Preparation and Characterization of a Methyl Methacrylate/Ethyl Acrylate/Acrylic Acid Terpolymer To Improve the Homogeneity of Ethylene–Propylene–Diene Monomer Rubber/Poly(vinyl chloride) Blends

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ABSTRACT: The emulsion terpolymerization of methyl methacrylate (MMA), ethyl acrylate (EA), and acrylic acid (AA) was carried out under a nitrogen atmosphere at 70°C. The final terpolymer conversion was determined gravimetrically. The synthesized MMA-EA-AA terpolymer was characterized with ¹H-NMR spectroscopy, thermal analysis, and gel permeation chromatography. Glasstransition temperatures of the MMA-EA-AA terpolymer were determined with a differential scanning calorimeter. Ethylene-propylene-diene monomer rubber (EPDM)/poly(vinyl chloride) (PVC) blends were prepared with different blend ratios (10/90, 20/80, 30/70, 40/60, and 50/50) in the presence and absence of MMA-EA-AA as a compatibilizer. The morphology of those blends was examined with the aid of a scanning electron microscope. The scanning electron micrographs in the presence of the MMA-EA-AA terpolymer illustrated the disappearance of the macroscale phase separation of EPDM/PVC blends as a result of the incorporation of MMA-EA-AA into that blend, indicating an improvement of the homogeneity. The mechanical properties of the EPDM/PVC blend films and the dielectric properties of the melt blends were investigated. The swelling behavior of the cured blends in the brake fluid was also discussed. The results illustrated that the mechanical properties, the weight swelling values, and the dielectric constant values showed linear behavior versus the blend ratios after the incorporation of the terpolymer. However, those values showed deviations from linearity in the absence of the terpolymer. That, in turn, ensured the results obtained with the scanning electron microscope. The results reveal that the MMA-EA-AA terpolymer prepared can be used successfully to improve the homogeneity of EPDM/PVC blends used in hose and oil seal applications. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 109: 1738–1745, 2008

Key words: blends; compatibility; compounding; copolymerization; elastomers

INTRODUCTION

The blending of immiscible polymers offers attractive opportunities for developing new materials with useful combinations of properties. However, simple blends often have poor mechanical properties and unstable morphologies. Compatibilization of such blends is necessary. Preformed graft or block copolymers have been traditionally added to act as compatibilizers.^{1–6} Some copolymers that carry some types of reactive groups such as carboxylic, primary and secondary amine, hydroxyl, and epoxide groups have been used as compatibilizers for different rubber/rubber and plastic/rubber blends.^{7,8} The segments of block, graft, or random copolymers can be identical to those in the respective phases. These

polymers reduce the macroscopic inhomogeneities and improve the morphological stability by decreasing the interfacial tension between incompatible phases.⁹⁻¹¹ The dynamics of phase separation in a binary mixture, following a thermal quench into the unstable coexistence region, proceeds by spinodal decomposition. Immediately after the quench, small domains, with local concentrations roughly corresponding to those of two immiscible phases, spontaneously form and grow without thermal activation energy and finally result in complete phase separation. However, when a surfactant molecule such as a block or random copolymer is added to an immiscible blend and absorbed at the immiscible interface, the kinetic and equilibrium properties of such a blend are significantly affected. $^{\rm 12-15}$ In this research, the homogeneity of poly(vinyl chloride) (PVC)/ethylene-propylene-diene monomer rubber (EPDM) was studied when a terpolymer of methyl methacrylate (MMA), ethyl acrylate (EA), and acrylic acid (AA) was coblended with PVC and EPDM in different proportions.

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EXPERIMENTAL

Materials

MMA, EA, and AA monomers, products of Merck (Darmstadt, Germany), were purified through stirring over active alumina (Fluka, Bushs, Switzerland) for 4 h and then filtration. Potassium persulfate $(K_2S_2O_8)$ and sodium dodecyl sulfate (SDS), products of Merck, were used as received. All other solvents were products of El-Nasr Pharmaceutical Co. (Egypt) and were analytical-grade. Vistalon-6505, EPDM with 9% ethylidene norbornene and a Mooney viscosity $[M_L(1 + 4) \text{ at } 100^{\circ}\text{C}]$ of 50, was a product of Esso Chimie (London, United Kingdom). Dicumyl peroxide [bis(1-methyl-1-phenylethyl) peroxide] was obtained from Aldrich Co. (St. Louis, MO) and used as curing agent. PVC was supplied by El-Amerria (Alexandria, Egypt). It was synthesized by suspension polymerization with a k value of 67. Dibutyltin maleate was used as a heat stabilizer. Other compounding ingredients were commercial grades.

