# Study of Heat Shrinkability of Crosslinked Low-Density Polyethylene/Poly(ethylene vinyl acetate) Blends

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**ABSTRACT:** In this study, the heat-shrinkage property in polymer was induced by first compounding low-density polyethylene/poly(ethylene vinyl acetate) (LDPE/EVA) blends with various amounts of peroxide in a twin-screw extruder at about 130°C. The resulting granules were molded to shape and chemically crosslinked by compression molding. A process of heating–stretching–cooling was then performed on the samples while on a tensile machine. Shrinkability and effective parameters were also investigated using thermal mechanical analysis. The results showed that the gel fraction was higher for the sample of higher EVA content with the same amount of dicumyl peroxide (DCP). A decrease in the melting point and heat of fusion ($\Delta H_f$), as determined from DSC, was observed with an increase in the DCP content. Studies on the heat shrinkability of the samples showed that samples stretched above the melting point had a higher shrinkage temperature than those stretched around the crystal transition temperature. The results showed that by increasing the peroxide content, the shrinkage temperature was decreased. These could be attributed to the formation of new spherulites as well as changes in the amount and the size of crystals. Furthermore, in samples elongated at 120°C (above the melting point), the rate of stretching had no effect on the shrinkage temperature. The results showed that the extent of strain had no effect on the temperature of shrinkage, but rather on the ultimate shrinkage value. © 2003 Wiley Periodicals, Inc. J Appl Polym Sci 91: 1389–1395, 2004

**Key words:** low-density polyethylene; poly(ethylene vinyl acetate); thermal properties; crosslinking; thermoplastics

## INTRODUCTION

Polyethylene is a one of the most important thermoplastics, although its use is restricted in certain applications because of its low melting point, solubility or swelling in hydrocarbons, and tendency to crack when stressed. In an effort to tackle these disadvantages there has been considerable work to improve the properties of polyethylene by crosslinking.[1–4] Crosslinking enables a thermoplastic polymer like polyethylene to exhibit a viscoelastic behavior, characteristic of an elastomer, at temperatures above the crystalline melting point of the thermoplastic.[5,6] This valuable property is widely exploited commercially in the preparation of heat-shrinkable materials. Heat-shrinkable components are widely used in the electric and cable industries for splicing, joining, and termination applications. Semicrystalline polymers such as low-density polyethylene (LDPE), like other polyolefins, are common polymers widely used in these applications.[7–9] At ambient temperature, the molded crosslinked polyethylene retains its shape because of the presence of plastic crystalline domains (see Fig. 1). It is believed that crosslinking usually occurs prefera-

bly in amorphous regions.[10,11] When crosslinked LDPE [Fig. 1, stage (C)] is heated above the melting point, crystalline regions disappear (D), but molecules do not flow because of crosslinking between them. This structure is similar to vulcanized rubber and can be easily deformed by only a small amount of stress. Stage (E) is the stretched case of stage (D). The change of (D) to (E) is reversible such as a vulcanized rubber. If stage (E) is cooled below its melting point under the stretched condition (F), crystalline regions appear again to maintain the deformed shape as in stage (E) even if the tension is released. That is, the stress in the stretched condition can be said to be "frozen." When stage (F) is heated again, the shape quickly recovers to stage (D) and then to stage (C) if cooled at that condition.

