Achieving Compatibility in Blends of Low-Density Polyethylene/Polyamide-6 with Addition of Ethylene Vinyl Acetate

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ABSTRACT: Polymer alloys and blends, whose major advantage is the potential of achieving a range of physical and mechanical properties, have continued to be a subject of interest over recent years. Addition of a block or graft copolymer, with chemically similar segments to those involved in the polymer blend considered, led to a variety of desirable properties. The copolymer added to the blend functioned to promote a homogeneous dispersion of the constituent phases and to enhance their mutual adhesion. Such agents that enable better dispersion in polymer blends are known as compatibilizers. In this study an attempt has been made to improve the compatibility in a polymer blend composed of two normally incompatible constituents, LDPE and PA_6, by addition of a compatibilizer. The compatibilizer agent, ethylene vinyl acetate (EVA), was added to the polymer blend in ratios of 1, 5, and 10% by using a twin-screw extruder. The effect of EVA on the crystallization of the polymer constituents was observed through DSC examinations. Furthermore, the control sample and all three blends of LDPE/PA_6/EVA were subjected to examinations to obtain their yield and tensile strengths, elasticity modulus, percentage elongation, izod impact strength, hardness, and melt flow index. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 82: 1748–1754, 2001

Key words: polymer alloys; polymer blends; compatibilizer

INTRODUCTION

Polymer blends are composed of either two different polymers or a polymer and a copolymer. Polymer alloys, on the other hand, are formed by adding a third constituent to increase the compatibility to a blend of two polymers that otherwise do not have mutual solubility. Blends of low-density polyethylene (LDPE) and polyamide-6 (PA_6) are such mixtures because the two polymers are not compatible. However, it is possible to impart compatibility by introducing ethylene vinyl acetate (EVA) or poly(ethylene-*co*-vinyl alcohol-*co*-vinyl mercaptoacetate) (EVASH) copolymers into the blend. To improve its physical and mechanical properties, PA_6 may be blended with EVA copolymer. In such a mixing no chemical reaction would take place. Three factors affect the physical and mechanical properties of this blend: the molecular weight, the vinyl acetate content of EVA copolymer, and the process conditions during preparation of the blend.[1–4] Depending on the area of use, polymer alloys and blends may be

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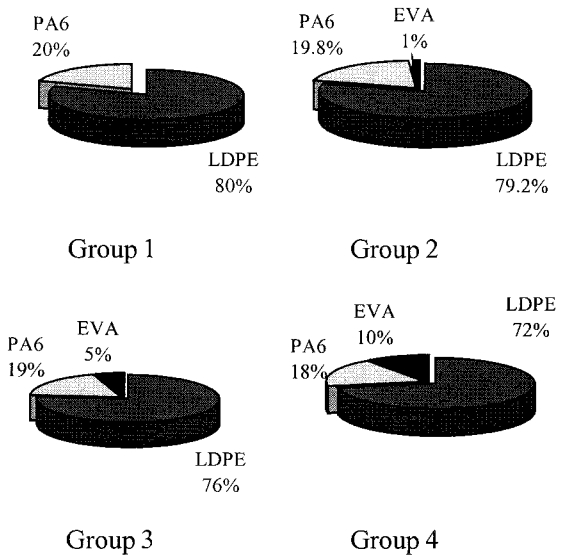


Figure 1 Pie charts showing the nominal compositions of the LDPE/PA_6/EVA alloys studied.

designed in terms of physical properties. Thus correct choice of the constituents becomes the most important issue followed by the choice of a suitable mixing method and a compatibilizer toward good control of the morphology for better performance.[5] Mixing affects the impact, yield, and tensile strengths; hardness; density; resistance to chemicals and wear; and flame-retardation properties. Recent studies also revealed machinability as an important criterion.[6] Mechanical properties of polymer alloys consisting of LDPE/PA_6/EVA may be altered not only by changing the ratios of each individual constituent present but also by employing various processing conditions. Based on this assumption in this study three alloys of LDPE/PA_6/EVA with increasing EVA proportion were prepared and examined in comparison with a control sample of LDPE/PA_6.

