Mechanical Properties of Recycled LDPE/EVA/Ground Tyre Rubber Blends: Effects of EVA Content and Postirradiation

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ABSTRACT: Thermoplastic elastomers were prepared from recycled low density polyethylene [rLDPE] and virgin low density polyethylene (LDPE), respectively, ground tyre rubber (GTR), and ethylene vinyl acetate (EVA) copolymer. The amounts of the rLDPE and GTR were fixed at 40 and 30 wt %, respectively, in the formulations, whereas the LDPE and EVA contents varied each between 0 and 30 wt %. The fresh LDPE served for reduction of the melt viscosity and EVA was used for improving the elastomeric properties. Blends of different compositions (by varying the LDPE/EVA ratio) were produced by twin-screw extrusion and pelletized. Specimens were produced by injection molding and subjected to tensile and instrumented falling weight impact (IFWI) tests. To improve the mechanical performance of the blends, the injection molded specimens were electron beam irradiated at 150 kGy absorbed dose. Static tensile and hysteresis, IFWI and dynamic mechanical thermal analysis tests were performed on the specimens and the fracture surface was inspected with a scanning electron microscope. The results indicated that better rubber-like properties were achieved with increasing EVA content. Moreover, postirradiation proved to be very beneficial, especially for blends containing relative high amounts of EVA. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 125: 512–519, 2012

Key words: polymer blends and alloys; ground tyre rubber; low density polyethylene; ethylene vinyl acetate copolymer; thermoplastic elastomers; irradiation

INTRODUCTION

The increasing number of used tyres, the growing environmental concerns, and strict legal regulations inspire researchers to find new fields of application for ground tyre rubber (GTR). Similarly to thermoplastic polymers, there are three ways of disposing used rubber products (e.g. tyres): dumping (this is illegal in most of the developed countries), energy recovery (mostly in cement kilns) or burning the pyrolysis products of the tyres, and the third category is the recycling. The latter involves retreading; reuse in other functions (buffers, soil stabilization, etc.). GTR with suitable "binders" are used for covers of sport and playing fields, blending with asphalt for road pavements and also added in small amount in rubbers, thermosets, or thermoplastics. The importance of the latter is continually growing as value-added products can be generated from GTR (up-cycling strategy).

One of the possible ways of up-cycling is to produce polymer (mainly thermoplastic) blends that contain GTR. Several thermoplastics and thermosets have already been tested as embedding matrices for GTR, but the most important of them are low density polyethylene (LDPE),[1–5] high density polyethylene (HDPE),[4,6–10] and polypropylene (PP).[11–15] However, the incorporation of GTR into polymer matrices significantly deteriorates the mechanical properties of the resulting blend due to poor GTR/matrix adhesion because of lacking reactive functional groups on the GTR surface. This problem can be reduced by the chemical modification of GTR: devulcanization (decreasing the degree of crosslinking) into reclaimed tyre rubber[2,16–18] or by the activation (oxidation) of rubber particle surfaces with high energy treatment (ultraviolet,[13] gamma irradiation,[4] and plasma or corona charge treatment[19]). The *in situ* vulcanization of rubbers in thermoplastic matrices in the presence of additional GTR may result

TABLE I
Components of the Investigated Blends

	Blend 1	Blend 2	Blend 3	Blend 4
GTR (wt %)	30	30	30	30
rLDPE (wt %)	40	40	40	40
LDPE (wt %)	30	20	10	0
EVA (wt %)	0	10	20	30

thermoplastic dynamic vulcanizates that have outstanding properties. Besides, the aforementioned chemical modification techniques, compatibilization of the GTR containing thermoplastic blend is also a very promising approach. As compatibilizers ethylene octene copolymer (POE),[7] ethylene vinyl acetate (EVA) copolymer,[7,20] chlorinated polyethylene, maleic anhydride-grafted-polyethylene, ethylene–propylene copolymer,[6] and ethylene-propene-diene monomer elastomer[6,21] were usually used. High energy irradiation also improved the compatibility of the blends.[9,22,23] The latter is a novel method which modifies the surface of GTR with γ or electron beam irradiation. In polymeric materials, irradiation results in the formation of free radicals, which may cause chain scission, grafting, or crosslinking, depending on the type of the corresponding polymer or blend. For example, electron irradiation in inert atmosphere is generally used for polyethylene crosslinking.