The use of poly(ethylene vinyl acetate) (EVA) in blends is most important from the technological point of view. Because of its low temperature flexibility, somewhat rubbery nature, low permeability, and good impact strength it has become the focus of increasing attention as a stretched film for packaging technology.[12] It has drawn interest as a cable insulating material because of the good resistance to stress cracking and because the polymer may be easily crosslinked.[13] The other characteristics that have led poly(ethylene vinyl acetate) to widespread use are low cost, easy processability, excellent chemical resistance, excellent electrical insulation properties, toughness
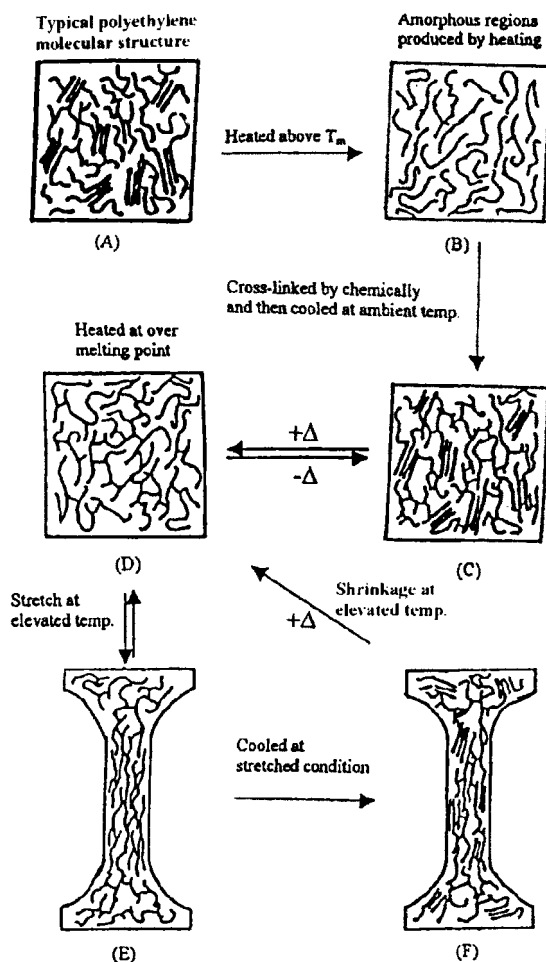
**Figure 1** Mechanism of heat shrinkage: (A) typical polyethylene molecular structure; (B) amorphous regions produced by heating, and then crosslinked chemically; (C) molded unstretched crosslinked polyethylene cooled at ambient temperature; (D) unstretched crosslinked polyethylene heated above the crystalline melting point; (E) polyethylene stretched above meltin point; (F) polyethylene cooled to room temperature in the stretched condition.

and flexibility even at low temperature, freedom from odor and toxicity, reasonable clarity of thin films, and sufficiently low permeability to water vapor. The most important features of poly(ethylene vinyl acetate) for which it has been selected here are its lower response temperature of the shape memory effect, good recovery, and good flow behavior (important for heat-shrinkable materials), and low heat conductivity.[14,15]

The blends prepared by melt mixing of thermoplastic materials have met industrial needs in recent years from the perspective of heat shrinkability.[10–12]

An enormous amount of work has already been carried out on the heat shrinkability of the above-mentioned semicrystalline polymers, but there is little systematic information concerning the shrinkability and effective parameters on the shrinkability of the blends. Recently Khonakdar et al.[19] investigated the

heat-shrinkable behavior of crosslinked low-density polyethylene. They showed that the stretching temperature had a substantial effect on the shrinkage behavior. Also, in another study they obtained the behavior for high-density polyethylene.[20] Das et al.[21] studied the heat-shrinkable behavior of polyolefin blends with elastomer such as chlorosulfonated polyethylene. They indicated the importance of a crosslinked elastomeric phase to enhance the heat shrinkability of the blend system and at the same time to improve flame retardancy. Mishra et al.[22,23] studied the heat-shrinkability behavior of polyolefin blends based on grafted low-density polyethylene and polyurethane elastomer. They suggested that the interchain crosslinking between grafted polyethylene and elastomer improved the shrinkability.

The objective of the present investigation was to develop blends of low-density polyethylene with poly (ethylene vinyl acetate) with respect to their heat shrinkability for commercial applications. In this study, we discuss the preparation of heat-shrinkable crosslinked LDPE/EVA blends as well as the thermal properties and their dependency on the concentration of the crosslinking agent, dicumyl peroxide (DCP). The thermal analysis of the blends was investigated by differential scanning calorimetry (DSC). The behavior of shrinkability and effect of process parameters were investigated by thermal mechanical analysis (TMA).

## EXPERIMENTAL

### Materials

Low-density polyethylene (LDPE; grade LF0200, melt flow index = 1.6 g/10 min, density = 0.923 g mL$^{-1}$) was obtained from BIPC, Iran.

EVA copolymer (with 18% vinyl acetate; trade name Escorene) was supplied by Exxon Chemical (Belgium).

Irganox 1010 antioxidant (density = 1.45 g mL$^{-1}$) was purchased from Ciba-Geigy Co. (Switzerland).

Carbon black masterbatch with the grade of 5003 was prepared from New Particle Color Corp. (Taiwan).

Dicumyl peroxide (DCP; purity = 98%, density = 1.02 g mL$^{-1}$) was obtained from Hercules Co. (Germany).

The blend compositions containing 10 and 20 wt % of EVA are shown in Tables I and II, respectively.