EXPERIMENTAL

Materials and Sample Preparation

Four different polymer alloys were prepared by changing the ratio of EVA in the LDPE/PA_6/EVA system by maintaining the ratio of LDPE/PA_6 nominally constant, as shown in Figure 1.

The alloy constituents used in this study were of industrial origin. LDPE had a melt flow index (MFI) value of 2.0 g/10 min (190°C, 2160 g), and melting point of 130°C. PA_6 had an MFI value of 160 cm^3/10 min (275°C, 5000 g), and vicat softening point of 201°C. EVA had a vinyl acetate content of 26–28%, MFI value of 3–4.5 g/10 min (190°C, 2160 g), and a melting point of 75°C. Granular samples of polymer mixtures were prepared by using a double-screw extruder at 170 rpm at a temperature range of 190–220°C under 24 bar pressure. The granular material was then used to prepare tensile test specimens using an injection press. Table I shows the conditions used in this process.

Testing and Characterization of Polymer Alloys

Tensile test samples were prepared in accordance with ISO 294 standard by using an NMC injection-molding machine (NMC Group, Canada). Tensile and impact tests were conducted according to ISO 527.2, at a cross-head speed of 50 mm/min, and ISO 180 standards, respectively, by using Zwick machines (Germany). MFI values were obtained according to ASTM D 1238 using CEAST test equipment (Torino, Italy). DSC studies were undertaken by using SETARAM DSC 131 (Scientex Pty Ltd., Victoria, Australia). SEM examinations were made by using a JSM-T330 JEOL scanning electron microscope (JEOL, Peabody, MA) operated at 15 kV after coating the samples with gold for conductivity.

RESULTS AND DISCUSSION

Mechanical Properties

Figure 2 shows the elasticity modulus, yield and tensile strengths, percentage elongation, impact strength, and hardness values of LDPE/PA_6 alloys containing EVA in various ratios. As can be seen in this figure, 1% EVA addition led to increases in elasticity modulus and tensile strengths, whereas 5 and 10% EVA caused a decrease. On the other hand, EVA addition seemed to decrease the yield strength and hardness. Im-

Table I Process Parameters Used in Injection Press Molding of Tensile Test Specimens

Parameter	Value
Injection temperature	180°C
Injection pressure	40 bar
Dwell time in mold	10 s

