

THERMOMECHANICAL PROPERTIES OF RADIATION-MODIFIED POLYETHYLENE/ETHYLENE-PROPYLENE-DIENE COPOLYMER/LIQUID-CRYSTALLINE COPOLYESTER BLENDS

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Received July 2, 1998

Accepted November 24, 1998

Radiation-modified blends of high-density polyethylene (PE) with ethylene-propylene-diene copolymer (EPDM; 10–65 wt.%) and thermotropic liquid-crystalline polymer (LCP; 10 wt.%) were investigated. The LCP was a liquid-crystalline copolyester of 40% poly(ethylene terephthalate) with 60% 4-hydroxybenzoic acid. The constituents were blended using a circular extruder, the specimens were prepared by compression molding and irradiated by ⁶⁰Co γ -radiation up to 200 kGy. Mechanical, thermal, and morphological properties in wide temperature range were investigated for the irradiated and non-irradiated specimens. The effects of irradiation on the thermomechanical behaviour of the PE matrix are discussed. The LCP addition significantly affected the stress-strain behaviour of PE at temperatures above its melting point resulting in some anomalies in kinetics of formation of thermorelaxation stresses, which arise if the material is previously oriented. Thermosetting properties (thermorelaxation stresses and residual setting stresses under conditions of isometric heating and cooling) of the PE/EPDM/LCP blends crosslinked by irradiation and then oriented were also established. The results obtained suggest that such ternary blends improve the application properties of irradiated polyethylene and obtain useful thermosetting materials.

Key words: Poly(ethylene-co-propylene-co-dicyclopentadiene); Poly[ethylene terephthalate-co-(4-hydroxybenzoic acid)]; Radiation-modified blends; Elastic modulus; Thermorelaxation stresses; Setting stresses; Polyethylene; Copolymers.

At present, development and investigation of blends of thermotropic liquid-crystalline polymers (LCP) with traditional and less expensive engineering thermoplastics is of increasing interest. It is known that the addition of LCP effects self-reinforcement directly during the processing, affording composites *in situ*, viz. thermoplastic composites reinforced with rigid and strong liquid-crystalline fibrils¹⁻³.

There exist few studies on the effect of addition of LCP on properties of polyethylene (PE) with various densities, representing the class of polyolefins. One of the reasons is that the melting point of crystalline PE is much lower than the temperatures at which the known LCP passes to a liquid-crystalline state. In such conditions, the disperse liquid-crystalline phase has not yet reached the liquid-crystalline state, the PE/LCP blend behaves as a filled system, *i.e.*, LCP particles are not deformed in the melt stream, and, therefore, highly oriented liquid-crystalline fibrils do not form. The ratios of viscosity of the system components can decisively affect the structure of processed blends. This was, for example, shown in ref.⁴, where the melt viscosity, thermal and mechanical properties, as well as the morphology of blends of PE with high- and low-density polyethylene and an LCP with melting point 229 °C, were investigated. This work also shows some interesting features of mechanical properties of the PE/LCP blends. The elongation at break of compression-molded specimens of high-density polyethylene/LCP blends displayed a strong reduction of that elongation upon addition of small (10%) amounts of LCP, whereas the tensile modulus did not vary appreciably, over the 0–20% LCP range investigated. Therefore, at small contents of LCP, as it was shown in ref.⁴, it was impossible to reach a significant modification of the PE/LCP composite properties.

It is well known that physicomechanical and chemical properties of polyolefins and their blends can be considerably changed by radiation modification, *i.e.*, by ionizing radiation^{5–8}. In PE exposed to γ -radiation or to accelerated electrons, a spatial molecular structure is formed the degree of crosslinking of which depends on the irradiation dose absorbed. One of the most useful modification manner is a preparation of thermosetting materials (TSM). At this stage there appears a striking property of the radiation-modified PE, the so-called form-memory effect. This property is manifested in the irradiated PE, when a previously oriented sample is heated at a temperature above the melting point of the crystalline phase and then cooled under isometric conditions.