In case of GTR, the irradiation induces rather chain scission and decomposition than formation of new crosslinks. The formed free radicals can react with the matrix material and thereby can decrease incompatibility.[9,21,22] Sharif et al.[24] investigated the effect of electron beam irradiation on LDPE/EVA blends. The presence of EVA increased both tensile strength and elongation at break without irradiation, while on the other hand irradiation resulted in higher tensile strength and lower elongation at break. These effects appear already at relatively low, 50–100 kGy absorbed doses.

In our previous work, LDPE/GTR blends (with various GTR and EVA contents) were investigated.[20] The results indicated that the LDPE is a good choice for the matrix and EVA is suitable to create good interfacial bonding to both LDPE[25] and GTR.[7] In addition, EVA has rubber-like properties. The goal of this study was to prepare LDPE/recycled LDPE (rLDPE) and GTR containing injection mouldable thermoplastic elastomers and to investigate the effect of the amount of EVA copolymer and postirradiation on the mechanical properties of LDPE/GTR blends.

EXPERIMENTAL

Materials and their processing

rLDPE (Holofon LDPE 4005, Holofon'95 Ltd., Tinnye, Hungary) with a melt volume rate of $MVR_{190°C/2,16 \ kg}$

$= 1 \ cm^3 (10 \ min)^{-1}$ was selected and used as the thermoplastic matrix. The rLDPE amount was kept at 40 wt % in each blend.

GTR powder was produced by mechanical grinding at ambient temperature by C.S.O. (Ócsa, Hungary). Then, it was screened and classified. For this study, the powder having particles size between 0.25 and 0.5 mm were selected and used. The GTR content in the blend was also fixed, namely at 30 wt %. Based on our previous study,[20] 0–30 wt % EVA copolymer (IBUCELL K-100, H. B. Fuller) with $MVR_{190°C/2,16 \ kg} = 10 \ cm^3 (10 \ min)^{-1}$ was added to improve the elastomeric properties of the blends.

As recycled LDPE and GTR have high viscosity (low fluidity), 0–30 wt % injection molding grade LDPE (Tipolen OF 2019, TVK, Tiszaújváros, Hungary) with $MVR_{190°C/2,16 \ kg} = 26 \ cm^3/10 \ min$ was added to the blend. As the EVA content was raised, the original LDPE ratio was decreased simultaneously; hence, the content of LDPE (original) and EVA together was always 30 wt % in each blend.

Different blends—listed in Table I—were prepared from the components in a Brabender Plasti-Corder PL 2100 type twin-screw extruder. Extrusion temperature was between 165 and 175°C and the number of revolutions was 10 min^{-1}. The extruded materials were subsequently granulated.

Specimens and their testing

Dumbbell type specimens (4 × 10 mm of cross section) for tensile and plaque specimens (with the dimensions of 80 × 80 × 2 mm) for instrumented falling weight impact (IFWI) tests were injection molded from the granulated blends in an Arburg Allrounder 320C 600–250 injection molding machine. Table II contains the related processing parameters.

A part of the produced tensile and IFWI specimens were postirradiated in a LUE-8-5V (NIEFA) type electron radiation device with an absorbed dose of 150 kGy. Note that this dose yielded crosslinked structure, for both LDPE[24] and EVA.[26]

TABLE II
Injection Molding Parameters for Both Specimen Types

Injection molding parameters	Tensile specimen	IFWI specimen
Temperature—Zone 1–5 (°C)	165...190	
Temperature—Mold (°C)	40	
Injection volume (cm^3)	44	50
Injection pressure (bar)	550–850	510–700
Switchover point (cm^3)	12	
Injection rate (cm^3 s^{-1})	50	
Holding pressure (bar)	400	
Holding time (s)	20	
Cooling time (s)	15	

Dynamic mechanical thermal analysis (DMTA) was performed on rectangular specimens cut from the IFWI specimens in tension mode on a Perkin Elmer Diamond DMA at a frequency of 1 Hz and heating rate of $2^\circ C$ min^{-1}. The temperature range was -120 to $120^\circ C$.

The cyclic tensile tests were performed on a Zwick Z020 universal testing machine according to EN ISO 527 (Dumbbell specimen–1A). Apart from the usual static mechanical tests, cyclic tensile tests are often used for testing thermoplastic polymers and their blends[27] to determine their viscoelastic characteristics. Cyclic investigation is performed at a constant loading rate—only the direction changes according to uploading and unloading. In this study, one cycle was set before a usual tensile test. During the cycle, a crosshead speed of 5 mm min^{-1} was applied up to 25 N, then unloading at 5 mm min^{-1} to zero force, where 15-s-long relaxation time was set. Thereafter, a tensile test was carried out at a loading rate of 50 mm min^{-1}. The tensile modulus was determined in the strain range of 0.1–0.5% at the beginning of the cycle.