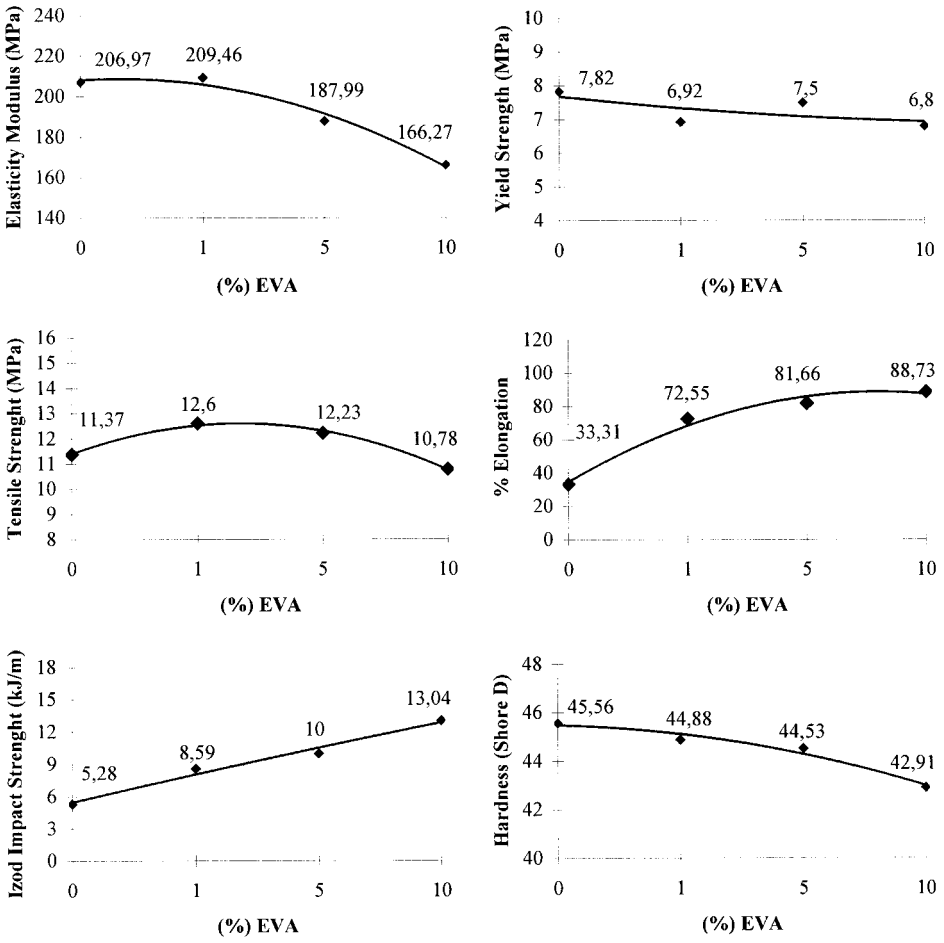


Figure 2 Changes in the mechanical properties of LDPE/PA_6/EVA alloys by addition of EVA.

pact strength and percentage elongation were both observed to increase with the addition of EVA.

Thermal Properties

Melt Flow Index

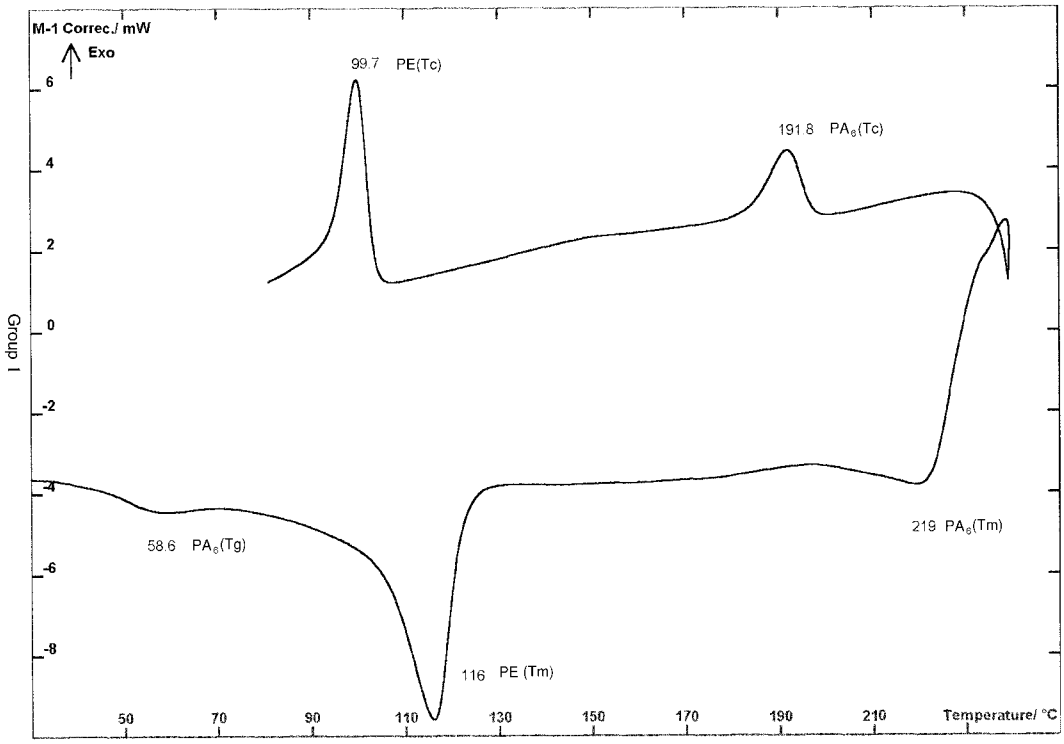
MFI values of the four groups of polymer alloys having various LDPE/PA_6/EVA ratios are given

