Use of a Sodium Ionomer as a Compatibilizer in Polypropylene/High-Barrier Ethylene–Vinyl Alcohol Copolymer Blends: The Processability of the Blends and Their Physical Properties

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ABSTRACT: The effect of a sodium ionomer (ion.Na⁺) on the compatibility of polypropylene (PP)/high-barrier ethylene–vinyl alcohol copolymer (EVOH) blends was studied in terms of the thermal, mechanical, and optical properties and morphology. The rheological behavior, tensile tests, and morphology of the binary blends showed that the miscibility of EVOH with PP was very poor. The miscibility of the polymers improved with the ionomer addition. In general, the ion.Na⁺ concentration did not alter the thermal behavior of the blends, but it did improve the ductility of the injection-molded specimens. Scanning electron micrographs dis-

INTRODUCTION

In recent years, the use of polymers in the packaging industry has steadily increased. A potential new market for polymeric materials is food packaging, a highly competitive area with great demands for performance and cost. Polymers offer advantages over other packaging materials, such as low cost, ease of processing, flexibility, and good physical properties, but many foods require specific atmospheric conditions to maintain their freshness and overall quality during storage, so the packing material needs to have certain gas barriers. This is an important impediment because conventional polymers usually have high permeability to various gases and solvents. An exception is the ethylene-vinyl alcohol copolymer (EVOH). EVOH is an excellent barrier to gases such as O_2 , N_2 , and CO_2 and hydrocarbons, and this property increases with the concentration of vinyl alcohol in the grade. Nevertheless, its barrier properties are greatly affected by

played better adhesion between the PP and EVOH phases in the samples with the ionomer. The mechanical improvement was better in the film samples than in the injection-molded samples. A 90/10 (w/w) PP/EVOH film with 5% ion.Na⁺ and an 80/20 (w/w) PP/EVOH film with 10% ion.Na⁺ presented better global properties than the other blends studied. © 2004 Wiley Periodicals, Inc. J Appl Polym Sci 94: 1763–1770, 2004

Key words: blends; extrusion; ionomers; mechanical properties; poly(propylene) (PP)

moisture because of the hygroscopic nature of vinyl alcohol.¹

The conventional approach to the production of high-barrier films for food packaging is to use multiple layers of different polymers to obtain the properties required.² A laminate that is often used in food packing consists of a layer of EVOH or polyamide 6 (PA6) combined with low-density polyethylene (LDPE), combining the gas barrier properties of EVOH or PA6 with the water vapor barrier, mechanical strength, and excellent sealing properties of LDPE.³ However, recent investigations of this issue have been directed toward polymer alloys, in which the properties of the blend components are synergistically combined so that certain homopolymer drawbacks and the cost/property ratio are minimized.

The blends of EVOH with hydrophobic polymers are examples.⁴ However, EVOH copolymers present bad miscibility and adhesion with other polymers because EVOH copolymers are strongly self-associated but form relatively weak interassociation with other polymers. Thus, it is important to study the compatibility of these blends and to determine how they could be improved to obtain satisfactory mechanical properties and alloys with better barrier properties.

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The purpose of this work was to study the compatibility of polypropylene (PP) and EVOH blends. The importance of PP and EVOH in the packaging industry validates the interest in these blends. To improve their miscibility, we used an ionomer as a compatibilizer.

Ionomers, which have a small number of ionic groups, have attracted interest for many years because of their unique properties as homopolymers and their ability to compatibilize certain incompatible blends.^{5–10} The basic structure is based on a poly(eth-ylene-*co*-methacrylic acid) terpolymer in which the acid groups are partially or fully neutralized by metal ions. In this work, a sodium ionomer (ion.Na⁺) was used.

In this work, we studied PP/EVOH and PP/ EVOH/ion.Na⁺ blends to determine the ionomer effect on the compatibilization of the blends, analyzing their thermal, mechanical, and optical properties and their morphology. The final purpose was to select a blend with better global properties.