### Preparation of blends

The mixtures of LDPE/EVA and other additives were prepared with different ratios of DCP as shown in Tables I and II, in a Turbo Mixer for 5 min at 1000 rpm. The mixing temperature was 75°C. In this state, DCP was melted and mixed with LDPE/EVA and carbon black masterbatch granules. The mixture was then extruded at relatively low temperature (above melting

### TABLE I
### Mixing Formulations with 10 wt % of EVA[a]

| Sample no. | LDPE (phr) | EVA (phr) | Masterbatch (phr) | Irgannox 1010 (phr) | DCP (phr) |
|---|---|---|---|---|---|
| 1 | 100 | 11.13 | 5.88 | 0.156 | 0.6 |
| 2 | 100 | 11.11 | 5.91 | 0.157 | 1.2 |
| 3 | 100 | 11.09 | 5.94 | 0.158 | 1.8 |
| 4 | 100 | 11.07 | 5.97 | 0.159 | 2.4 |
| 5 | 100 | 11.05 | 6.00 | 0.160 | 3.1 |
| 6 | 100 | 11.03 | 6.04 | 0.161 | 3.6 |

[a] phr, parts per hundred g of polyethylene.

temperature, ~ 130°C). The extrudate was pelletized by a knife pelletizer. The crosslinking of sheet samples was carried out by compression molding on a Davenport 25 ton laboratory hot press at a temperature of 185°C for 5 min under 15 bar pressure, and the samples were then cooled to room temperature.

### Gel measurement

The gel content of the crosslinked LDPE/EVA blends was determined gravimetrically, using a 16-h Soxhlet extraction cycle with $p$-xylene as the solvent at 140°C according to ASTM D 2765. Irganox 1010 was added at 0.5 wt % to inhibit polymer degradation during the extraction. Approximately 0.3 g of the crosslinked polymer sample was cut into small pieces and placed in a preweighed stainless-steel fine-wire mesh. After the extraction cycle, the sample was washed with acetone and vacuum dried to constant weight. The gel fraction was calculated as the percentage ratio of the final weight of the polymer to its initial weight.

### Stretching

The crosslinked samples were stretched at 90 and 120°C at different rates according to DIN 53504 in an Instron 6025 tensile machine (Instron, Canton, MA). Marked lengths of 40 mm on the central length of the dumbbells were taken as the gauge length. The samples were stretched up to 200 and 300% strain, and then were cooled in the stretched condition to room temperature, after which they were removed from the apparatus and conditioned at ambient temperature.

### Thermal analysis

DSC was carried out on a DSC PL thermal analyzer in a nitrogen atmosphere. Samples were heated from 25 to 160°C at a heating rate of 10°C/min and then cooled at the same rate. The specific enthalpy and melting point of samples were determined from the DSC thermograms. TMA tests were done on the specimens to study the heat shrinkability (shrinkage temperature and dimensions of the samples).

### Shrinkage studies

The lengthwise shrinkage (Sh) was measured in nonisothermal condition by TMA (model PL, England) test according to ISO 11501 using the following relationship:

$$Sh\ (\%) = \frac{L_{str} - L_{shr}}{L_{str}} \times 100 \qquad (1)$$

where Sh (%) is the percentage of shrinkage, $L_{str}$ is the original length of the specimen before heating, and $L_{shr}$ is the length of the specimen during shrinking as a function of increasing temperature.

The ultimate shrinkage, sh∞, is the maximum shrinkage achieved and indeed is obtained from the length of the fully shrunk sample. The ultimate shrinkage can be expressed by the following equation:

$$Sh_{\infty} = \frac{L_{str} - L_{shr,\infty}}{L_{str}} \times 100 \qquad (2)$$

where $L_{shr,\infty}$ is the final length of the fully shrunk sample before overheating.

Three parameters—the shrinkage temperature, the shrinkage constant (maximum slope of the curve of changes of shrinkage percentage versus temperature), and the ultimate shrinkage—characterize the shrinkage behavior of stretched crosslinked LDPE/EVA blends.

## RESULTS AND DISCUSSION

### Gel fraction measurement

Peroxide crosslinking takes place randomly at elevated temperatures in the molten state where the polymer has only an amorphous structure. Figure 2 shows the gel content (%) against DCP concentration (wt %) of the crosslinked LDPE/EVA blends with two different percentages of EVA. As the curve indicates, there is a continuous increase of the gel content with peroxide concentration up to about 2.5%, after which the curve gradually levels off; that is, an exponential variation of gel content was observed with increasing the DCP content. It was also observed that the gel fraction is higher for the sample of higher EVA content