Experimental techniques

Polymerization

The emulsion terpolymerization of MMA (1.0 mol), EA (1.5 mol), and AA (0.5 mol) was carried out under a nitrogen atmosphere at 70°C. The polymerization was carried out in a 2-L, three-necked, round-bottom flask equipped with a mechanical stirrer and thermostated water bath. An MMA (50 mL) and EA (50 mL) mixture was first seeded in the initial charge, 1 L of a 3.4 mM SDS surfactant solution and 3.7 mM K₂S₂O₈. After a 30-min induction period, the rest of the monomer phase was added dropwise over 4 h with a stirring rate of 100 rpm. The terpolymerization was continued for another 2 h at 80°C for complete polymerization. The final polymer conversion was determined gravimetrically.

Purification

The MMA–EA–AA terpolymer was precipitated in methanol from a tetrahydrofuran (THF) solution and then subjected to Soxhlet extraction of the homopolymers with acetone. The prepared terpolymer was then vacuum-dried at 40°C for 24 h until a constant weight was obtained.

Gel permeation chromatography (GPC)

The number-average molecular weight (M_n), weightaverage molecular weight (M_w), and molecular weight distribution (M_w/M_n) values for the prepared terpolymer were obtained with an Agilent (Germany) GPC instrument equipped with a series of three Styragel columns $(10^2, 10^3, \text{ and } 10^4 \text{ Å})$ calibrated with a series of narrow-molecular-weight polystyrene standards and a refractive-index detector (model 1100 differential refractometer, Agilent).

Thermal analysis

Glass-transition temperatures (T_g 's) of the MMA– EA–AA terpolymer were determined with a Shimadzu (Foster City, CA) model DSC-50 calorimeter. The specimens were cooled to -100° C and heated up to 110°C to remove their thermal history. The second runs were measured at a heating rate of 10°C/ min under a nitrogen atmosphere. Thermal degradation was studied with a model TGA 50 thermal gravimetric analyzer through the heating of samples X_{1–5} from the ambient temperature to 700°C.

¹H-NMR spectroscopy

The terpolymer composition was determined with $CDCl_3$ and a 500-MHz ¹H-NMR spectrometer (model EX-270, JEOL Technics Co., Ltd., Tokyo, Japan). The terpolymer sample was dissolved in THF and precipitated in a water/methanol mixture (50/50 v/v) twice, then subjected to Soxhlet extraction for homopolymers with acetone/water (70/30 v/v) as the mobile phase for 24 h, and then dried in a vacuum oven at 50°C for 48 h.

Preparation of the PVC/EPDM blends by the solution-casting technique

The required polymer solutions were prepared by the dissolution of 5 g each of the PVC and MMA– EA–AA terpolymer as well as 1.2 g of EPDM separately in 100 mL of THF. Polymer blend films were prepared through the casting of mixed solutions of different ratios (10/90, 20/80, 30/70, 40/60, and 50/ 50) of EPDM to PVC with and without 10 wt % MMA–EA–AA terpolymer on a glass plate with slow drying at room temperature. Care was taken to control the uniform film thickness for all compositions.

Mechanical testing

The physicomechanical properties of the films obtained were determined with a Zwick 1425 tensile tester (Munchen, Germany) at $25 \pm 1^{\circ}$ C and at a crosshead speed of 500 mm/min according to ASTM D 412-98a.

Mixing and processing of the EPDM/PVC blends

EPDM/PVC blend mixes with different blend ratios (10/90, 20/80, 30/70, 40/60, and 50/50) were pre-

pared in the presence and absence of the MMA-EA-AA terpolymer (10 parts by weight). MMA-EA-AA was first mixed with EPDM on an open two-roll mill (with a diameter of 170 mm and a working distance of 300 mm) at a slow-roll speed of 24 rpm and a gear ratio of 1:1.25 at 90°C for 2 min. The EPDM/ MMA-EA-AA mixture was then added to a PVC/ DOP (100/25) melt in the presence of 3% dibutyltin maleate as a heat stabilizer onto a Brabender plasticorder at 180°C for 2 min at a rotor speed of 30 rpm. The EPDM/PVC blends were then mixed with dicumyl peroxide (3 phr) on the two-roll mill for 2 min at 90°C. EPDM/PVC mixes were then subjected to sheeting on the mill. The blends were then pressed in a hydraulic press for 5 min at $165 \pm 1^{\circ}C$ with a pressure of 4 MPa on the mold.

Swelling test of the cured EPDM/PVC blends

The swelling test of the cured blends in brake fluid was conducted at $100\pm1^{\circ}$ C for 7 days in a thermostated electric oven according to ASTM D 471-97. Swelling data were measured in five replicates.