Table II MFI Values of LDPE/PA_6/EVA Polymer Alloys

Type	MFI (g/10 min) (190°C, 2160 g)
Group 1	7.9
Group 2	7.8
Group 3	7.7
Group 4	7.0

Table III Temperature Data Obtained from DSC for LDPE/PA_6/EVA Polymer Alloys

Group	Parameter (°C)	LDPE	PA_6
1	T_g	—	57.60
	T_m	116.00	219.00
	T_c	99.70	191.80
2	T_g	—	54.40
	T_m	115.50	220.00
	T_c	99.50	192.00
3	T_g	—	53.50
	T_m	116.70	221.00
	T_c	98.70	191.60
4	T_g	—	55.00
	T_m	116.20	219.80
	T_c	98.40	191.00



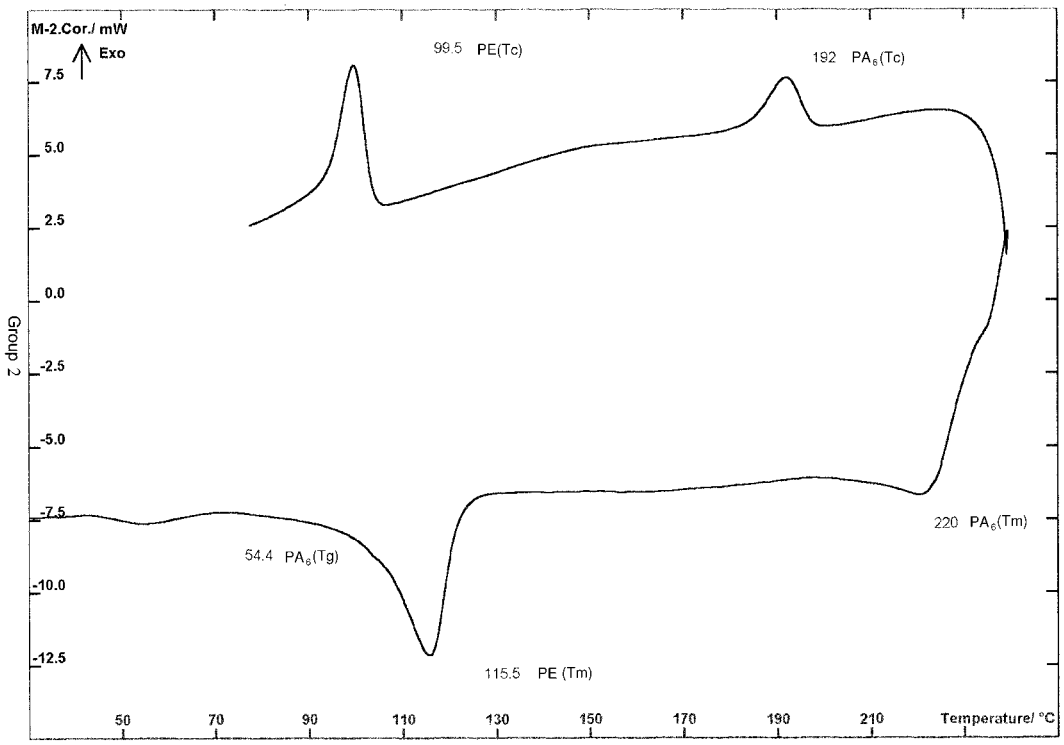


Figure 3 DSC curves for LDPE/PA_6/EVA polymer alloys: (a) Group 1; (b) Group 2; (c) Group 3; (d) Group 4.