The curves of uploading and unloading taken during the cyclic tests do not fit on each other, that is, a hysteresis area can be observed (Fig. 1). The total deformation (ε_{tot}) can be divided into three components: the instantaneous elastic (ε_0), the time-dependent viscoelastic (ε_v), and in the time-dependent viscous (or relaxation) deformation components (ε_{rel}), and they can be determined with the help of the cyclic diagram (Fig. 1).

$$\varepsilon_{tot1} = \varepsilon_{01} + \varepsilon_{v1} + \varepsilon_{rel1} \qquad (1)$$

where "1" in subscript indicates the first cycle.

At the Point A, the uploading is stopped and started unloading. At the Point B, the force in the specimen becomes zero, which means total recovery of the instantaneous elastic deformation component and partial recovery of the time-dependent visco-elastic deformation component. To assess the components more accurately, the sample has to be relaxed for a given time (in this case for 15 s) before starting the next cycle (longer time is available for the restitution of the viscoelastic deformation component). The next cycle will start from Point C and the developed viscous (ε_{rel1}) deformation in the first cycle can be approached by the distance between Points 0 and C (Fig. 1). The instantaneous elastic component (ε_{01}) can be estimated with the section (between Points D and E) determined by the tangent fit to the initial point of the unloading curve (thereby, its real value is usually overestimated). The behavior of rubber-like materials can be characterized primarily by the instantaneous component.
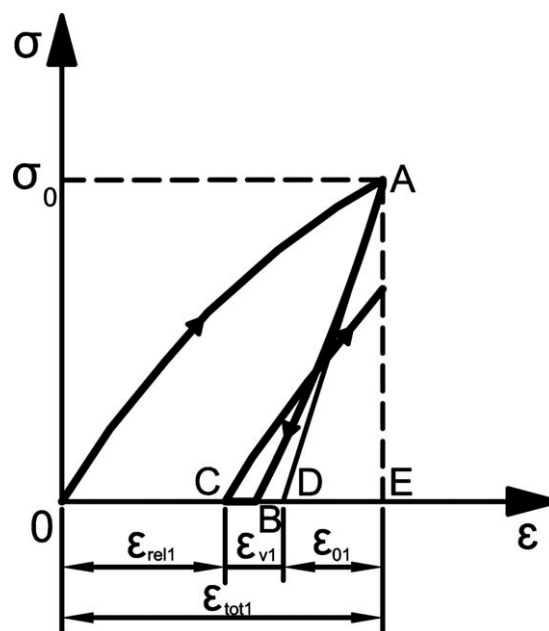

Figure 1 Deformation components and hysteresis area in case of given relaxation time.

Instrumented falling weight impact (IFWI) tests were performed on a Fractovis 6785 instrument (Ceast, Pianezza, Italy) using the following settings: maximal energy: 229.05 J; diameter of the dart: 20 mm; diameter of the support rig: 40 mm; weight of the dart: 23.62 kg, and drop height: 1 m. All mechanical tests were performed at $23^\circ C$. The fracture surface of the tensile specimens was inspected with a scanning electron microscope (SEM; JEOL 6380 LA, Japan) after coating with an Au/Pd alloy.

RESULTS AND DISCUSSION

DMTA tests

To investigate the temperature-dependent behavior of the blends, DMTA tests were performed. Figures 2 and 3 show the tan δ–T and storage modulus–T curves for LDPE, EVA, Blend 1, and Blend 4, respectively. It can be seen from the tan δ–T curves that EVA possesses the highest peak around $-20^\circ C$, which belongs to the glass transition temperature (T_g) of the EVA. In case of Blend 1, there is a shoulder in the tan δ–T trace around this temperature meaning that the rLDPE itself contained EVA. The tan δ–T curves of the blends are superpositions of the corresponding curves of the pure components (EVA, LDPE, and GTR). The run of the tan δ–T curves differs from each other because of changing content of the components and the quality of interfacial connection between the phases. Note that the interfaces between the components is determinant in the final properties of the blend.[28] If the bonding between two phases becomes stronger, tan δ peaks show an
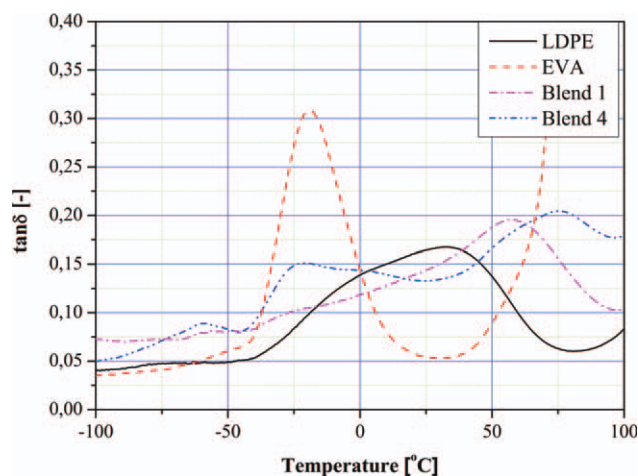
Figure 2 tan δ curves of the components and Blends 1 and 4 (nonirradiated). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Figure 4 tan δ curves of the components and Blends 1 and 4 (irradiated). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

