

EFFECT OF VINYL ACETATE CONTENT ON THE MECHANICAL PROPERTIES AND MORPHOLOGICAL STRUCTURES OF POLYPROPYLENE

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Abstract The effect of vinyl acetate (VA) content in ethylene vinyl acetate (EVA) copolymer on the mechanical properties of polypropylene was investigated. Three different EVA copolymers with concentrations of 3 wt%, 6 wt%, 9 wt%, 12 wt% and 15 wt%, were blended to polypropylene. The mechanical properties such as yield and tensile strengths, elastic modulus, Izod impact strength, hardness and melt flow index of the blends were investigated. Relationship between type of vinyl acetate and concentrations, mechanical, MFI and morphological properties were explored.

Keywords: Compound; Mechanical properties; Morphology.

INTRODUCTION

Blending of polymers has become an increasingly important area of materials research. The blending or mixing of polymers is the most cost effective ways to upgrade material properties and hence, has the potential to be a reliable and cheap way to recycle them. However, their blends always show rather poor mechanical properties^[1–3]. Compatibility is essential in improving mechanical performance and making recycling favorable. However, the properties of the polymer blends are closely related to their morphology and structures. In recent years, there have been many studies in this field. The composition ratio, viscosity, interfacial tension, and many other factors were adjusted to obtain polymer materials with the desired properties^[3–6].

Polypropylene (PP) is a semi-crystalline and multi-purpose thermoplastic with a wide variety of characteristics such as good mechanical, thermal, corrosion resistance, low cost, easy processing, low density, etc. Properties of polypropylene depend on their compositions, crystalline structure, the ratio of amorphous and crystalline phases, crystal size, molding conditions, chemical composition of the additives, fillers content and its processing technique used. PP is regarded as engineering plastic in industrial applications, however, due to its high transition temperature and in particular, at low service temperatures, notch toughness is not sufficient and thus limits its applications. For this reason, not only the price/performance characteristics but also the low service temperature characteristics can be considerably improved by blending in elastomers and particularly ethylene-propylene copolymer (EPR), ethylene-propylene-diene terpolymer (EPDM), ethylene-vinyl acetate copolymer etc^[7–11]. By blending elastomer to polymeric structures will cause a decrease in the elastic modulus, tensile and yield strengths but an increase in elongation and impact strength properties. Because of this reason, multi-purpose fillers can be used to overcome this problem by ensuring better toughness and dimensional stability. Ethylene vinyl acetate copolymer is the fastest growing general-purpose material on market. This copolymer has wide range of melt index values and good adhesion to a variety of materials, and permits the designing of tailored material. Polypropylene, ethylene vinyl acetate copolymer blends and composites are well suited for many indoor and outdoor applications due to their ozone and weathering resistance, as well as good mechanical properties for

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general applications, such as household goods, electrical insulators, electronic parts, medical supplies, machine parts^[12–16].

In this study, the effect of vinyl acetate content on the mechanical, thermal, and morphological properties of polypropylene/ethylene vinyl acetate copolymer blends were investigated. For this aim, polypropylene/ethylene vinyl acetate copolymer blends containing 0%, 3%, 6%, 9%, 12% and 15% ethylene vinyl acetate copolymer have been prepared and their properties were studied.

EXPERIMENTAL

Materials and Blending Process

PP and EVA copolymer are all commercial products and their characteristic properties are listed in Table 1. PP homopolymer used as matrix phase was Petoplen MH 418, pellet, supplied by Petkim, Turkey. Three kinds of EVA copolymer were used with VA content of 9 wt% (VA 9), 18 wt% (VA 18) and 28 wt% (VA 28). They are supplied by Greenflex (VA 9 and 18) and Evatane respectively. It should be noted that the melt flow index of EVA copolymer increases with increasing of VA content. The additive component, *i.e.* 0.3 wt% Irganox B 225 (CIBA) was added into PP/EVA blends as synergistic processing and long-term thermal stabilizer during the melt processing. PP, EVA copolymer and stabilizer (0.3%, CIBA B 225) were blended by single-screw extruder (Microsan MTV) according to the parameters listed in Table 2. The processing parameters used in extrusion of blends are listed in Table 3. The extrudate was frozen in-line in a water bath ($\sim 20^{\circ}\text{C}$), pelletized and dried in oven at 100°C for 2 h. Tensile and impact specimens (ISO procedures 527 and 180) were prepared by injection molding on a Yonca YMS 100 ton machine. The processing conditions are listed in Table 4.