Our previous investigation shows that the LCP addition significantly affected the stress–strain behaviour of the irradiated high-density polyethylene at temperatures above the melting point and also thermosetting properties of the high-density polyethylene/LCP blends previously irradiated and oriented suggesting that the LCP addition makes it possible to improve properties of TSM (ref.⁹).

The aim of the present paper is to continue the study on the thermomechanical properties of γ -irradiated high-density polyethylene/LCP blends on addition of elastomer phase to modify the properties of composite. The

main attention is paid to the influence of LCP additives on the form memory effect and to the possibilities of increasing the efficiency of thermosetting products made from these composites.

EXPERIMENTAL

Materials

High-density polyethylene (PE) of trademark 20708-016 produced by Novopolotsk Chemical Enterprise (Belarus) with the following characteristics was used: density 0.952 g/cm^3 , melt index 2.0 g/10 min , melting point $131 \text{ }^\circ\text{C}$ (from DSC data), softening temperature $125 \text{ }^\circ\text{C}$.

Ethylene-propylene-diene (EPDM) commercial copolymer SKEPT-40 (Russia) with propylene contents 40 mole %, dicyclopentadiene content 0.5–2 mole %, density 0.87 g/cm^3 , and viscosity 40° Moony was used as an elastomer.

The 40 : 60 copolyester of poly(ethylene terephthalate) (PET) with 4-hydroxybenzoic acid (HBA) (LC-3000, Unitika Ltd., Kyoto, Japan), was used as an LCP. This copolymer has several relaxation transitions¹⁰ because it comprises two phases, the first enriched by PET, the other by HBA. In the first phase, the α -transition occurs at $62 \text{ }^\circ\text{C}$ and the α' -transition in the range of 80 – $100 \text{ }^\circ\text{C}$. The glass transition temperature in the second phase is $160 \text{ }^\circ\text{C}$, while the melting point is $199 \text{ }^\circ\text{C}$.

PE/EPDM/LCP blends were prepared in a circular extruder at $230 \text{ }^\circ\text{C}$ by compression molding at $200 \text{ }^\circ\text{C}$, plates $100 \times 70 \times 1 \text{ mm}$ were made of neat PE and of ternary blends of PE containing 0, 10, 25, 50, and 70 wt.% of EPDM and 10 wt.% of LCP (for all composites). For comparison, PE/EPDM blends mixed and molded at $160 \text{ }^\circ\text{C}$ were used.

Radiation treatment. Samples in the form of plates were irradiated by an experimental ^{60}Co RHM-20 γ -radiation source in an argon atmosphere to obtain various levels of the absorbed dose. The maximum absorbed dose was 200 kGy ($1 \text{ Gy} = 100 \text{ rad}$) at 10 kGy/h . The dose and degree of crosslinking were controlled by an ultrasonic method¹¹.

Methods of Investigation

Calorimetric tests. The melting parameters (temperature, T_m , and enthalpy, ΔH_m) were determined by DSC using a Mettler TA 4000 instrument. Indium was taken as a standard for the heat capacity calibration. The temperature-scanning rate was $10 \text{ }^\circ\text{C/min}$. The 10 mg samples were heated up to $250 \text{ }^\circ\text{C}$.

Morphological tests. The morphology of samples of the blend composites before and after irradiation (also oriented) was investigated using a microscope LEITZ LABORLUX 11 POL S.

Thermomechanical tests. The dependence of the elastic modulus on temperature was examined in the range of 20 – $200 \text{ }^\circ\text{C}$ by quasistatic loading on a stand designed for creep tests and fitted with a heating chamber. The sample was periodically loaded as the temperature increased; then the strain was measured and the load immediately removed. The loading-unloading cycle took 1 s. The assigned strain did not exceed 1% since the applied load was reduced with increasing temperature. At all temperatures, the strains in loading and unloading were approximately the same, *i.e.*, no marked residual strains were found. The elastic modulus was defined as the ratio of the applied stress to the measured strain.