### TABLE II
### Mixing formulations with 20 wt % of EVA

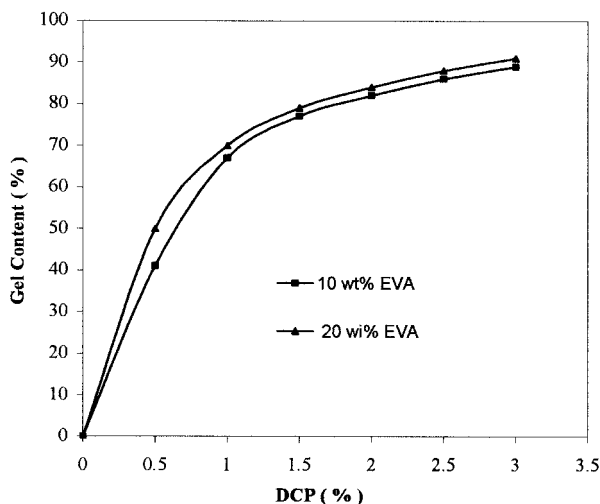| Sample no. | LDPE (phr) | EVA (phr) | Masterbatch (phr) | Irgannox 1010 (phr) | DCP (phr) |
|---|---|---|---|---|---|
| 1 | 100 | 25.05 | 6.62 | 0.176 | 0.7 |
| 2 | 100 | 25 | 6.65 | 0.177 | 1.3 |
| 3 | 100 | 24.94 | 6.68 | 0.178 | 2.1 |
| 4 | 100 | 24.90 | 6.71 | 0.179 | 2.7 |
| 5 | 100 | 24.86 | 6.76 | 0.180 | 3.4 |
| 6 | 100 | 24.81 | 6.79 | 0.181 | 4.1 |

**Figure 2** Plots of the gel content of the cured samples versus concentration of DCP at two levels of EVA content.

for the same DCP concentration (wt %). The increase in gel content with increasing EVA content is attributed to the greater number of tertiary type of carbons, which readily take part in crosslinking reaction.

**Thermal properties**

The amount of crystallinity has a substantial effect on the mechanical and thermal properties of polymeric materials. Table III shows the melting temperature and heat of fusion ($\Delta H_f$), which are related to the amount of crystallinity as determined from DSC measurements, for crosslinked LDPE/EVA blends with different percentages of peroxide content. A decrease in the melting point and $\Delta H_f$ was observed with an increase in DCP content. Therefore, it seemed as if crosslinking reduced the polyolefin crystallinity. The effect is more pronounced with increasing the amount of peroxide.

**Effect of the stretch temperature on shrinkage of the blends**

Figures 3 and 4 show the percentage shrinkage as a function of temperature of LDPE/EVA blends con-

**TABLE III**
**Melting Temperature and $\Delta H_f$ of Crosslinked Low-Density Polyethylene/Poly(ethylene vinyl acetate) Blends**

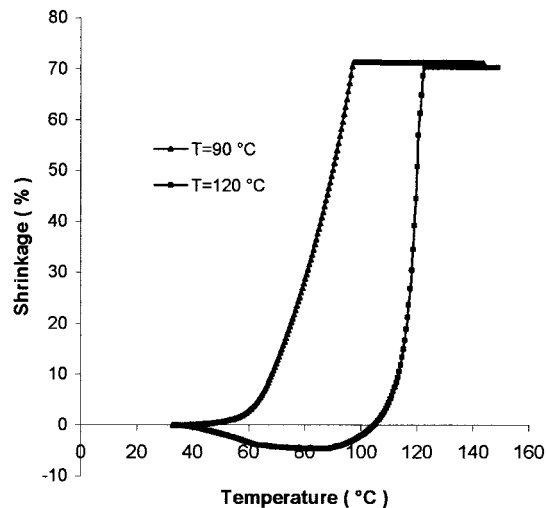| Percent of peroxide (%) | $\Delta H_f$ (mcal/mg) | | Melting temperature (°C) | |
|---|---|---|---|---|
| | 10% EVA | 20% EVA | 10% EVA | 20% EVA |
| 0.5 | 13.24 | 12.63 | 115.8 | 113.7 |
| 1 | 12.52 | 12.21 | 113.8 | 112.7 |
| 1.5 | 12.11 | 11.17 | 112.8 | 112.2 |
| 2 | 11.18 | 10.93 | 111.7 | 111 |
| 2.5 | 10.32 | 10.16 | 110.3 | 110.1 |
| 3 | 9.89 | 9.62 | 109.8 | 109.4 |



**Figure 3** Effect of the stretching temperature on the shrinkability of crosslinked blend of low-density polyethylene with 10% EVA and 0.5% DCP.

taining 10% EVA with 0.5 and 1.5% DCP at two different stretching temperatures (90 and 120°C), respectively. The samples were elongated four times at these temperatures and in the stretched state were rapidly cooled. It may be seen that the sample stretched at 120°C, compared to the sample stretched at 90°C, shows a higher shrinkage temperature when heated. This behavior is the same as for samples with different contents of peroxide and different contents of EVA.