inward shift with respect to the parent components. The small tan δ peak in Blends 1 and 4 around −60°C refers to the GTR phase reflecting its synthetic (mainly styrene butadiene rubber) and natural rubber components (T_g of natural rubber is around −70°C and the T_g of synthetic rubber is around −52°C[29]). The tan δ–T curve of the pure LDPE is a superposition of two peaks, one around −10°C and the other around 35°C. The first is associated with the transition of branch points (β-transition) and the second to the start of the movement of the molecules connected to the crystalline phase (α-transition; Ref. [30] and references therein).

The storage modulus–temperature curves show that at room temperature, the LDPE and Blend 1 (0 wt % EVA) have relatively high modulus (around 300 MPa). By the addition of EVA to the blend
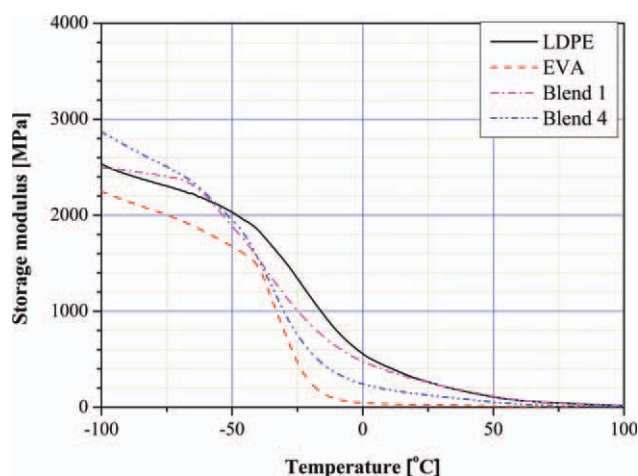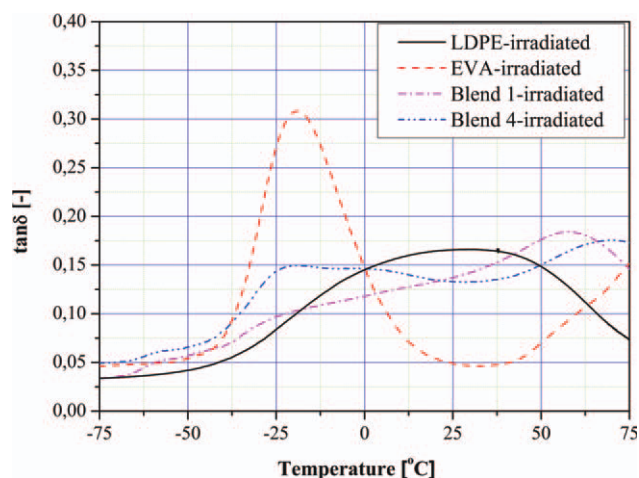
(Blend 4) results in a rubber-like material with low modulus (around 150 MPa). In case of Blend 1 and 4, higher storage modulus can be obtained below −60°C. This is due to the glassy state of GTR (its T_g is around −60°C).

Figures 4 and 5 show the effect of postirradiation on tan δ and storage modulus–T curves, respectively. It can be seen that the irradiation affected the intensity and the position of tan δ peaks in case of LDPE and LDPE based blends, and especially the α-transition of LDPE. For EVA, the irradiation affected mostly its molecular structure, and therefore, hindered melting can be resolved in the studied range due to crosslinking (cf. the tan δ curve of EVA around 70°C in Figures 2 and 4).