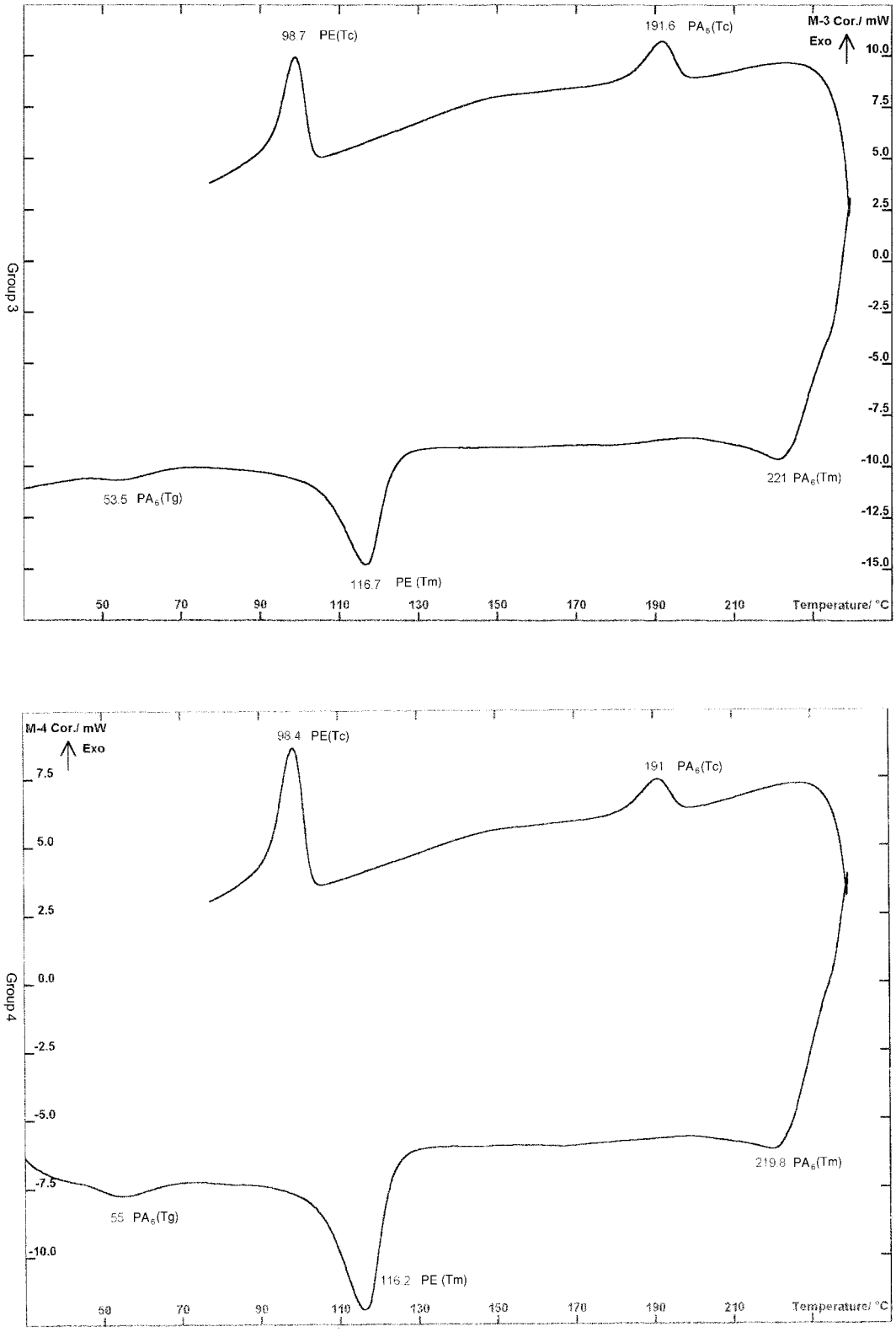


Figure 3 (*Continued from previous page*)

