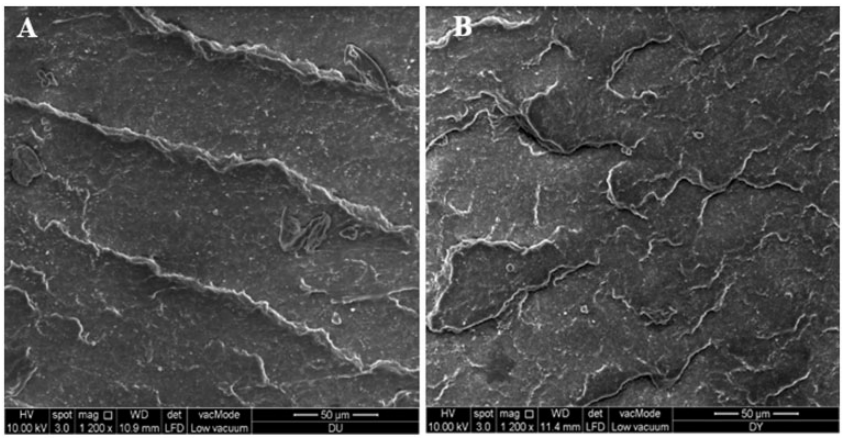


Figure 12. Impact fractograph for 20% PP blend: (a) before radiation and (b) after radiation at 80 kGy.

Table 3. Skin sensitization, dermal reaction grades for 50:50 PP/EPDM blend at 25 kGy.

	Test extract				Positive control				Negative control			
	Hours after patch removal											
	24		48		24		48		24		48	
Animal test number	*E*	*O*	*E*	*O*	*E*	*O*	*E*	*O*	*E*	*O*	*E*	*O*
1	0	0	0	0	3	1	3	1	0	0	0	0
2	0	0	0	0	3	1	3	2	0	0	0	0
3	0	0	0	0	1	1	1	1	0	0	0	0
4	0	0	0	0	2	1	2	1	0	0	0	0
5	0	0	0	0	3	2	3	3	0	0	0	0

E: erythema; O: oedema.

Table 4. Skin sensitization, dermal reaction grades for 50:50 PP/EPDM blend at 100 kGy.

	Test extract				Positive control				Negative control			
	Hours after patch removal											
	24		48		24		48		24		48	
Animal test number	*E*	*O*	*E*	*O*	*E*	*O*	*E*	*O*	*E*	*O*	*E*	*O*
1	0	0	0	0	2	0	2	0	0	0	0	0
2	0	0	0	0	2	0	1	0	0	0	0	0
3	0	0	0	0	3	0	2	0	0	0	0	0
4	0	0	0	0	3	0	2	0	0	0	0	0
5	0	0	0	0	3	0	2	0	0	0	0	0

E: erythema; O: oedema.

Dermal maximization test

The dermal sensitization was performed on 50% PP blends radiated at 25 and 100 kGy. The skin reaction during the challenge phases after patch removal was graded from 0 to 4, according to Magnusson and Kligman scale.[27] A score of zero indicated no irritation and the highest of four indicated the occurrence of intense erythema and edema and/or swelling.[28] The grades of test extract material, positive and negative controls assessed after 24 h and 48 h of patch removal are listed in Tables 3 and 4. From the tables, it can be

witnessed that positive controls exhibited moderate and confluent erythema and edema, and negative controls did not show any effect as anticipated. However, from Tables 3 and 4 both the test materials (25 kGy and 100 kGy PP/EPDM) were graded a score of zero, indicating no skin sensitization reaction after 24 h and 48 h of patch removal. It is also noted that the body weights of the guinea pig increased.

Acute systemic toxicity test

Acute systemic toxicity test is used to study the possibility of any potential health hazards that are about to arise from short-time exposure caused by fluids, bloods, or drugs that are stored in PP/EPDM blends and enters intraperitoneal route. The treated group and control group were administered daily for 14 days. Acute toxicity test was performed for samples radiated at 25 kGy and 100 kGy. The clinical signs in fur and skin changes, eyes, respiratory effect, motor activity, mucous membrane, tremor, involuntary contraction of the voluntary muscle, body movements, diarrhea, and death were examined. Gross necropsy was performed on both groups following euthanasia at termination day. All the animals survived the course of study period with no evidence of clinical or toxicity. All the animals did not show any weight loss. Gross examination on treated groups (for 25 kGy and 100 kGy test material) and control groups showed no remarkable abnormalities in brain, kidneys, lungs, liver, stomach, spleen, heart, and pancreas during necropsy. Thus, 25 kGy and 100 kGy test materials did not show any adverse toxic reaction at 50 ml/kg body weight under the condition of this study.

Conclusion

It can be concluded that the changes in the mechanical properties of the PP/EPDM blends upon radiation was influenced by the EPDM content and absorbed radiation dose. The PP-rich blends witnessed a decrease in tensile properties upon increasing radiation due to radiation-induced chain scissioning of PP. On the other hand, EPDM-rich blends found to reach maximum tensile strength at 20–40 kGy followed by a drop, due to excessive crosslinking of the EPDM. The impact properties of PP found to increase with addition of EPDM although it showed a decline with radiation dose. The highest elongation at break and impact strength obtained for 20% PP blend as compared to all other samples studied in this work suggests that 20% PP blend is most compatible for 20–40 kGy radiation sterilization. Enhancement in tensile strength believed to be associated with increased compatibility between PP and EPDM along with the radiation-induced crosslinking. SEM micrographs of fractured surfaces of the PP/EPDM blend complemented the observed changes in mechanical properties of the blends upon radiation. Further, in vivo studies performed confirm that no sensitization persisted. Thus, PP/EPDM blend radiated by E-beam provides us an instinct that these blends could be a potential alternate for PVC in manufacturing medical devices. The radiation dose depends upon the application of the targeted blends. However, further studies including thermal stability and other in vivo studies including minimum essential medium (MEM) elution, hemolysis, oral mucous membrane irritation tests need to be performed to confirm their biocompatibility to humans and other high-order species.

Acknowledgement

The authors wish to thank Mr Wan Ali of Polymer Technology Laboratory, Nuclear Malaysia, for the assistance during the experimental work.

Declaration of Conflicting Interests

The author(s) declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

Funding

The author(s) disclosed receipt of the following financial support for the research, authorship, and/or publication of this article: The authors sincerely thank the Ministry of Science, Technology and Innovation, Malaysia, MOSTI, for granting this project (code: 03-03-01-SF0242).

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