Another difference is in the shape of the pure LDPE curve, the tan δ values around 0°C increased

Figure 3 Storage modulus curves of the components and Blends 1 and 4 (nonirradiated). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
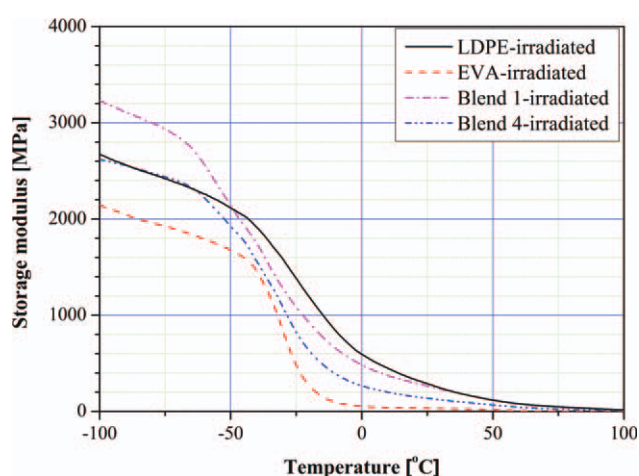

Figure 5 Storage modulus curves of the components and Blends 1 and 4 (irradiated). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
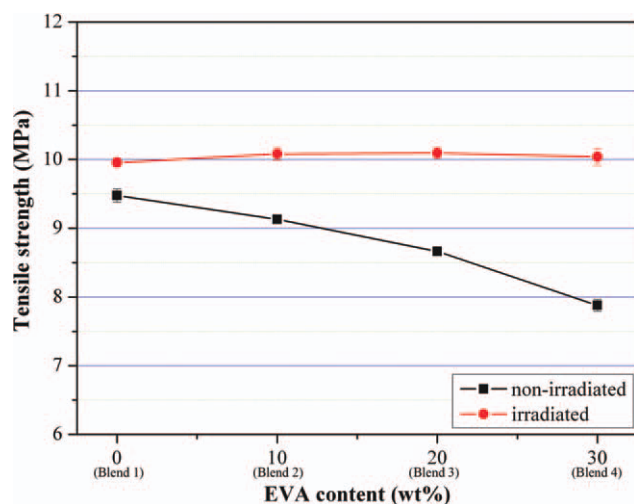
Figure 6 Tensile strength as a function of EVA content in irradiated and nonirradiated blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Figure 8 Tensile modulus as a function of EVA content in irradiated and nonirradiated blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

slightly when compared with the nonirradiated sample. It can be explained by the higher branching content caused by the crosslinking effect of the irradiation. The storage modulus curves reveal that the postirradiation increased this parameter in case of Blend 1 and decreased in case of Blend 4 below the T_g. This result can be explained by the irradiation caused crosslinking effect between the rLDPE and LDPE. In case of Blend 4, the irradiation may cause bonds between the EVA and the rLDPE, so the storage modulus decreased. Above the T_g of EVA, irradiation resulted in practically no difference in the storage modulus compared with the nontreated materials.
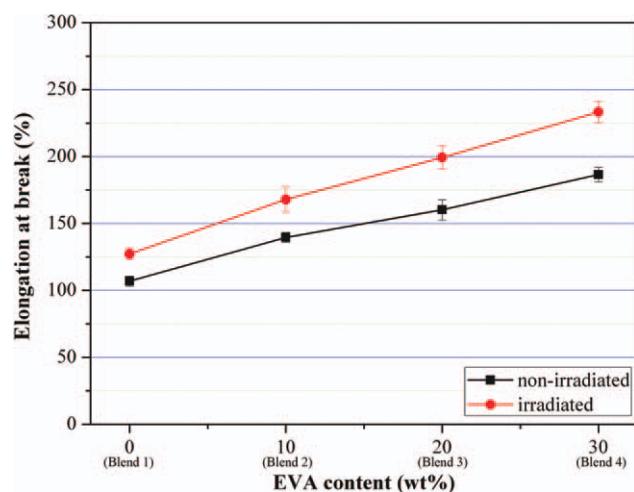