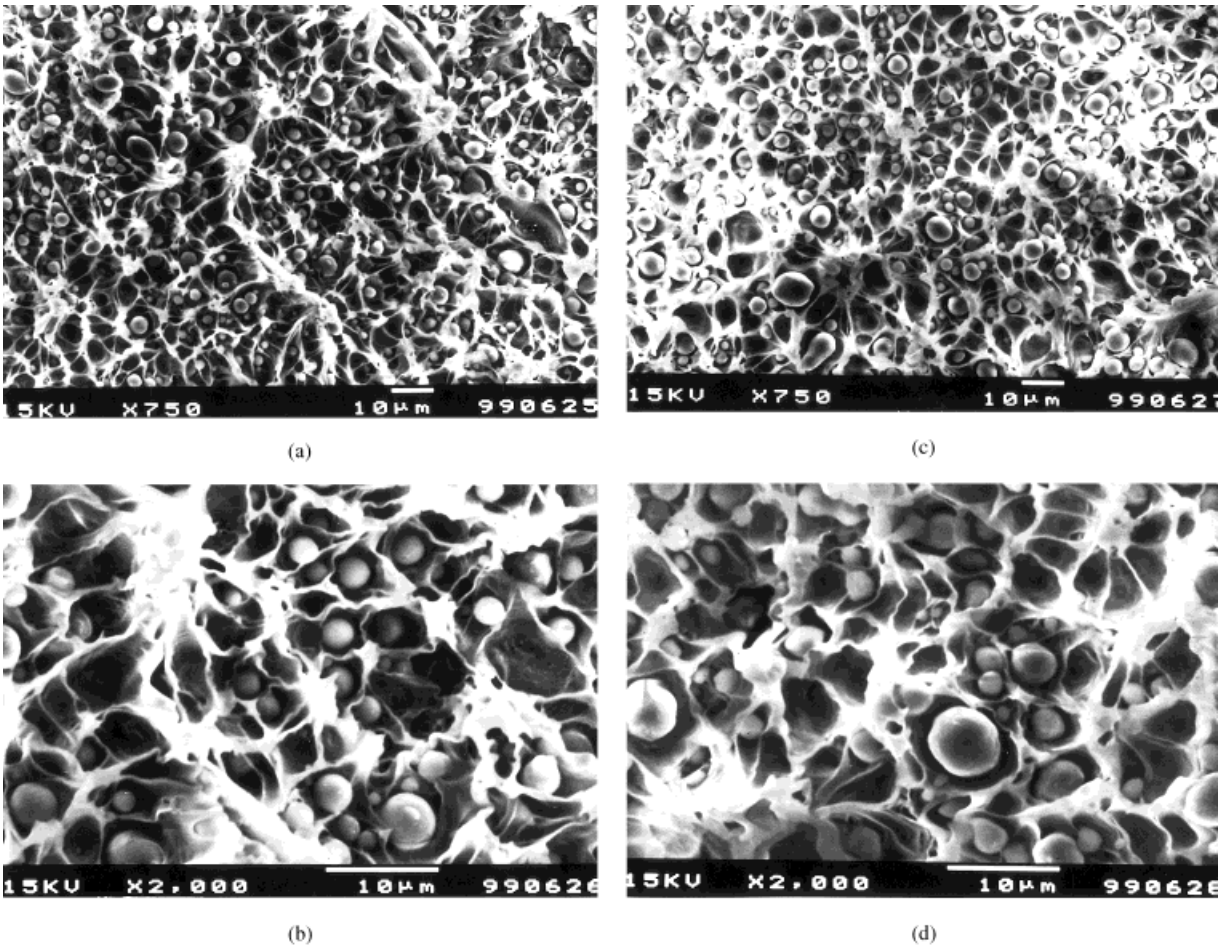


Figure 4 SEM micrographs revealing the appearance of the fracture surfaces of LDPE/PA_6/EVA alloys. (a) LDPE/PA_6, Group 1 polymer blend (×750); (b) LDPE/PA_6/EVA, Group 2 polymer alloy (×2000); (c) LDPE/PA_6/EVA, Group 3 polymer alloy (×750); (d) LDPE/PA_6/EVA, Group 4 polymer alloy (×2000).