Thermosetting effects in the radiation-crosslinked specimens were investigated after their preliminary orientation at increased temperatures (140 – $200 \text{ }^\circ\text{C}$) by extension up to 100%

and subsequent cooling to room temperature in the isometric fixed state. Thermo-mechanical behaviour of the specimens prepared with repeated heating was determined in the fixed state. In such isometric conditions, thermorelaxation stresses, σ_t , which arose when the specimens was heated, as well as setting stresses, σ_s in the subsequent isometric cooling were estimated. After cooling to room temperature, residual setting stresses are obtained. The specimens were $50 \times 8 \times 1$ mm strips. The tests were carried out on a special apparatus fitted with a heating chamber and a mechanism for the force indication. The thermosetting force was measured tensometrically: one of the clamps was fastened to a graduated springy steel plate, with four tensoresistors attached to it. The sensitivity of the device ± 0.01 N. The heating and cooling rates were $1^\circ\text{C}/\text{min}$.

RESULTS AND DISCUSSION

The initial-molded specimens of neat PE and 90 PE/10 LCP blends were tested in our previous work⁹. Specimens preliminary tested in tension at room temperature showed that on addition of LCP only slightly affected the general shape of tensile diagrams. For initial-molded specimens of 90 PE/10 LCP, the limit of recrystallization, σ_c , increases from 23.6 to 26.1 MPa, elastic modulus, E , increases from 0.81 to 1.00 GPa, but elongation-at-break, ε_b decreases from 1 560 to 1 000%, compared with neat PE (in this investigation the high-density PE of trademark BP Rigidex HD 5740 3UA was used).

Figure 1 shows the temperature dependences of modulus for the investigated specimens⁹. The temperature dependence of the elastic modulus of a non-irradiated 90 PE/10 LCP blend is similar to the dependence $E(T)$ for neat PE. Within the investigated range of temperatures (20–140 °C), the value of E for the composite is only 10–20% higher than for PE (see curves

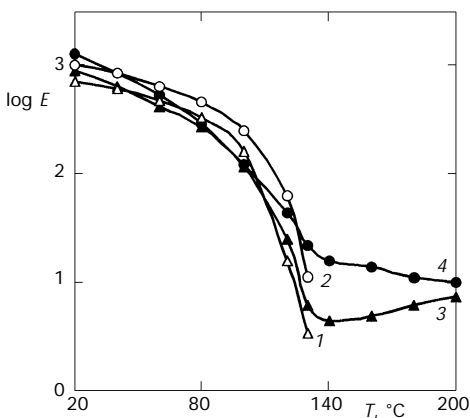


FIG. 1
Temperature dependence of the elastic modulus (E , MPa) of non-irradiated PE (1) and 90 PE/10 LCP blend (2), and of irradiated PE (3) and 90 PE/10 LCP blend (4) (irradiation with an absorbed dose 150 kGy)

1 and 2 in Fig. 1). This is not strange because the LCP fraction in the blend is quite small and fibrillation of the LCP particles in the PE matrix during molding is not significant.

After irradiation with an absorbed dose of 150 kGy, the elasticity of PE prior to the melting of crystallites is mainly determined by the presence of the crystalline phase. At this irradiation dose, transverse bonds appears mainly in the amorphous regions of PE while the degree of crystallinity varies insignificantly, which is confirmed by the DSC data⁹. An accelerated decrease in the elastic modulus of PE is observed above 90–100 °C that is caused by a decrease in the crystalline phase content. If the melting is completed, the irradiated PE passes into a rubberlike state with its typical conformational elasticity (see curve 3 in Fig. 1). Specimens of 90 PE/10 LCP show a different kind of curve (curve 4). Within the range 140–200 °C (after complete melting of PE crystallites), the value E of the blend continues to decrease monotonically, in contrast to the elastic modulus of irradiated PE.

The nature of adherence between the particles of LCP and the PE matrix is not clear enough; however, we can assume the existence of crosslinking of LCP with PE. As final copolyester of LCP contained segments HBA and PET (ref.¹¹), it is possible, by analogy with neat PET, that a mean active radical, $-C_6H_4-O-CO-\dot{C}H-CH_2-O-$ (ref.¹²), is grafting to the PE matrix. The results of morphologic tests (SEM) show pronounced homogenization of the structure, formation of transition phase, and fibrillation growth⁹. Similar results we discover using optical microscope (see Figs 9 and 10).