Figure 7 Elongation at break as a function of EVA content in irradiated and nonirradiated blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
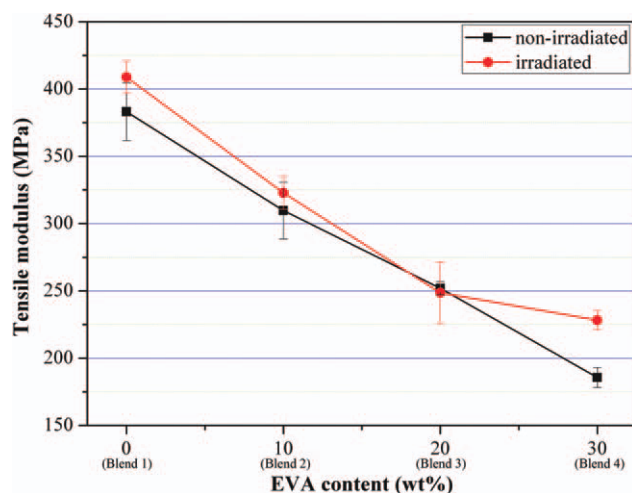
Figure 9 Average values of the deformation components as a function of EVA content for nonirradiated (a) and irradiated (b) blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Figure 10 Perforation energy as a function of EVA content in irradiated and nonirradiated blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
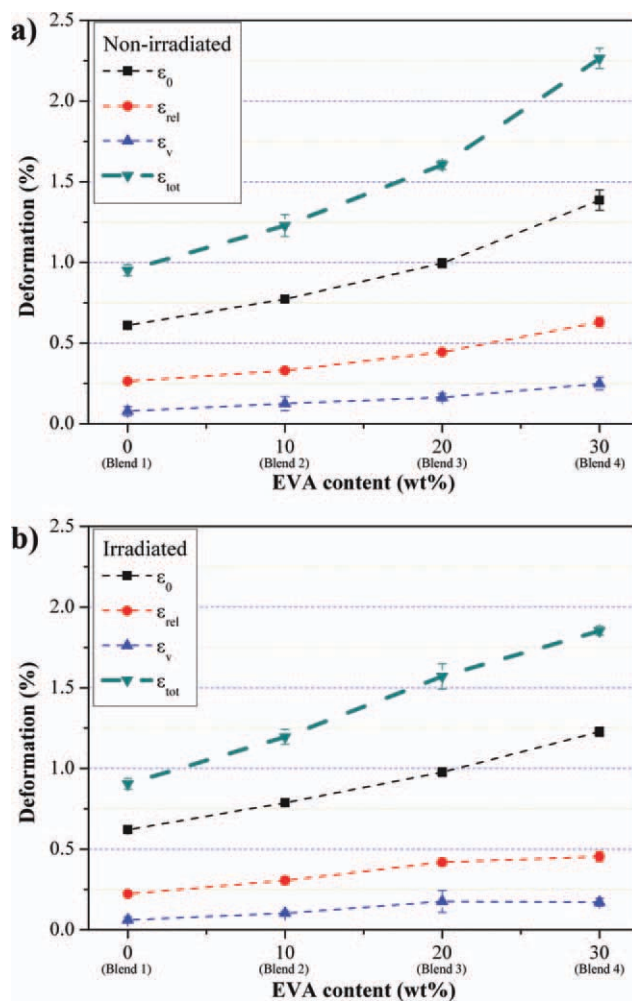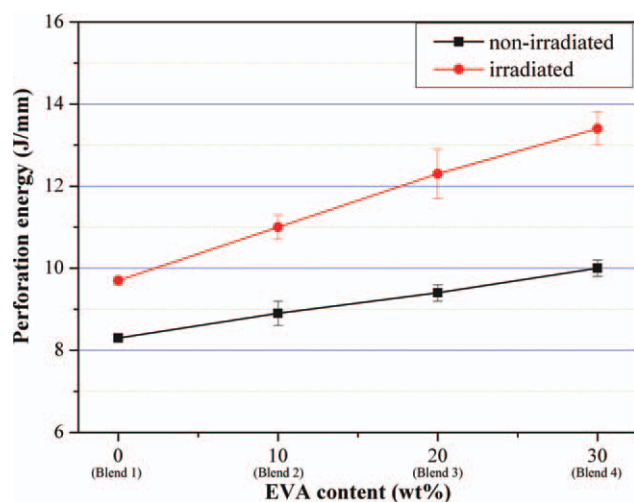
Hysteresis and tensile tests

Figure 6 shows the tensile strength values for different blends before and after irradiation. Based on the results, it can be stated that in case of nonirradiated blends the tensile strength decreases (maximum of 20%) monotonously with increasing EVA content. Recall that parallel to increasing EVA content, the fresh LDPE content decreases. In case of irradiated blends, there is no change in the tensile strength as a function of EVA content. This means that irradiation can compensate the effect of changes in the material composition on the properties. The most significant positive effect can be noticed for Blend 4: as a result of postirradiation about 30% higher tensile strength is obtained. Hence, the tensile strength of each investigated blend approaches that of the original LDPE (10 MPa) provided in the datasheet of the producer.