EXPERIMENTAL

Materials

Extrusion-grade PP (Isplen PP044W3f) was synthesized by Repsol-YPF. Its melt-flow index (MFI) was 3.02 g/10 min (230° C, 2160 g), and its density was 0.90 g cm^{-3} .

EVOH (F101B), from EVAL Europe (Kuraray Company Ltd., Kuroashiki, Japan), had an ethylene concentration of 32.9%, an MFI of 6.33 g/10 min (230°C, 2160 g), and a density of 1.19 g cm⁻³.

Ion.Na⁺ (Surlyn resin 8528), from DuPont, Wilmington, DE, was a random ethylene/methacrylic acid copolymer with an MFI of 1.10 g/10 min (190°C, 5000 g) and a density of 0.93 g cm⁻³.

Mixing

Before the processing, EVOH and ion.Na⁺ were dried in a vacuum oven for 24 h at 80°C and for 8 h at 60°C, respectively. Blends of PP and EVOH and PP, EVOH, and ion.Na⁺ were prepared with a corotating twinscrew extruder (DSE-20; C.W. Brabender Instruments, South Hackensack, NJ) operating at 45 rpm. The barrel temperature was 215°C, and the die temperature was 220°C. All the components were premixed by tumbling and were simultaneously fed into the twin-screw extruder.

Binary blends were prepared in the following proportions: 90/10, 80/20, 70/30, and 60/40 (w/w) PP/ EVOH. The ternary blends were made through the addition of different concentrations of ion.Na⁺ to the blends (2–20% with respect to EVOH).

The tensile specimens, according to ISO 527, were molded with a Battenfeld Plus 350 injection-molding machine (Battenfeld Extrusionstechnik GmbH, Bad Oeynhandsen, Germany). The temperature of the barrel and die was 200°C.

For the preparation of the extruded films, a Brabender DSE-20 twin-screw extruder with a flat die was used (the barrel and die temperature was 200°C). The melted polymer was cooled on a chilled roll and stretched in the machine direction. The chilled roll was kept at 70°C with a rotation speed of 5.5 rpm.

Apparatus and procedures

The rheological properties of the PP/EVOH blends and EVOH/ion.Na⁺ were obtained from the torque during kneading in a Brabender plasticorder at 200°C.

The MFIs of the samples were measured in a meltflow tester (Ceast S.p.A., Torino, Italy) according to ISO 1113. The temperature of the test was 230°C, and it was used at a load of 2160 g.

Differential scanning calorimetry (DSC) measurements were carried out with a DSC 7 (Perkin Elmer Cetus Instruments, Norwalk, CT). The influence of the EVOH and ion.Na⁺ concentrations on the melting and crystallization of the blends was studied. The following thermal cycle was applied to the samples: a first heating scan from room temperature to 210°C at 10°C min⁻¹, followed by an isothermal scan at 210°C for 5 min to erase the thermal history and a subsequent cooling scan to room temperature at 10°C min⁻¹. The melting thermograms of the samples were recorded from the second heating scan at 10°C min⁻¹.

Tensile tests of the injection-molded specimens were performed at a crosshead speed of 10 mm min⁻¹ and at 23°C with an Instron 5566 mechanical tester (Instron, Canton, MA) according to ISO 527. The mechanical properties of the films were measured at room temperature according to DIN 53457 with a Zwick 1425 universal testing machine (Zwick-GmbH, Germany) and rectangular strips measuring 150 mm long and 15 mm wide. The crosshead speed was 100 mm min⁻¹. For both tensile tests, eight specimens were tested for each reported value, and the results were averaged to obtain a mean value.

The haze of the films was measured in with a Diffusion Systems SH MOD.57 (Diffusion Systems, U.K.) according to ASTM D 1003.

Scanning electron microscopy (SEM; JSM-6400, JEOL, USA Inc., Peabody, MA) was used to study the morphologies of the fractured surfaces of the blends at an accelerating voltage of 20 kV. The samples were sputter-coated with a thin layer of gold. Cryofractured surfaces, obtained with liquid nitrogen, were examined at a tilt angle of 30°.



Figure 1 Logarithm of the torque of the PP/EVOH blends versus the EVOH content.