Let us compare the temperature dependences of the elastic modulus for non-irradiated and irradiated molded specimens of PE/EPDM/LCP blends

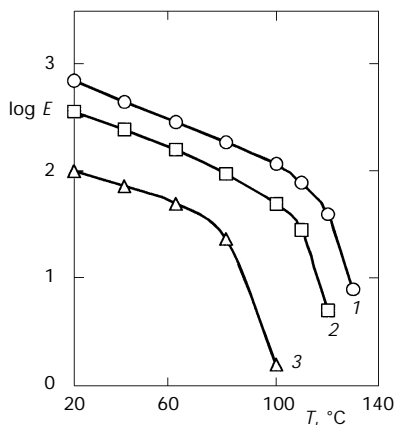


FIG. 2

Temperature dependence of the elastic modulus (E , MPa) of non-irradiated blends 80 PE/10 EPDM/10 LCP (1), 45 PE/45 EPDM/ 10 LCP (2), and 25 PE/65 EPDM/10 LCP (3)

(Figs 2 and 3). The values of E at 20 °C for the blend decrease with increasing of EPDM content. Equally decreases of E values are evident at higher temperatures (see curves 1–3 in Fig. 2). Crosslinking (mostly PE and EPDM) of both phases affects E at higher (90–200 °C) temperatures. In such cases (in the presence of elastomer EPDM), we can not see the monotonically decreasing E values, which appear for blends without EPDM, *i.e.* PE/LCP blends (see curve 4 in Fig. 1). Strangely enough, we cannot see a typical rubber-like curve (see curve 3 in Fig. 1) even though the crosslinked elastomer content increases (see curves 1–3 in Fig. 3). Probably, the structure features in the presence of LCP affect elastic properties of the blend at higher temperatures. Therefore, it is of special interest to compare tensile diagrams of the irradiated neat PE with PE/LCP and PE/EPDM/LCP blends at elevated temperatures; the dependence of stress, σ , on strain, ϵ , is shown in Fig. 4. The tests were carried out at 150 °C, *i.e.*, after complete melting of PE crystallites. It can be seen that irradiated PE behaves like a typical representative of rubber-like polymers. Blends with the EPDM content 10–65 wt.% show the same form of curves, however, the presence of 10 wt.% LCP affected the stress–strain characteristics. With extension of 200–400%, the neat PE and all PE/EPDM/LCP composites fail. We can notice that the slopes of the initial parts of the stress–strain curves in Fig. 4 are in good agreement with the results shown in Fig. 3.

The tensile diagram for the composite 90 PE/10 LCP is quite different. At 100% strain, the stress growth sharply slows down, and the specimen continues deforming up to 1 000% exhibiting signs of flowing (see curve 2 in Fig. 4). Outwardly, the dependence $\sigma(\epsilon)$ for PE/LCP blend is similar to that

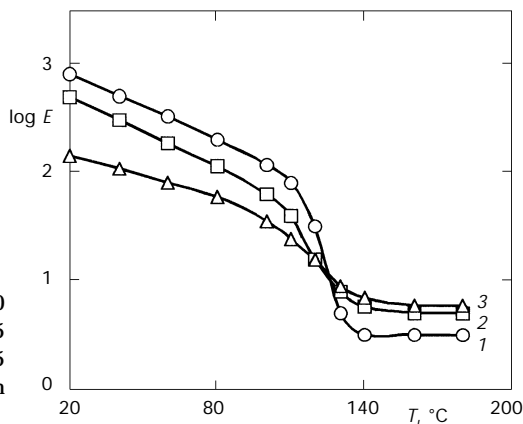


FIG. 3

Elastic modulus (E , MPA) of blends 80 PE/10 EPDM/10 LCP (1), 45 PE/45 EPDM/ 10 LCP (2), and 25 PE/65 EPDM/10 LCP (3) (irradiation with an absorbed dose of 150 kGy)

of a tetrafunctional structure strongly solvent-swollen, which satisfies the known Mooney–Rivlin equation. Apparently, the particles of LCP dispersed within the volume of a crosslinked structure of PE shape the transition phase (as a result of crosslinking of PE and LCP) and produce a plasticizing effect on the blend. Therefore, we can claim that the addition of LCP considerably increases the deformability of the radiation-crosslinked PE at temperatures above the melting point of the crystallites. The presence of considerably crosslinked elastomer EPDM phase reduces these properties.