Because measurements of thermal shrinkage involve dimensional changes, it is necessary to discuss the possibility of overlapping of the shrinkage effect and the thermal expansion of the material being investigated. Thermal expansion arises from thermal
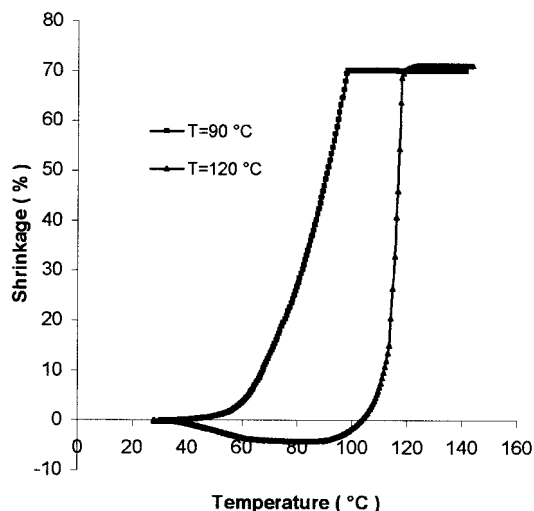


**Figure 4** Effect of the stretching temperature on the shrinkability of crosslinked blend of low-density polyethylene with 10% EVA and 1.5% DCP.

TABLE IV
Thermal Properties of Crosslinked LDPE with 20% EVA as a Function
of Peroxide Content and Stretching Temperature

| Peroxide content (%) | Crosslinked LDPE/EVA blends | | Crosslinked LDPE/EVA blends stretched at 90°C | | Crosslinked LDPE/EVA blends stretched at 120°C | |
|---|---|---|---|---|---|---|
| | $T_m$ | $\Delta H_f$ | $T_m$ | $\Delta H_f$ | $T_m$ | $\Delta H_f$ |
| 0.5 | 113.7 | 12.63 | 116.8 | 13.41 | 114.60 | 12.89 |
| 1.5 | 112.2 | 11.17 | 113.8 | 11.97 | 112.9 | 11.59 |

vibrations of molecules, and this process indeed plays an important role below the temperatures at which molecular orientation is still stable. At higher temperatures, when extensive shrinkage occurs, the influence of thermal expansion of the sample on the overall effect is negligibly small.[24,25]

This effect may be seen in Figures 3 and 4. For the sample stretched and oriented at 120°C (see Fig. 3), shrinkage upon heating does not commence at temperatures below 100°C, and thus a thermal expansion for this sample has been found to be a negative curve in the temperature range 50–110°C before the onset of the main shrinkage curve. However, for the other samples whose onset of shrinkage was at much lower temperatures, thermal expansion has been compensated by the large shrinkage effect and only the sum of the two effects is seen as a curve (with initial zero) and then as a positive slope.

The crosslinked samples, which were heated and stretched at 120°C (a temperature above the melting point), were in an amorphous rubbery state. Cooling the samples, while still being kept in the stretched state, resulted in the formation of large spherulites. In fact, a semicrystalline structure was uniformly formed throughout the sample. According to Hoffman theory, the melting point is related to the thickness of crystal lamella as[26]

$$T_m = T_m^o \left( 1 - \frac{2\sigma_o}{\rho \Delta H_f L} \right) \qquad (3)$$

where $L$ is the thickness of the crystallite, $\rho$ is the crystal phase density, $\sigma_e$ is the fold surface equilibrium free energy, $\Delta H_f$ is the heat of fusion, $T_m^o$ is the melting temperature of the polymer, and $T_m$ is the melting temperature of all crystalline regions. This equation yields the melting point in terms of the fundamental parameters.

Cooling samples from high temperatures (above the melting point) to ambient temperature causes an increase in the thickness of crystal lamella compared to rather small spherulites obtained at lower tempera-

tures and thus resulting in a higher value of the crystal melting temperature ($T_m$).

The crystallite's uniformity, thickness of the crystallite, and total amount of crystalline regions largely affect the thermal properties of the polymer.

The samples, which are stretched at 90°C, below the polymer crystal transition temperature and then cooled, have a different crystalline structure.