RESULTS AND DISCUSSION

Rheological behavior

Figures 1 and 2 show the logarithm of the torque of the PP/EVOH blends and EVOH/ionomer blends as a function of their composition. The reported torque values correspond to the steady state obtained after the full mixing, and they are related to the melt viscosity of the blends, although the relation is complex. Some authors have suggested that the log additivity



Figure 2 Logarithm of the torque of the EVOH/ion.Na⁺ blends versus the ionomer content.

TABLE I MFIs PP/EVOH Blends and PP/EVOH/Ion.Na⁺ Blends

PP/EVOH/Ion.Na ⁺	MFI (g/10 min)
100/0/0	2.59 (0.07)
90/10/0	4.04 (0.57)
90/10/2	2.85 (0.05)
90/10/5	2.83 (0.05)
90/10/10	2.64 (0.01)
80/20/0	4.92 (0.08)
80/20/2	3.17 (0.15)
80/20/5	2.86 (0.01)
80/20/10	2.73 (0.05)
70/30/0	6.26 (0.16)
70/30/5	3.23 (0.01)
70/30/10	2.90 (0.02)
70/30/15	2.79 (0.02)
70/30/20	2.83 (0.02)
60/40/0	8.53 (0.26)
60/40/5	3.16 (0.07)
60/40/10	2.96 (0.22)
60/40/15	3.14 (0.03)
60/40/20	3.00 (0.06)
0/100/0	5.29 (0.29)

rule might be used to classify the flow behavior of polymer blends. As shown in Figure 1, the viscosity of pure PP was reduced with the addition of EVOH, and all the PP/EVOH blends displayed a negative deviation with respect to the log additivity rule, which was a signal of poor compatibility between the blend components. A reduction in the viscosity is typical for immiscible blends.^{11–13}

In the EVOH/ionomer blends (Fig. 2), a positive deviation with respect to the log additivity rule was observed. This proved that the components were miscible and that the ionomer could improve the miscibility of PP/EVOH blends.

The MFI data of the binary and ternary blends are summarized in Table I. For the binary blends, the MFI values increased with the EVOH concentration, with a positive deviation of the linear relationship between the values of the pure components. This fact is directly related to the diminution of the melt viscosity of the PP/EVOH blends previously studied.

With the addition of the ionomer, the MFI values decreased in all the blends studied, and the melt-flow behavior of the ternary blends was similar to that of pure PP. Theoretically, their processing parameters would be very close to the PP parameters.

Thermal properties

Figure 3 shows the exotherms and endotherms of pure EVOH, PP, and ion.Na⁺, which correspond to the cooling scans and second heating scans, respectively. From the heating scan of EVOH, the melting peak (at 181.2°C) and T_g (at 55.4°C) were measured. The endotherm of ion.Na⁺ displays a melting peak with a max-



Figure 3 Crystallization curves (continuous) and melting curves (dotted) of PP, EVOH, and ion. Na^+ .

imum at 98.1°C and a weak shoulder at 64.0°C, which can be associated with cluster dissociation during the heating scan; it was clearly detected during the first heating scan. The PP thermogram shows a melting point at 159.3°C.

The cooling scans plotted in Figure 3 show the crystallization peaks at 60.8°C for ion.Na⁺ and 109.3°C for PP. In the EVOH curve, two exotherms can be observed at 146.2 and 106.1°C (very weak); the latter crystallization peak could be due to molecular heterogeneity of the copolymer and was previously detected by other authors.¹⁴

The temperatures of the melting peaks (T_m) and the maximum crystallization temperature (T_c) for the PP/ EVOH blends are summarized in Table II. Figure 4 shows the endotherms of 60/40 (w/w) PP/EVOH with different concentrations of the compatibilizer. The curves correspond to the second heating scan of the samples. In all the blends, two melting peaks were observed, corresponding to the PP phase and EVOH phase. There was not a significant change in T_m with the EVOH concentration. These results indicated the presence in the blends of two crystalline phases and the absence of strong interactions between the blend components, which should have affected the crystalline perfection.