If the EVA—the elongation at break of which is several hundred percentages—content is increased, this parameter increases for the blends, as well. Accordingly, elongation at break increases significantly for both nonirradiated and irradiated blends (Fig. 7). The positive effect of irradiation can be seen again: for each blend this parameter increases by at least 20%. Note that for multicomponent blends, the fracture behaviour depends strongly on the interfacial bonding between the phases. The electron beam irradiation can increase the interaction between phases, so the resistance to crack propagation can be enhanced. This results in higher elongation at break that refers to the effectiveness of electron beam irradiation in compatibilization. Furthermore, with increasing EVA content, the effect of irradiation becomes more significant. To sum it up, it can be stated that both EVA and irradiation increase elongation at break.

Figure 8 shows the tensile modulus as a function of EVA content. It can be seen that the tensile modulus decreases considerably with increasing EVA content, regardless of irradiation. Compared the tensile moduli of treated and untreated blends, it can be seen that irradiation improved the modulus slightly (except for Blend 3). For Blend 4 (containing 30 wt % EVA), irradiation enhanced the modulus from 185 to 228 MPa, and, for Blend 1, from 383 to 408 MPa.

Based on the description in Specimens and Their Testing section, the estimated values of the deformation components were determined with the help of the hysteresis. The total and the derived deformation components are shown in Figure 9(a). A definite increment can be seen for all the three components in case of all blends with increasing EVA content. For each blend, ε_{01} (this may characterize the rubber-like properties) accounts to two third of ε_{tot1}. By increasing EVA content from 0 to 30 wt %, ε_{tot1} increases
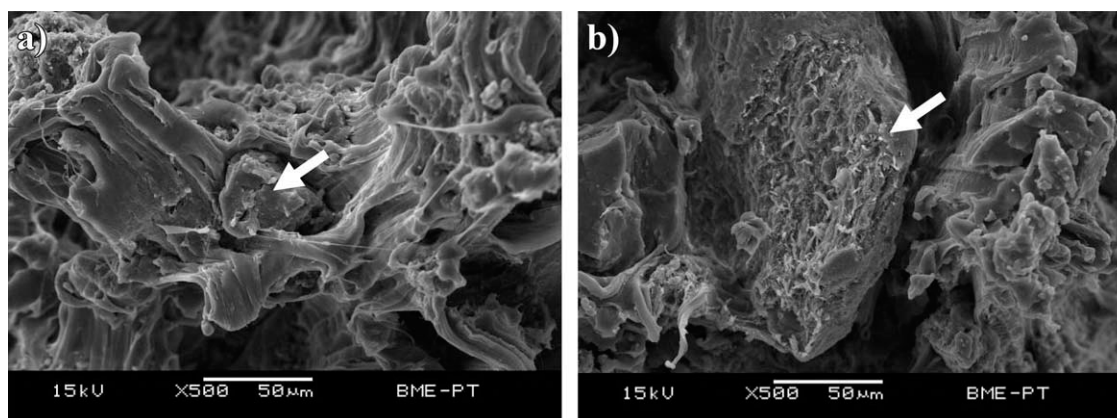

Figure 11 SEM pictures of Blend 1 without irradiation (a) and with irradiation (b) (white arrows designate the GTR particles).
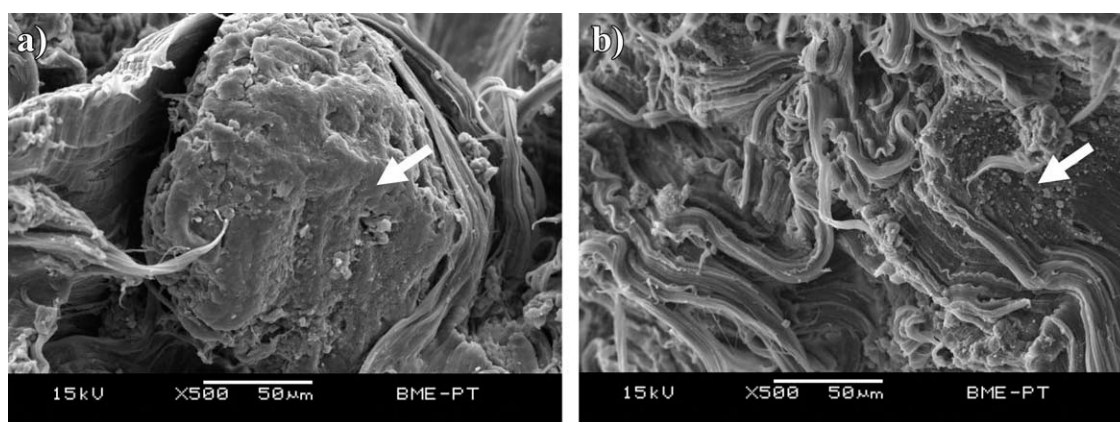
Figure 12　SEM pictures of Blend 4 without irradiation (a) and with irradiation (b) (white arrows designate the GTR particles).