The thickness of crystallites formed is smaller and thus according to eq. (3), $T_m$ would be decreased. However, the shrinkage temperature has a close tie with $T_m$ and this would account for the decrease in shrinkage temperature for the sample stretched at 90°C compared to the sample stretched at 120°C. Table IV shows the melting point and the heat of fusion obtained from DSC measurements for the LDPE/EVA blends containing 20% EVA crosslinked with 0.5 and 1.5% DCP and also for the same samples but stretched four times at 90 and 120°C. It is shown that $T_m$ and $\Delta H_f$ for crosslinked LDPE/EVA blends stretched at 90°C are higher than those for samples stretched at 120°C. The crosslinked LDPE/EVA blends stretched at 90°C, near the crystal transition temperature and cooled, not
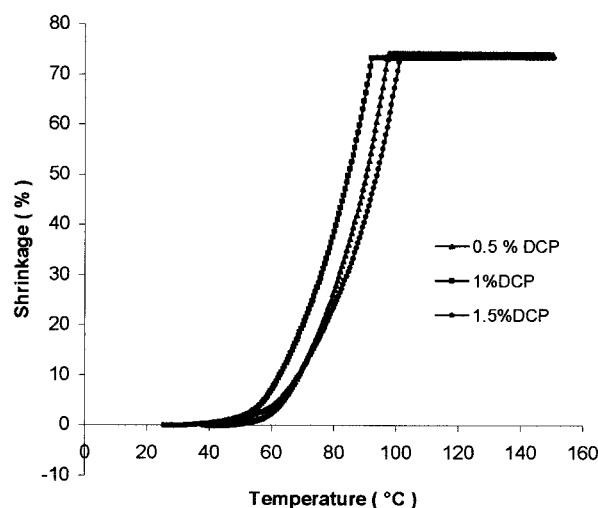


**Figure 5** Effect of DCP content on the temperature-dependent shrinkage of crosslinked blend of low-density polyethylene with 20% EVA stretched at 90°C.
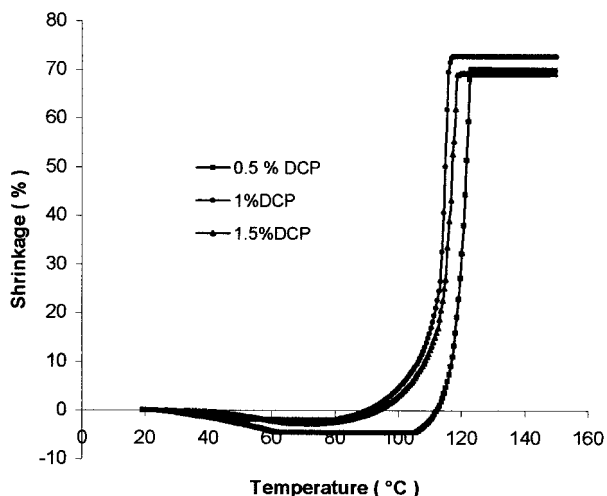
**Figure 6** Effect of DCP content on the temperature-dependent shrinkage of crosslinked blend of low-density polyethylene with 20% EVA stretched at 120°C.
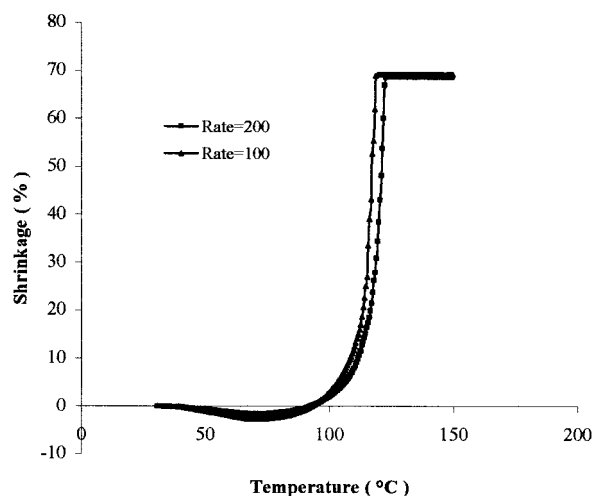


**Figure 8** Effect of rate of stretch on the temperature-dependent shrinkage of crosslinked blend of low-density polyethylene with 10% EVA and 1.5% DCP.

only have smaller spherulites but also a larger number of crystallites because of strain-induced crystallization, which is more pronounced than when the sample cools from the amorphous state (120°C). This fact would account for the increase of average $T_m$ and $\Delta H_f$ for the 90°C sample, although its onset of melting is at lower temperature.

For the same reason the stretched oriented crosslinked LDPE/EVA blends have higher $T_m$ and $\Delta H_f$ values than those of unstretched crosslinked LDPE/EVA blends.