Table 1. Characteristic properties of the polypropylene and ethylene vinyl acetate copolymer

Materials	Supplier	MFI (g/10 min)	Density (g/cm ³)	T_m ($^{\circ}\text{C}$)	Shape
PP Petoplen MH 418	Petkim	4–6*	0.925	165	pellet
9 EVA (VA 9)	Greenflex	2.5**	0.950	102	pellet
18 EVA (VA 18)	Greenflex	2.5**	0.950	87	pellet
28 EVA (VA 28)	Evatane	4.5**	0.950	75	pellet

* 230°C , 2.16 kg; ** 190°C , 2.16 kg

Table 2. Compositions of the blends (wt%)

Polypropylene (PP)	100	97	94	91	88	75
EVA (9, 18, 28 VA)	0	3	6	9	12	15

Table 3. Processing parameters used in extrusion of PP/EVA blends

Parameter	Value
Screw diameter (D , mm)	30
Length-to-diameter ratio (L/D)	25
Screw speed (r/min)	40
Extrusion pressure (MPa)	0.5
Extrusion temperature profiles from feed zone to the die ($^{\circ}\text{C}$)	210–225

Table 4. Processing parameters in injection molding of PP/EVA blends

Parameters	Value
Injection pressure (MPa)	50
Injection pressure time (s)	10
Mold temperature ($^{\circ}\text{C}$)	40
Dwell time in the mold (s)	10
Temperature range ($^{\circ}\text{C}$)	210–230

CHARACTERIZATION

The effects of EVA copolymer on PP were evaluated through their mechanical properties (yield stress, tensile stress, elastic modulus, Izod impact resistance, hardness (Shore D), and MFI index). The tensile tests of the blends were measured with Zwick Z010 tensile test machine according to ISO procedure 527 at a crosshead speed of 50 mm/min at room temperature. A computer was connected to the Zwick load cell and a data acquisition program read the force recorded by the load cell. At least five specimens were tested for each blend. Impact strengths were measured with a Zwick impact test machine according to the ISO procedure 180 at room temperature with a notch radius of 1 mm. The angle of the notch was 45° and its depth was 2.0 mm. At least ten specimens were tested for each blend. Prior to the mechanical tests, all specimens were kept at room temperature for at least 72 h. The values of the impact strengths were calculated as averages over seven specimens for each composition of blend. The hardness of the blends was measured with Zwick hardness tester according to ISO procedure 868 at room temperature. The values of the hardness tests were calculated as averages over four specimens for each composition of blend. Melt flow indexes (MFI) of PP/EVA copolymer blends were determined by a Zwick 4100 MFI test instrument according to the ISO procedure 1133. The values of the MFI were calculated as averages over four specimens for each composition of blend. The morphology of the blends was observed with the aid of JSM 5410LV JEOL scanning electron microscope (SEM) operated at 20 kV. Before the SEM investigation, the samples were coated with gold to a thickness of 2.5 nm.

RESULTS AND DISCUSSION

The changes in the yield and tensile strengths of the PP/EVA copolymer blends are shown in Figs. 1(a) and 1(b), respectively. As can be seen, the yield and tensile strengths tend to decrease in direct proportion with the increase in VA ratio in the blends. As it is observed that the addition of EVA copolymer has a significant effect on the yield and tensile strengths and the addition of 9%–15% of EVA copolymer causes such a decrease to become more pronounced. As it is shown in Figs. 1(a) and 1(b), 3% EVA copolymer addition cause a slight decrease in yield and tensile strengths, whereas 6%–15% EVA copolymer (in all ratios) cause a significant decrease. Figure 1(c) shows the effect of EVA content on the elastic modulus of blends. Significant decrease in elastic modulus was observed for EVA content = 0%–3%. More than 3% a decrease is observed for VA 9. The reason for the decreasing maybe related to the poor compatibility. When the content of VA = 6%–12% VA 18 and VA 28 blends have similar effect on elastic modulus. The effect of ethylene vinyl acetate copolymer range 15% (VA 18, 28) on the decreased elasticity modulus is much more significant as compared with elasticity modulus of range 3%–12%.

The changes in the Izod impact strength of the PP/EVA copolymer blends containing various ratios of EVA copolymer are shown in Fig. 1(d). In general, impact strength of PP is improved by blending with an elastomer. However, the extent of improvement varies with the nature of the second component of the blends^[17]. It is obvious that blending with EVA copolymer (in various vinyl acetate content) produces greater improvement on the impact strength of PP. EVA copolymer with 9% and 18% VA tends to increase the impact strength of the blends in direct proportion and similar effect on property with the increase in the vinyl acetate ratio in the polypropylene. VA 28 was found to be a better Izod impact modifier than VA 9 and VA 18. Here, if we consider a PP/EVA copolymer blend containing 0% EVA as reference; it becomes apparent that the impact resistance changes significantly depending on the amount of vinyl acetate in a blend containing 15% (VA 28). The increase of the izod impact strength may be the result of the more flexibility of the vinyl acetate. Figure 1(e) shows the hardness of PP/EVA copolymer blends. As it is apparent in the figure, the hardness of blends decreases with the increase of the content of EVA. Figure 1(f) shows the changes in the melt flow indexes (MFI) of PP/EVA copolymer blends. The MFI values increase with the increase in the amount of EVA copolymer. The MFI values increase insignificantly with the addition of VA 9 and VA 18 EVA copolymers, and even a slight decrease is observed in the 3%–12% range. A significant increase of MFI values appears for VA 28.