by approximately 130%, while ε_{01} by approximately 100%; however, the proportion of ε_{01} remains 60–65% within the total deformation.

In case of the blends irradiated with an absorbed dose of 150 kGy, a similar tendency is obvious [Fig. 9(b)]. The total deformation is lower in Blend 4, so the derived deformation components are also lower compared with nonirradiated blends [Fig. 9(a)]. The time-dependent viscous deformation shows slightly lower values for each irradiated blends. The irradiated blends having lower EVA content (up to 20 wt %), the ε_{01} values did not change compared to the nonirradiated ones except Blend 4. The time-dependent component decreased slightly owing to the irradiation. This means that the instantaneous deformation became even more dominant after irradiation.

IFWI tests

Falling weight impact tests were performed to assess the materials' response for out-of-plane loading. Perforation energy values were determined from the IFWI tests. Figure 10 shows the thickness related perforation energy as a function of EVA content. One can see that Blend 4 has the highest energy absorbing capability, that is, the highest perforation energy. Both the increasing EVA content and postirradiation increase the perforation energy. However, with increasing EVA content (with decreasing fresh LDPE content) the effect of irradiation is getting more and more significant (difference between treated and untreated specimens is between 16.5 and 34.5%). By adding 30 wt % EVA and thereafter irradiating the blends, the perforation energy can increase by more than 60% compared with the nonirradiated Blend 1.

Scanning electron microscopy

The fracture surfaces of broken tensile specimens were investigated with a SEM. It can be seen that postirradiation did not improve the poor interfacial connection between the rubber and polyethylene phases [Fig. 11(a,b)], when EVA was not added to the blend. Therefore, the improvement in the mechanical properties can be assigned to the crosslinking of LDPE. Note that the original and recycled LDPE are connected by primary bonds as a result of crosslinking, so the poorer mechanical property of the rLDPE was compensated by crosslinking.

When 30 wt % EVA was added to the blend (Blend 4—by substituting the fresh LDPE), the matrix material (LDPE + EVA)—besides its crosslinking process—yielded a better interfacial bonding as a result of electron irradiation [Fig. 12(b)] when compared with the nonirradiated Blend 4 [Fig. 12(a)]. This duality is especially advantageous and its effect is also reflected by the improvement in the mechanical properties. The small improvement in the characteristics of Blends 2 and 3 can be explained with the same reasons.

CONCLUSIONS

In this study, thermoplastic elastomers (with 70 wt % recyclate content) based on LDPE and GTR blends were prepared and investigated. EVA copolymer was added to improve the adhesion and to achieve improved rubber-like properties. To enhance the mechanical performance, postirradiation with an absorbed 150 kGy dose was applied.

Based on the tensile test results, it was found that postirradiation affected the parameters advantageously compared with the related nonirradiated blends. It was also established that the tensile strength of irradiated blends was not affected by the EVA content in the blend. The cyclic test results showed that the total deformation increased with increasing EVA content, analogously to the static elongation at break values. The proportion of the deformation components remained constant, but all of

them increased with increasing EVA content. After irradiation the instantaneous deformation became even more dominant. Results of the DMTA tests showed that both increasing EVA content and postirradiation beneficially influenced the thermomechanical properties due to the improved interfacial connection between the components of the blends.

SEM pictures revealed that the GTR particles were embedded in the matrix material (blend of rLDPE/LDPE/EVA); however, the interfacial connection was poor. EVA incorporation and postirradiation yielded better interfacial bonding between GTR and matrix, which was manifested in improved rubber-like properties compared with those of the nonirradiated blends with less or no EVA.

This work is connected to the scientific program of the "Development of quality-oriented and harmonized R + D + I strategy and functional model at BME" project.

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