For the blends with high concentrations of ion.Na⁺, a small peak could be observed at 96°C (see Fig. 4), which corresponded to the melting endotherm of ion.Na⁺. This fact proved that the mixing did not alter the melting behavior of the ionomer.

Some authors have found that T_m of EVOH decreases with high ionomer compositions because of

		TABLE II
Thermal	Properties	of PP/EVOH Blends and PP/EVOH/
	-	Ion.Na ⁺ Blends

PP/EVOH/ion.Na ⁺	T_m	(°C)	<i>T_c</i> (°C)
90/10/0	160.4	180.6	126.7	162.1
90/10/2	161.7	179.9	117.5	160.5
90/10/5	160.7	180.1	118.0	160.2
90/10/10	162.7	180.3	111.6	158.5
80/20/0	160.9	181.1	125.1	162.1
80/20/2	161.7	181.7	119.4	160.9
80/20/5	161.7	181.6	116.6	159.7
80/20/10	161.7	181.0	117.3	160.1
70/30/0	160.5	180.9	122.7	159.1
70/30/5	159.8	180.4	112.1	160.4
70/30/10	162.1	181.3	110.7	160.7
70/30/15	159.6	179.2	110.5	160.0
70/30/20	159.7	179.9	110.4	160.4
60/40/0	161.7	184.6	123.7	160.4
60/40/5	160.1	181.3	111.1	160.4
60/40/10	162.3	182.8	110.4	160.4
60/40/15	161.4	181.6	110.4	160.4
60/40/20	157.5	182.2	109.5	160.0

morphological or thermodynamic miscibility, but in this study, the presence of the ionomer did not seem to alter the thermal behavior of the copolymer. However, a decrease in T_c corresponding to the PP phase with the concentration of ion.Na⁺ in the ternary blends could be observed in the experiments. This reduction indicated that interactions between the components existed and that the presence of the ionomer hindered the PP crystallization.



Figure 4 DSC thermograms of 60/40 (w/w) PP/EVOH with different concentrations of ion.Na⁺.

Compatibilizer and	ompatibilizer and with Different Amounts of Ion.Na ⁺		
PP/EVOH/ion.Na ⁺	E (MPa)	σ_{B} (MPa)	$arepsilon_B$ (%)
100/0/0	854 (12)	25.0 (0.4)	575 (1)
90/10/0	1025 (25)	10.4 (0.4)	33.0 (14.3)
90/10/2	880 (36)	18.6 (0.3)	165.1 (27.8)
90/10/5	896 (48)	25.0 (0.1)	577.8 (0.7)
90/10/10	883 (13)	22.6 (3.2)	566.9 (19.1)
80/20/0	1102 (16)	25,.2 (0.2)	11.8 (1.1)
80/20/2	958 (22)	17.9 (0.2)	30.7 (7.8)
80/20/5	933 (34)	18.2 (0.4)	41.3 (20.5)
80/20/10	959 (17)	18.4 (0.3)	119.7 (25.5)
70/30/0	1044 (26)	27.4 (0.3)	8.4 (0.4)
70/30/5	1026 (21)	30.7 (0.6)	12.6 (1.3)
70/30/10	945 (18)	29.8 (0.5)	14.6 (1.6)
70/30/15	1003 (34)	29.4 (0.3)	11.5 (1.1)
70/30/20	904 (42)	29.4 (0.1)	14.7 (1.2)
60/40/0	1236 (23)	27.6 (0.3)	7.4 (0.6)
60/40/5	1150 (21)	32.3 (0.5)	11.2 (0.9)
60/40/10	1125 (24)	32.4 (0.8)	11.9 (2.1)
60/40/15	1067 (41)	32.4 (0.2)	13.7 (1.8)
60/40/20	1050 (36)	29.2 (0.6)	8.4 (0.5)
0/100/0	2009 (248)	39.7 (2.9)	28 (2)

 TABLE III

 Tensile Properties of PP/EVOH Blends Without a

 Compatibilizer and with Different Amounts of Ion.Na⁺

Tensile properties

Binary blends

The tensile properties [Young's modulus (*E*), strength at break (σ_B), and strain at break (ε_B) obtained from the load–elongation curves] for the injection-molded specimens are summarized in Table III.