in Table II. The MFI values appeared to decrease with increasing EVA ratio. The highest MFI value, 7.9 g/10 min, was achieved with the mixture containing PA_6 of high MFI and LDPE of low MFI, and no EVA.

DSC Observations

During heating the LDPE/PA_6 mixture the glass-transition temperature (T_g) of PA_6 was observed at 58.6°C. The melting temperature (T_m) of LDPE, contrary to the expected value of 130°C, was determined to be 116°C. Upon further heating the melting temperature of PA_6 was observed at 219°C. During controlled cooling the crystallization peaks for PA_6 and LDPE were detected at 191.8 and 99.7°C, respectively. The temperature data obtained from DSC work are given in Table III. The DSC curves are presented in Figure 3.

Morphological Characteristics

The fracture surfaces of the polymer alloys were examined via SEM in an attempt to correlate the mechanical properties to the microstructural characteristics. Figure 4 shows the micrographs taken from the fracture surfaces of the four polymer alloys studied.

CONCLUSIONS

It was shown that increasing addition of EVA copolymer to LDPE/PA_6/EVA mixture enhanced the elastic properties of the alloy. Particularly, the impact strength was observed to have increased considerably—as a desired result—with the increased addition of EVA. The initial increase in the tensile strength at 1% EVA ratio

followed by a gradual decrease with further addition, and especially the increase in percentage elongation ascribed to EVA, may be attributed to increasing compatibility. Such effects may further be interpreted in terms of the elastomeric structure of the compatibilizer agent. In a study on PE/PA_6/EVASH by Silva et al., similar effects on the mechanical behavior were obtained.[3,4,7] As an expected result the MFI values were seen to have dropped with increased EVA addition, given that EVA itself has a low melt flow index. EVA addition, on the other hand, did not cause a significant shift of the heating or cooling curves obtained in DSC experiments. This result is also in agreement with the findings of a reported study on PA_6/PVF/EPDM blends by Tang and Huang[1] SEM examinations of the fracture surfaces indicated the lack of compatibility in LDPE/PA_6 polymer blend, whereas a much better dispersion of the two phases was observed in the alloy containing 5% EVA. Furthermore, SEM micrographs revealed that the adhesion between LDPE and EVA was better developed compared to that between PA_6 and EVA, eventually leading to improved compatibility between LDPE and PA_6 molecules.

REFERENCES

1. Tang, T.; Huang, B. J Appl Polym Sci 1994, 53, 355.
2. Martuscelli, E.; Musto, P.; Ragosta, G. Advanced Routes for Polymer Toughening; Elsevier: The Netherlands, 1996.
3. Barbosa, R. V.; Soares, B. G.; Gomes, A. S. J Appl Polym Sci 1993, 47, 1411.
4. Barbosa, R. V.; Moraes, M. A. R.; Gomes, A. S.; Soares, B. G. Macromol Rep A 1995, 32, 663.
5. Ultracki, L. A. Melt Flow of Polyethylene Blend, Multiphase Polymer: Blends and Ionomers; ACS Symposium Series 395; American Chemical Society: Washington, DC, 1989; pp. 153–210.
6. Ultracki, L. A. Progress in Polymer Processing in Frontiers of Materials Research: Electronic and Materials Research; Hanser: Amsterdam, 1991; pp. 39–48.
7. Silva, E. F.; Bluma, G. S. J Appl Polym Sci 1996, 60, 1687.
8. Frensch, B.; Jungnickel, B. J. Colloid Polym Sci 1989, 267, 16.