### Effect of DCP content on shrinkage of the blends

Figures 5 and 6 show the effect of DCP content, influencing the crosslinking degree, on shrinkage behavior

of crosslinked blend of low-density polyethylene with 20% EVA stretched at 90 and 120°C, respectively. Both figures show that increasing the crosslinking degree reduces the shrinkage temperature and of course, as noted above, the higher the stretching temperature, the higher the shrinkage temperature. As mentioned before (Table III) because $\Delta H_f$ decreases with increasing DCP content, according to eq. (3), $T_m$ and thus the shrinkage temperature decrease.

### Effect of rate of stretch on shrinkage of the blends

Figures 7 and 8 show the shrinkage percentage versus temperature for samples with different peroxide content with 20% EVA elongated with different rates and
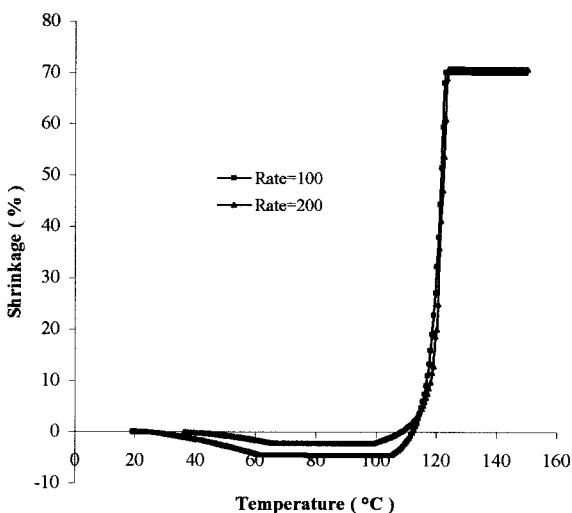


**Figure 7** Effect of rate of stretch on the temperature-dependent shrinkage of crosslinked blend of low-density polyethylene with 10% EVA and 0.5% DCP.
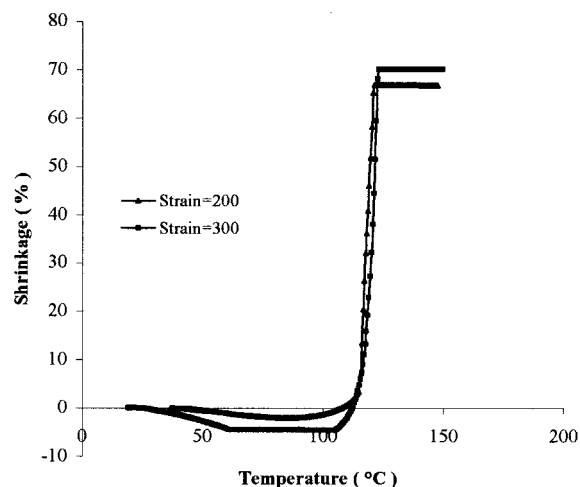


**Figure 9** Effect of percentage of strain on the temperature-dependent shrinkage of crosslinked blend of low-density polyethylene with 10% EVA and 0.5% DCP.
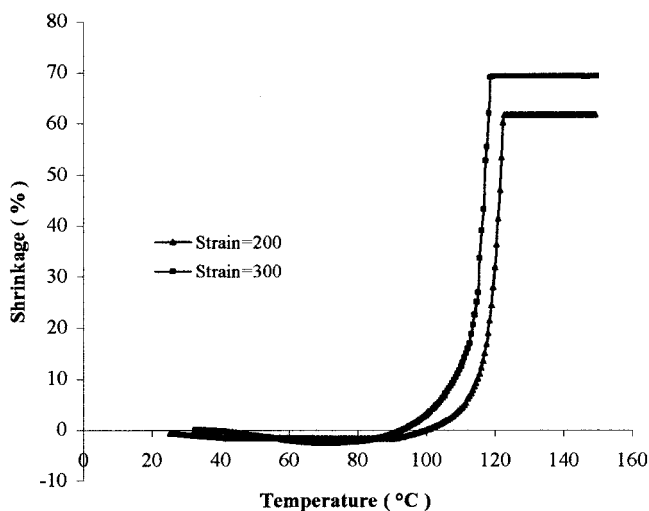
**Figure 10** Effect of percentage of strain on the temperature-dependent shrinkage of crosslinked blend of low-density polyethylene with 10% EVA and 1.5% DCP.

cooled very fast. The result shows that for the samples elongated at 120°C (above the melting point), the rate of stretch has no effect on the shrinkage temperature. Above the melting point, the samples are in the amorphous state. In this state, the stretch of sample results in molecular orientation, on which the rate of stretch has no substantial effect.