The EVOH incorporation increased the material modulus at the expense of a severe decrease in the ductility of the blends. The modulus increased with the EVOH amount after the ideal behavior of the blends, with the exception of the 70/30 composition. This behavior was fairly usual in immiscible blends. The principal effect of the EVOH incorporation on the mechanical behavior of the blends was a strong reduction of ε_{B} . It could be explained by the presence of large EVOH particles in the PP matrix, and it showed the incompatibility of both polymers due to the poor interfacial adhesion between the dispersed phase (EVOH) and the matrix of PP. Although the 90/10(w/w) PP/EVOH blend showed a relatively high $\varepsilon_{\rm B}$ value, the other blends with higher EVOH contents displayed brittle fracture. The tensile strength slightly increased with respect to the PP value in the samples with higher EVOH concentrations because of the strong decrease in ε_B . Because of the low ε_B values of these compositions, the tensile strength increase was probably due to the high value of EVOH in comparison with that of PP.

Ternary blends

To improve the mechanical properties and compatibility of the blends, we added ion.Na⁺ to the formulations. For each blend, the effect of the ionomer quantity on the mechanical behavior was studied with injection-molded specimens. Table III shows the data obtained. The modulus increment observed in the binary blends with the EVOH concentration was reduced in the ternary blends. This was due to the low modulus of the ionomer. In general, the modulus of the materials improved with respect to pure PP.

The tensile strength increased with respect to the values of the binary blends (except in the 80/20 blends), although the trend with the quantity of the ionomer in the blends was different. The ductility of the 90/10 (w/w) PP/EVOH blends improved with the ionomer addition; this allowed important growth of σ_B in the blends with 5 and 10% ion.Na⁺ with respect to the corresponding binary blend. For the 70/30 and 60/40 (w/w) PP/EVOH blends, the ionomer allowed an enhancement in the σ_{B} values; it was probably due to the slight improvement in ε_B produced by the compatibilizer and to the higher modulus of these samples with respect to the others. However, these blends exhibited brittle fracture. In the 80/20 (w/w) PP/ EVOH blends, the mechanical behavior changed with the ionomer in the blends. The brittle fracture in the binary blend was converted into more ductile fracture, although the mechanical tests showed great dispersion (which could be seen in the high deviations for $\varepsilon_{\rm B}$). This fact could explain the reduction in the tensile strength when the ionomer was added to the blend.

The addition of ion.Na⁺ to the PP/EVOH blends raised ε_B . It was evident that the ionic aggregates acted like compatibilizers in the polymer blends, but the results were not similar in all the ternary blends.

For the 90/10 (w/w) PP/EVOH blends, 2% ion.Na⁺ was enough to obtain a good value for ε_B , but 5% produced a better value close to the value obtained for pure PP.

For the 80/20 (w/w) PP/EVOH blends, the ionomer addition enhanced ε_B , but the improvement even with 10% ionomer was not enough to achieve high values (near that of PP). The high deviations (in the blends with 5 and 10% ionomer) showed that the ionomer incorporation was not as effective as in the previous blend, even though the mechanical behavior corresponded to a ductile material.

For the 70/10 and 60/40 (w/w) PP/EVOH blends, ionomer concentrations up to 20% were studied. Although an increase in ε_B was measurable, the samples exhibited a fragile fracture, with ε_B near that of the blends without the ionomer. The added ionomer was not enough to obtain an effective compatibilization of the PP/EVOH blends.