The results of defining the stresses in irradiated oriented of PE/EPDM and PE/EPDM/LCP blends, which arise upon repeated isometric heating and cooling, examine their form-memory. The specimens irradiated with 150 kGy were previously stretched at 150 °C and then isometrically cooled to 20 °C. The dependence of thermorelaxation stresses on temperature shows different curve forms for PE/EPDM and PE/EPDM/LCP blends (Figs 5 and 6). Addition of LCP considerably affects the development kinetics and values of σ_t . The $\sigma_t(T)$ curve for the 90 PE/10 LCP blend, in contrast to the curves for neat PE and PE/EPDM/LCP blends, has a peculiar three-stage form (see curve 1 in Fig. 6); the composition 80 PE/10 EPDM/10 LCP shows a negligible effect. As it was defined in ref.⁹, the stepwise form of the σ_t curve and the extended temperature range of the σ_t development were caused by the relaxation of the α - and α' -transitions in LCP. This relaxation also affects the kinetics of the isometric cooling process. Thermorelaxation stresses increase with the increasing content of EPDM because the considerably crosslinked amorphous phase of EPDM increases these stresses (Fig. 5). In the presence of LCP, the crosslinking in EPDM phase is probably impeded,

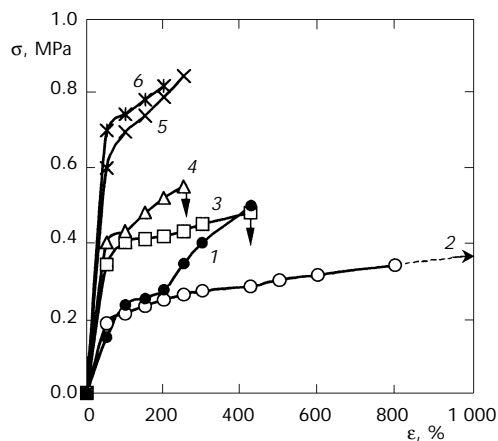


FIG. 4

Tensile stress–strain behaviour at 150 °C of PE (1), 90 PE/10 LCP (2), 80 PE/10 EPDM/10 LCP (3), 65 PE/25 EPDM/10 LCP (4), 45 PE/45 EPDM/10 LCP (5), and 25 PE/65 EPDM/10 LCP (6) (irradiation with an absorbed dose of 150 kGy)

and the mentioned increase in σ_t is not observed (Fig. 6, curve 1 in Fig. 7). The maximum thermorelaxation stresses are much greater than for neat PE; thus, the claimed twice-greater stresses withstand heating up to 250–280 °C without failing and allow to manufacture and use thermoshrinkable products with enhanced application parameters. Figure 8 shows the dependence of residual setting stresses, on the EPDM content; a decrease is a rule because crystallinity of blend decreases.

Higher absorbed doses, up to 200–250 kGy, were also used in the radiation treatment but in this case, parameters such as thermorelaxation stresses decrease because decomposition of LCP proceeds, the fact being de-

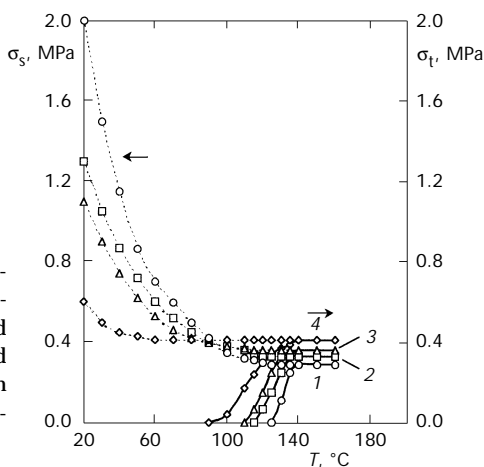


FIG. 5
Thermorelaxation stresses, σ_t (—), and setting stresses, σ_s (- -), upon isometric heating and cooling for PE (1) and for irradiated 70 PE/30 EPDM (2), 50 PE/50 EPDM (3), and 30 PE/70 EPDM (4) blends (irradiation with an absorbed dose 150 kGy) and 100% oriented