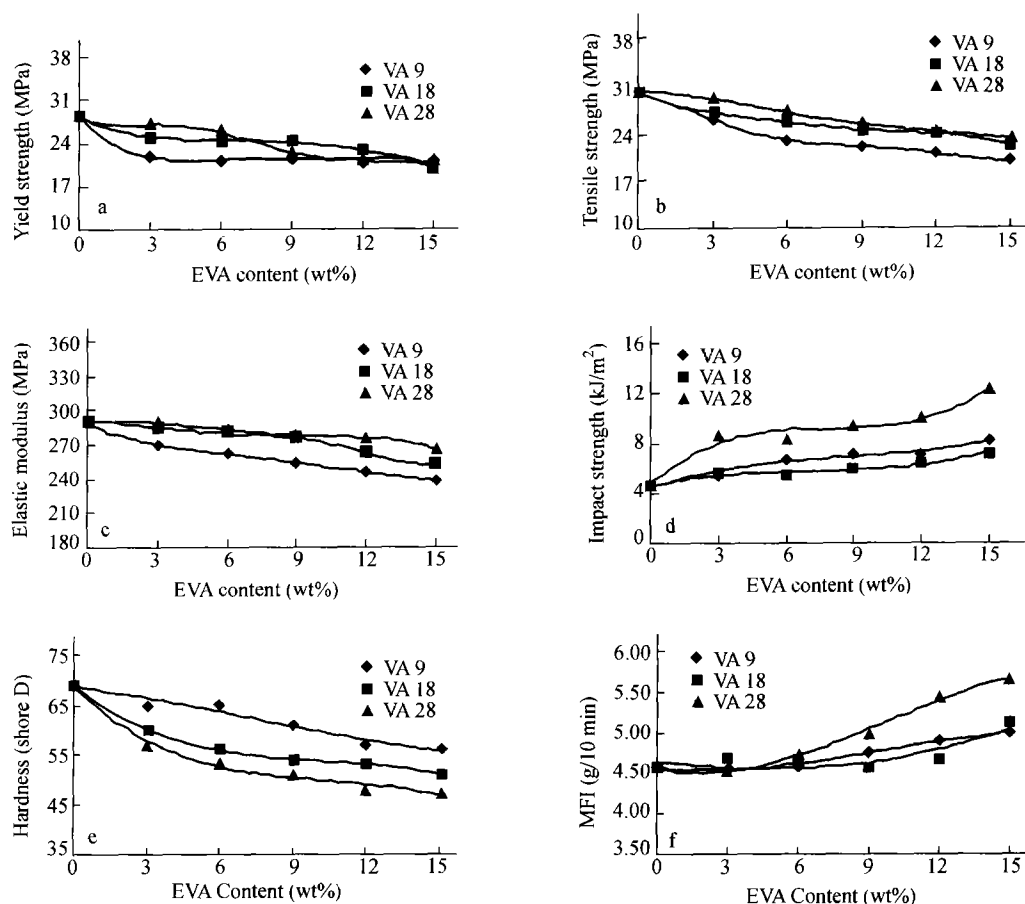


Fig. 1 Properties of polypropylene/ethylene vinyl acetate copolymer blends

The properties of polymer blends are strongly influenced by the morphology of the blending system^[16]. The SEM micrographs of fractured surface of PP/EVA samples with various EVA contents after Izod impact testing are shown in Figs. 2a–2h. In the absence of EVA copolymer, the fractured surface of PP shows an even texture, as shown in Fig. 2(a). The presence of ethylene vinyl acetate copolymer has quite a fine distribution and appears to adhere well in the polypropylene matrix (in Figs. 2b–2h). The adhesion seemed to be enhanced as the ethylene vinyl acetate copolymers were added at 6% and 9%, and the level of homogeneity in the dispersion of phases still appeared to be insufficient (Figs. 2b–2h). When 12%–15% ethylene vinyl acetate copolymer was added, the adhesion and distribution of the phases considerably enhanced polypropylene (Figs. 2c and 2d). Fracture surface topologies of specimens for Izod impact test seem to be dependent on the ethylene vinyl acetate copolymer content. Fracture surfaces are quite smooth with lower ethylene vinyl acetate copolymer content and there is insignificant plastic deformation on the fracture surfaces. Figures 2(e), 2(f) and 2(g) show fracture surfaces of polypropylene that contain ethylene vinyl acetate copolymer VA 18. There is a quite strong adhesion between the matrix material and the ethylene vinyl acetate copolymer. Figure 2(h) shows the fracture surface of the blend containing 15% of ethylene vinyl acetate copolymer VA 28. Furthermore, the increase in the amount of ethylene vinyl acetate copolymer also makes the distribution within the matrix more easy and thus strong adhesion between the matrix and the ethylene vinyl acetate copolymer increases the impact strength. This is due to the fact that ethylene vinyl acetate copolymers are impact modifier in polypropylene/ethylene vinyl acetate copolymer blends in the amorphous region, which is favorable for improving impact properties^[16].