Films

The mechanical properties of the extruded films are summarized in Table IV. The films were analyzed in

	a Companioniz		
PP/EVOH/ion.Na ⁺	E (MPa)	σ_{B} (MPa)	$arepsilon_B$ (%)
100/0/0	1122 (100)	38.3 (4)	538 (22)
90/10/0	1266 (124)	27.8 (4)	236 (53)
90/10/2	1392 (99)	44.8 (6.1)	425 (90)
90/10/5	1197 (145)	38.2 (8.2)	471 (86)
90/10/10	1111 (94)	40.0 (3.0)	454 (18)
80/20/0	1127 (87)	25.3 (4)	31 (5)
80/20/2	1268 (60)	30.6 (1.9)	8,8(1)
80/20/5	1261 (149)	38.6 (2.9)	471 (44)
80/20/10	1388 (178)	50.3 (1.3)	439 (9)
70/30/0	1391 (127)	31.1 (3.4)	5(1)
70/30/15	1741 (90)	46.8 (3.0)	449 (10)
70/30/20	1506 (90)	33.8 (2.5)	20 (18)
60/40/0	1405 (129)	34.7 (1.1)	7 (2)
60/40/15	1561 (74)	38.7 (3.1)	349 (38)
60/40/20	1822 (152)	45.6 (6.3)	352 (44)

TABLE IVTensile Properties of PP/EVOH Films with and Without
a Compatibilizer

the stretching direction. In the binary blends, E increased with the EVOH concentration. Although the moduli of the films were higher than the moduli of the injection-molded specimens, these differences were probably due to the different geometries of the samples and the higher crosshead speed used in these mechanical tests. With the ionomer addition, the modulus of the films showed slight variations in the 90/10and 80/20 (w/w) PP/EVOH blends. In the other ternary blends, the modulus increased with respect to the binary blends. This shows that the compatibilizer promoted the modulus increment because of the copolymer presence, and only the 70/30 (w/w) PP/EVOH blend with 20% ionomer showed a reduction caused by the low value of the ionomer modulus and the high concentration of the ionomer.

The mechanical behavior of the films at the break point was better than the behavior of the injectionmolded samples. The tensile strength of the PP/ EVOH films with ion.Na⁺ increased with respect to the injection-molded samples, but the values did not show a clear trend. In the films of the binary blends, ε_{B} was higher than in the injection-molded specimens for the 90/10 and 80/20 blends, but similar values were obtained in the 70/30 and 60/40 blends. With the ionomer addition, the strain values were acceptable in the 90/10 blends. Except in the 60/40 (w/w) PP/ EVOH blends, high concentrations of the ionomer produced a reduction in the ductility, probably because of ionomer agglomeration.¹⁵ The more important improvement with respect to the injectionmolded specimens was achieved in the 70/30 (w/w) PP/EVOH blend with 15% ionomer and in the 60/40 (w/w) PP/EVOH blend with 20% ionomer; the compatibilization could reach high strain values at the break point. However, these blends were not very interesting because they were not mechanically compatible without high ionomer concentrations, and it was demonstrated that the high permeability of the ionomer reduced the beneficial barrier properties of EVOH. In general, the molecular orientation suffered by the materials during the stretching improved the tensile properties of the ternary blends.

Optical properties

An estimation of the blend miscibility was obtained from haze measurements.¹³ Haze is the percentage of transmitted light through a specimen that is deviated from the incident beam by forward scattering, and haze data can be related to the heterogeneity of the surface and its internal defects, which contribute to the diffusion or light deviation. The haze values corresponding to the blends with better mechanical properties are summarized in Table V. In the binary blends, the haze values increased with the EVOH concentration, showing that the immiscibility of the polymers produced no homogeneous materials. When ion.Na⁺ was added to the formulations, the haze of the films diminished. This indicated that the blends with high ionomer concentrations were more homogeneous than the materials with lower ionomer contents.

Morphology

Figure 5 shows the fracture surfaces of injectionmolded specimens for the binary blends. The micrographs show that the blends were biphasic, and EVOH was dispersed across the PP matrix. The dispersed EVOH phase was generally elongated and oriented, probably because of the shear flow during the injection-mold filling. As expected, the size of the dispersed structures increased with the EVOH concentration in the blends.