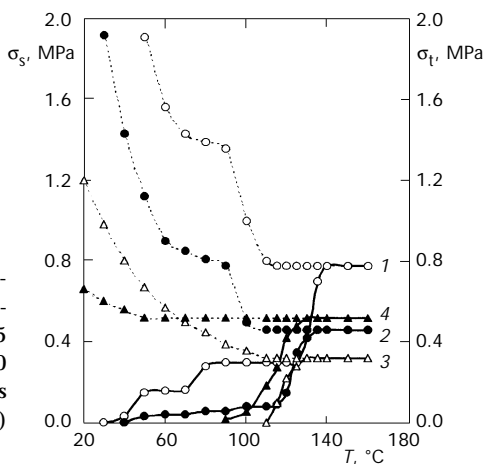


FIG. 6
Thermorelaxation stresses, σ_t (—), and setting stresses, σ_s (- -), upon isometric heating and cooling for 90 PE/10 LCP (1), 45 PE/45 EPDM/10 LCP (2), 80 PE/10 EPDM/10 LCP (3), 25 PE/65 EPDM/10 LCP (4) blends (irradiation with an absorbed dose 150 kGy) and 100% oriented

terminated in ref.⁹. For comparison, Fig. 7 shows the dependence of σ_t on the content of EPDM for PE/EPDM/LCP blends irradiated with absorbed doses 150 and 200 kGy.

The results of morphological and calorimetric tests have already been mentioned. The microphotos (Fig. 9) present the microstructure of 90 PE/10 LCP specimens, both non-irradiated and irradiated with an absorbed dose 150 kGy, as well as irradiated and stretched up to 100%. On a microphoto of the non-irradiated specimens, LCP particles dispersed in the matrix can be

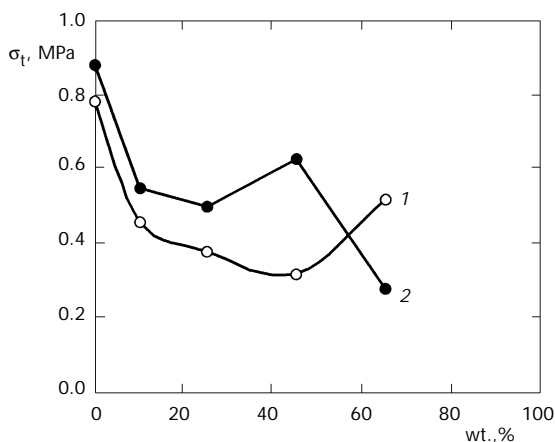


FIG. 7

Thermorelaxation stresses, σ_t , of PE/EPDM/LCP (10 wt.%) irradiated with absorbed doses 150 (1) and 200 kGy (2) versus EPDM content

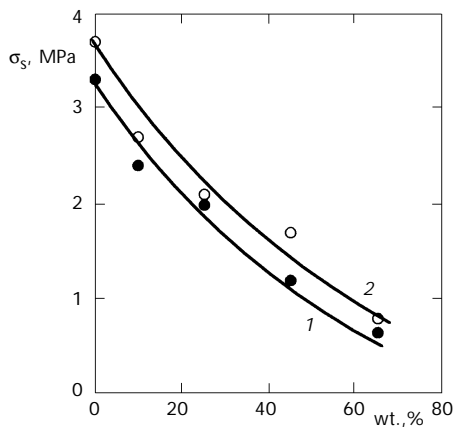


FIG. 8

Residual setting stresses of PE/EPDM/LCP (10 wt.%) irradiated with absorbed doses 150 (1) and 200 kGy (2) versus EPDM content

seen. The morphology of the irradiated specimens differs in a pronounced homogeneity of the structure, formation of a transition phase, and growth of fibrillation. The structure homogeneity may be caused by structural modifications due to direct exposure to ionizing radiation. Another reason may be the orientation effects developed in the matrix in the presence of LCP particles by increasing temperature in the irradiation chamber ($\approx 60^\circ\text{C}$). Its temperature is similar to the temperature of α -transition of LCP. The stretching of the sample at 150°C shows that orientation of LCP particles does not appear; only some of them were observed. This may be because they affect, as was seen, the deformation and shrinking properties of the blend. Figure 10 presents the blend microstructure on addition of 45 wt.% of EPDM, where also the homogeneity of the structure can be seen. In the case of stretched specimens partial orientation of LCP particles can be observed. Probable reason is the crosslinking of LCP with the EPDM phase.