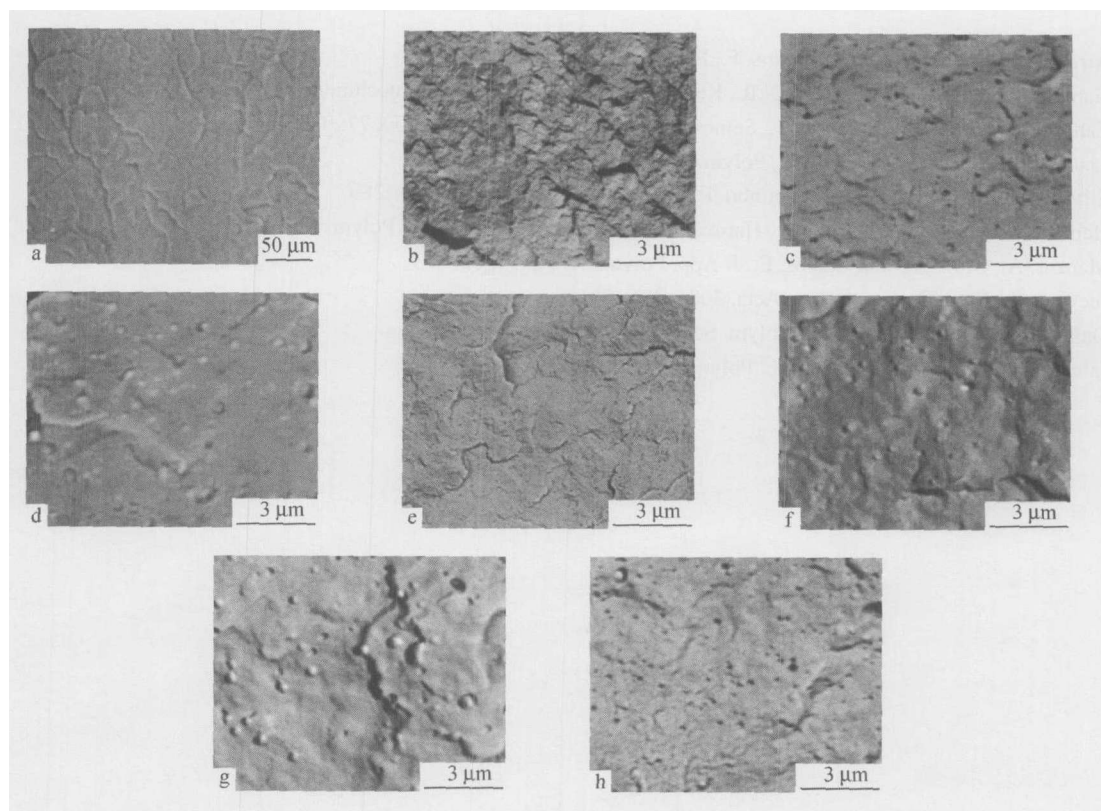


Fig. 2 Scanning electron micrographs (SEM) of the impact fractured surfaces of PP/EVA copolymer blends

a) Polypropylene; b) 6 wt% VA 9; c) 12 wt% VA 9; d) 15 wt% VA 9; e) 6 wt% VA 18
f) 9 wt% VA 18; g) 12 wt% VA 18; h) 15 wt% VA 28

CONCLUSIONS

The effects of ethylene vinyl acetate copolymer on the mechanical properties, thermal and micro structural properties of ethylene vinyl acetate copolymer (9, 18 and 28) toughened polypropylene have been studied. The following conclusions can be drawn from the present investigation. Yield and tensile strength decreased gradually as the ethylene vinyl acetate copolymer content increased. Maximum impact strength was obtained with 12%–15% ethylene vinyl acetate copolymer (VA 28). Hardness values of blends decreased as the ethylene vinyl acetate copolymer content increased. Melt flow index increased as ethylene vinyl acetate copolymer content increased. SEM examinations in this study indicated that ethylene vinyl acetate copolymer especially vinyl acetate 28 type is a good modifier for polypropylene blends in terms of compatibility.

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