### Effect of percentage of strain on shrinkage of the blends

Figures 9 and 10 show the shrinkage percentage versus temperature for samples with different peroxide contents containing 20% EVA elongated by different strains and cooled very fast. The results show that the strain or indeed the stretching ratio has no effect on the temperature of shrinkage, but on the ultimate shrinkage value. For the sample elongated four times, the ultimate shrinkage was 75%. In fact, for this case, $L_{str} = 16$ and $L_{shr,\infty} = 4$; thus the ultimate shrinkage according to eq. (2) was 75%. Also, for the sample elongated three times, the ultimate shrinkage was 66%.

### CONCLUSIONS

The melting temperature and the heat of fusion ($\Delta H_f$) of LDPE/EVA blends are reduced by increasing the dicumyl peroxide (DCP) concentration, which is accompanied by an increase in the gel content because of the higher degree of crosslinking. The results show that the gel fraction is higher for the sample of higher

poly(ethylene vinyl acetate) content for the same DCP concentration (wt %). Moreover, the effect of increasing poly(ethylene vinyl acetate) on the gel content is more pronounced at lower concentrations of DCP. Studies on the heat shrinkability of the samples show that samples stretched above the melting point have a higher shrinkage temperature than those stretched around the crystal transition temperature. Also, by increasing the peroxide content, the shrinkage temperature is decreased. These phenomena may be attributed to the formation of new spherulites as well as changes in the amount and the size of crystals. In fact, the onset of shrinkage and shrinkage temperature of the samples stretched above the melting temperature are higher than those of the samples stretched below the melting temperature. Furthermore the rate of stretching has no effect on the shrinkage temperature at 120°C (above the melting temperature). The percentage of strain has no effect on the temperature of shrinkage but on the ultimate shrinkage value. Generally the influencing parameters investigated on the shrinkability have the same pattern at different peroxide contents.

### References

1. Lyons, B. J. Radiat Phys Chem 1983, 22, 135.
2. Rado, R. Int Polym Sci Technol 1993, 20, 78.
3. Chodak, I. J Prog Polym Sci 1995, 20, 1165.
4. Khonakdar, H. A.; Morshedian, J.; Wagenknecht, U.; Jafari, S. H. Polymer 2003, 44, 4301.
5. Ota, S. Radiat Phys Chem 1981, 18, 81.
6. Lendlein, A.; Kelch, S. Angew Chem Int Ed Engl 2002, 41, 2034.
7. Morton-Jones, D. H.; Ellis, J. W. Polymer Products; Chapman & Hall: London, 1986.
8. Hoffman, J. W. IEEE Elect Insul Magazine 1991, 7, 33.
9. Rado, R.; Zelenak, P. Int Polym Sci Technol 1992, 19, 72.
10. Kampouris, E. M.; Andereopoulos, A. G. J Appl Polym Sci 1987, 34, 1209.
11. Lazar, M.; Rado, R.; Rychly, J. Adv Polym Sci 1990, 95, 149.
12. Brydson, J. A. Plastic Material; Butterworth: London, 1982.
13. Kumar, S.; Pandya, M. V. J Appl Polym Sci 1997, 64, 823.
14. Arsac, A.; Carrot, C. J Appl Polym Sci 1999, 74, 2625.
15. Weil, E. D. Additivity, Synergism in Flame Retardance; Plastic Institute of America: New York, 1971.
16. Chowdury, S. R.; Das, C. K. J Appl Polym Sci 2000, 77, 2088.
17. Patra, P. K.; Das, C. K. J Reinforced Plast Compos 1997, 16, 1111.
18. Patra, P. K.; Das, C. K. J Appl Polym Sci 1998, 68, 597.
19. Khonakdar, H. A.; Morshedian, J.; Mehrabzadeh; Eslami, H. J Adv Polym Technol 2003, 22, 112.
20. Khonakdar, H. A.; Morshedian, J.; Wagenknecht, U.; Jafari, S. H. Eur Polym J, to appear.
21. Patra, P. K.; Das, C. K. J Polym Eng 1997, 17, 231.
22. Mishra, J. K.; Raychowdhury; Das, C. K. Polym Int 2000, 49, 1615.
23. Mishra, J. K.; Das, C. K. J Elastomers Plast 2001, 33, 137.
24. Pakula, T.; Trznadel, M. Polymer 1985, 26, 1011.
25. Trznadel, M.; Ktyszewski, M. Polymer 1988, 29, 418.
26. Gedde, U. W. Polymer Physics; Chapman & Hall: London, 1995.