The values of melting point, T_m , and enthalpy of melting, ΔH_m , determined from the results of calorimetric tests of non-irradiated and irradiated samples, are presented in Table I. The ΔH_m of neat LCP is relatively small

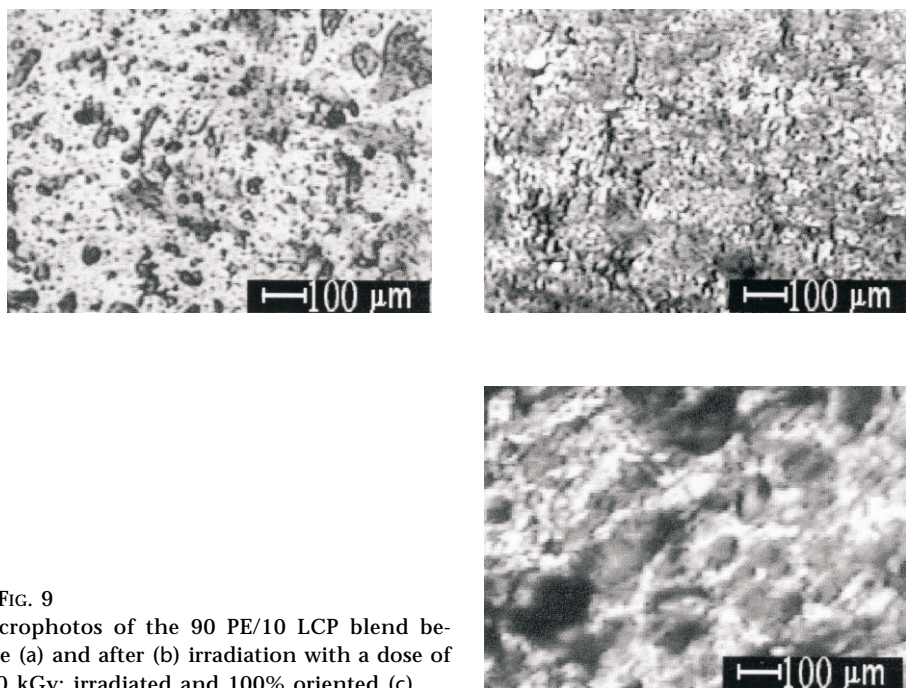


FIG. 9

Microphotos of the 90 PE/10 LCP blend before (a) and after (b) irradiation with a dose of 150 kGy; irradiated and 100% oriented (c)

TABLE I
Values of melting temperature, T_m , and melting enthalpy, ΔH_m , for PE/EPDM/LCP blend specimens

PE/EPDM/LCP Composition	Treatment	T_m °C	ΔH_m J/g
100/0/0	non-irradiated	131.0	171.3
100/0/0	150 kGy	125.6	187.6
90/10/0	non-irradiated	127.0	134.1
90/10/0	150 kGy	128.5	168.1
50/50/0	non-irradiated	126.9	87.1
50/50/0	150 kGy	124.0	83.5
90/0/10	non-irradiated	130.3	176.7
90/0/10	150 kGy	127.8	164.7
80/10/10	non-irradiated	132.1	153.6
80/10/10	150 kGy	131.4	153.1
45/45/10	non-irradiated	130.9	103.9
45/45/10	150 kGy	120.3	103.2

TABLE II
Tensile strength-at-break, σ_b , and elongation-at-break, ϵ_b , of PE/EPDM/LCP blends

PE/EPDM/LCP Composition	Treatment	σ_b MPa	ϵ_b %
90/0/10	non-irradiated	16.5	46
90/0/10	150 kGy	16.5	47
65/25/10	non-irradiated	9.7	50
65/25/10	150 kGy	12.7	65
45/45/10	non-irradiated	6.9	97
45/45/10	150 kGy	9.6	120
25/65/10	non-irradiated	2.4	240
25/65/10	150 kGy	4.4	250

(6 J/g); therefore, the second melting point of LCP in blends containing 10 wt.% of LCP is not observed. Mechanical properties at 20 °C are shown in Table II.