The dispersed structures underwent debonding on fracture; there were no rest of particles upon the fracture surface. These facts indicated poor interfacial ad-

TABLE V Haze of PP/EVOH/Ion.Na⁺ Films

PP/EVOH/ion.Na ⁺	Haze
100/0/0	35.0 (1.0)
90/10/0	59.9 (3.9)
90/10/2	57.2 (8.5)
90/10/5	51.3 (4.8)
90/10/10	49.5 (2.3)
80/20/0	62.8 (0.8)
80/20/5	51.7 (1.0)
70/30/0	66.6 (1.4)
70/30/15	50.5 (1.2)
60/40/0	69.7 (0.5)
60/40/20	51.8 (2.1)



Figure 5 SEM micrographs (scale = $30 \ \mu$ m) of binary PP/ EVOH blends: (a) 90/10, (b) 80/20, (c) 70/30, and (d) 60/40.

hesion between PP and EVOH; consequently, the incompatibility of the blends was demonstrated, in good agreement with the previously discussed mechanical and optical properties.

Figures 6 and 7 show the morphologies of the 90/10 and 60/40 (w/w) PP/EVOH blends with different concentrations of ion.Na⁺; in this way, the effect of the compatibilizer on the morphology of the blends was studied. The addition of the ionomer had a clear effect on the morphology. Figure 6(a–c) shows that low quantities of the ionomer drastically reduced the dimensions of the EVOH phase and improved its dispersion into the PP matrix. The further addition of the compatibilizer did not alter these features. For the 60/40 (w/w) PP/EVOH blends, at least 15% ion.Na⁺ was necessary to observe the effect of the compatibilization on the morphologies.



Figure 6 SEM micrographs (scale = $20 \ \mu$ m) of $90/10 \ (w/w)$ PP/EVOH with different ion.Na⁺ concentrations: (a) 0, (b) 2, (c) 5, and (d) 10%.



Figure 7 SEM micrographs (scale = 20 μ m) of 60/40 (w/w) PP/EVOH with different ion.Na⁺ concentrations: (a) 0, (b) 10, (c) 15, and (d) 20%.

In summary, the distribution of the dispersed phase became more homogeneous in the compatibilized blends. The size of the dispersed particles decreased upon compatibilization, and the fracture was progressively more cohesive. The EVOH particles appeared firmly bonded to the PP matrix, and this indicated an increase in the interfacial adhesion as a result of the compatibilizing effect of the ionomer.

CONCLUSIONS

The compatibilization of the PP/EVOH blends with an ionomer of sodium was studied. The following conclusions were drawn:

- 1. The rheological behavior was studied, and the results showed poor compatibility between the PP/EVOH blends. However, with the addition of the ionomer, the blend miscibility was clearly improved.
- 2. The investigated ionomer concentrations did not modify the melting behavior of the PP/ EVOH blends. However, a reduction of T_c of the PP phase with an increase in ion.Na⁺ in the ternary blends was observed. This fact was characteristic of strong interactions between the components.
- 3. The tensile properties of the binary blends showed that PP and EVOH were incompatible: the modulus increased and ε_B strongly decreased with the concentration of the copolymer in the blends. The morphology, studied by SEM, proved the incompatibility of both polymers due to the poor interfacial adhesion between the dispersed phase (EVOH) and the matrix of PP.

- 4. The ionomer in the ternary blends improved the mechanical behavior in the injection-molded blends. The SEM micrographs showed that ion.Na⁺ improved the adhesion between EVOH and PP and that the distribution of the EVOH phase in the matrix was more regular in the blends with the compatibilizer. The best results were obtained in the 90/10 blends with ionomer concentrations of 5 or 10%.
- 5. The mechanical behavior of the films was better than that of the injection-molded samples. The more important improvement with respect to the injection-molded specimens was achieved in the 70/30 PP/EVOH blend with 15% ionomer and in the 60/40 (w/w) PP/EVOH blend with 20% ionomer, for which high values of ε_B were measured. However, the high concentration of the ionomer in these films was undesirable for the improvement of the barrier properties.
- 6. In summary, the 90/10 (w/w) PP/EVOH film with 5% ion.Na⁺ and the 80/20 (w/w) PP/EVOH film with 10% ionomer were the best of the films studied. Moreover, the concentrations

of the copolymer and ionomer were not excessive for possible industrial applications.

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