Scanning electron microscopy (SEM)

The morphology of the EPDM/PVC blends was studied with a model JXA-840A scanning electron microscope (JEOL Technics) at a magnification of $500 \times$ for scanning electron observations. The surface of the polymer was mounted on a standard specimen stub. A thin coating (10 Å) of gold was deposited onto the polymer surface and attached to the stub before examination in the microscope to enhance the conductivity.

Differential scanning calorimetry (DSC)

 T_g 's of blends were determined with a Shimadzu DSC-50 differential scanning calorimeter. It was operated at a heating rate of 10°C/min within the temperature range of -100 to 100°C. Rubber blend specimens were heated up to 100°C and then cooled to -100°C twice to remove their thermal history.

Dielectric measurements

Dielectric measurements were carried out in the frequency range of 100 Hz to 100 kHz with an AG-411 B LCR meter (Ando Electric, Ltd., Japan). The capacitance (*C*) and loss tangent (tan δ) were obtained directly from the bridge from which the permittivity (ε') and dielectric loss (ε'') were calculated. An NFM/5T type guard ring capacitor (Wiss Technical Werkstatten GmbH, Germany) was used as a measuring cell. The cell was calibrated with standard



Figure 1 Total conversion versus the time for the emulsion polymerization of MMA, EA, and AA.

materials, and the experimental errors in ε' and ε'' were found to be ±3 and ±5%, respectively. The cell temperature was controlled within ±0.1°C by the circulation of water from an ultrathermostat through a jacket surrounding the cell.

RESULTS AND DISCUSSION

Polymer synthesis

The conversion-time curve of the emulsion polymerization of MMA, EA, and AA is shown in Figure 1. The conversion determined after Soxhlet extraction of the homopolymers increases as the time of polymerization increases, reaching a maximum of 86%. The value would reach 99% under these reaction conditions in the absence of AA. The presence of AA as a comonomer increases the homopolymer conversion because of the solubility of the AA monomer in the dispersing phase, which results in AA drifting toward the shell rather than the polymer particle core. The presence of the poly(acrylic acid) (PAA) shell increases the latex stability and reactivity.¹⁶

Characterization

GPC

The GPC trace of the MMA–EA–AA terpolymer prepared is shown in Figure 2. The molecular weights $(M_n \text{ and } M_w)$ and polydispersity index were found to be 3.27×10^5 , 9.62×10^5 , and 2.91, respectively. The polydispersity index is quite low because of the presence of two polydispersity index raising factors: the nonlivingness of the system and the heterogene-



Figure 2 GPC trace of the MMA–EA–AA terpolymer. W: refractive index of polymer solutions. M: minus mvolts.

ous polymerization of AA in both emulsion and solution states.

Thermal analysis

A DSC scan of the MMA–EA–AA terpolymer is shown in Figure 3. The T_g transition endothermic peak appears at 17.5°C. The low T_g value of the terpolymer is attributed to the EA monomer, which reduces chain–chain interactions. This T_g is convenient for the application of this terpolymer because it lies between the T_g 's of EPDM and PVC.

A thermogravimetric analysis scan of the terpolymer is shown in Figure 4. The MMA–EA–AA terpolymer shows very good thermal stability: it loses 4.09 wt % at 267.58°C. This thermal stability of the prepared MMA–EA–AA terpolymer allows the application of this terpolymer not only in solution blending but also in melt blending for rubber/rubber, rubber/ plastic, and plastic/plastic blends.



Figure 3 DSC scan of the MMA–EA–AA terpolymer. Δ H: enthalpy.



Figure 4 Thermogravimetric analysis scan of the MMA–EA–AA terpolymer.

Terpolymer composition

The terpolymer composition was determined from the integral values of the metheine groups of AA at



Figure 5 SEM micrographs of EPDM/PVC blends with different blend ratios. (a) 50/50, (b) 40/60, (c) 30/70, (d) 20/80, and (e) 10/90 (magnification = $1000 \times$).

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Figure 6 SEM micrographs of EPDM/PVC blends with different blend ratios containing the MMA–EA–AA terpolymer: (a) 50/50, (b) 40/60, (c) 30/70, (d) 20/80, and (e) 10/90 (magnification = $1000 \times$).

 δ = 2.65 ppm, the methoxy group of MMA at δ = 3.5, and the methylene group of EA at δ = 4.1 ppm. This yielded 76.42% EA, 23.19% MMA, and 0.39% AA. The low incorporation of AA in the terpolymer is attributed to the PAA homopolymer and copolymer solubility in the polymerization aqueous medium. The absence of the carboxylic group proton in the spectrum is attributed to carboxylic protons



Figure 7 DSC traces of EPDM/PVC (40/60) blends (a) without and (b) with the MMA–EA–AA terpolymer.

exchangability with remaining H_2O protons; H^+ can cause peak brodening and sometimes disappearance from the spectra.