Changes in ΔH_m observed for blends with similar EPDM content (45–50 wt.%) suggest that LCP affect a possible partial degradation of PE at the mixing temperature (230 °C), which causes the growth of crystallinity degree as well as of the enthalpy.

CONCLUSIONS

1. The PE/EPDM, PE/LCP, and PE/EPDM/LCP blends previously irradiated with the absorbed dose 150 kGy and 100% oriented by tension demonstrate the form-memory effect in repeated heating.

2. An addition of the LCP phase up to 10 wt.% allows to increase considerably (several times) the deformability (the achieved degree of extension) of radiation-crosslinked PE at temperatures above the melting point of its crystallites.

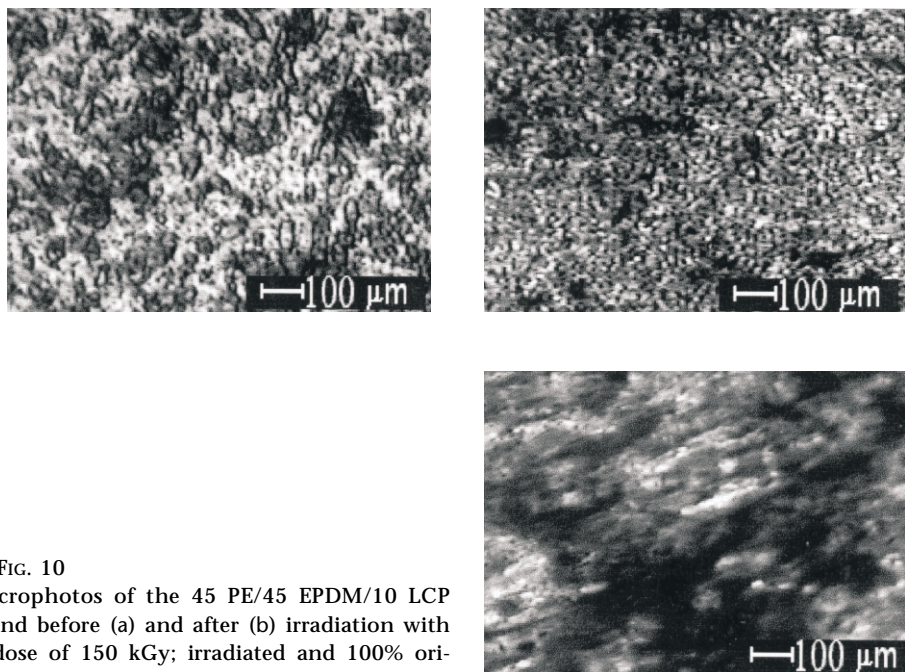


FIG. 10
Microphotos of the 45 PE/45 EPDM/10 LCP blend before (a) and after (b) irradiation with a dose of 150 kGy; irradiated and 100% oriented (c)

3. On addition of LCP affects the development kinetics and the value of thermal relaxation stresses (σ_t). Specimens of neat PE fail due to the σ_t arising at temperatures above 200 °C, whereas the 90 PE/10 LCP specimens subjected to σ_t twice as great as the thermorelaxation stresses withstand heating up to 250–280 °C without failure.

4. On addition of the EPDM phase (10 wt.% and more) considerably decreases the deformability of radiation-crosslinked PE/LCP blend at temperatures above the 150 °C, at the same time showing a sufficient form-memory effect, which allows to produce rubber-like thermosetting materials.

5. The described features of the stress development in radiation-modified blends of high-density polyethylene with elastomer and liquid-crystalline copolyester in isometric heating and cooling agree, in general, with the results of morphological and calorimetric tests and are important for improving performance of thermoshrinkable products.

The Latvian Council of Science financially supported this study.

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