SEM

EPDM/PVC blend films were prepared with different blend ratios (10/90, 20/80, 30/70, 40/60, and 50/50) in the presence and absence of the MMA-EA-AA terpolymer as a compatibilizer. The morphology of those blends was examined with the aid of a scanning electron microscope. The SEM micrographs in the absence of the MMA-EA-AA terpolymer are illustrated in Figure 5(a-e). Figure 5(a) illustrates the appearance of two different phases for the EPDM/PVC (50/50) blend, and phase separation took place. Also, the formation of two different morphologies for the EPDM/PVC (40/60) blend is obvious from Figure 5(b). The phase separation is also clear for the EPDM/PVC (30/70) blend as shown in Figure 5(c). As the amount of EPDM decreased to 20 parts and the amount of PVC increased to 80 parts [Fig. 5(d)], the EPDM agglomerates separated from the PVC phase. The EPDM agglomerates spread through the PVC phase as the amount of EPDM decreased to 10 parts and the amount of PVC increased to 90 parts [Fig. 6(e)]. The morphology of the EPDM/PVC blends with different blend ratios was examined in the presence of MMA-EA-AA to test it as a compatibilizer. The SEM micrographs are illustrated in Figure 6. The phase separation disappeared for EPDM/PVC blends with different blend ratios containing the MMA-EA-AA terpolymer. The morphology shows an improvement of the EPDM/PVC blend homogeneity for all the blend ratios investigated. This can be explained as a result of the interaction between



Figure 8 Tensile strength at yield and tensile strength and elongation at rupture of EPDM/PVC blend films versus the EPDM content in the blend.



Figure 9 Tensile strength at yield and tensile strength and elongation at rupture of EPDM/PVC blend films containing 10 parts by weight MMA–EA–AA terpolymer versus the EPDM content in the blend.

the nonpolar part of the terpolymer with the nonpolar EPDM and the interaction between the polar part of the terpolymer with the polar PVC; this in turn results in decreasing the surface tension of the two phases, leading to the improvement in the blend homogeneity.

DSC

DSC was used to detect qualitatively the homogeneity of the EPDM/PVC blends. Figure 7(a,b) illustrates DSC traces of EPDM/PVC (40/60) blends with and without MMA–EA–AA. T_g 's of EPDM and PVC in the blend appear at -50 and $+50^{\circ}$ C, respectively, with a T_g difference of 100°C. However, T_g 's of EPDM and PVC in the blend containing MMA–EA– AA appear at -45° C and $+49^{\circ}$ C, respectively, with a T_g difference of 94°C. These data illustrate that T_g 's of EPDM and PVC became closer to each other upon the incorporation of MMA–EA–AA. This may be attributed to a sufficient interaction between the polar segments of MMA–EA–AA with the PVC phase and with the EPDM phase, reducing the interfacial tension that leads to the increase in adhesion between the phases. Therefore, MMA–EA–AA can be used successfully for a reduction of the macroscopic inhomogeneity and improvement of the homogeneity and for increasing the stability of the morphology during the mixing of an EPDM/PVC blend.

Mechanical properties

EPDM/PVC blend films were cut with a dumbbellshaped cutter. Cross-section areas of the dumbbell specimens were measured accurately with the aid of a thickness gauge. The mechanical properties of the blend films with different blend ratios in the presence and absence of the MMA-EA-AA terpolymer were measured and are illustrated in Figures 8 and 9. As the EPDM amount increases in the blend, in the absence of the terpolymer (Fig. 8), elongation at rupture increases. This can be attributed to the elasticity of EPDM. However, the tensile strengths at yield and at rupture decrease because of the increase in the amount of uncured rubber. In the presence of the terpolymer (Fig. 9), elongation at rupture increases linearly, whereas tensile strengths at yield and at rupture change linearly as the amount of EPDM increases in the blend. The linear behavior of the mechanical properties versus the blend ratios indicates the homogeneity of EPDM/PVC blends due to the incorporation of the terpolymer.

Swelling behavior in brake fluid

Cured EPDM/PVC blends with different blend ratios (10/90, 20/80, 30/70, 40/60, and 50/50) were prepared in the presence and absence of the MMA–EA–AA terpolymer (10 parts by weight). The formu-

TABLE I Formulations of EPDM/PVC Blends with Different Blend Ratios in the Presence and Absence of the MMA–EA–AA Terpolymer

Ingredient (phr)	Designation									
	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10
EPDM	10	20	30	40	50	10	20	30	40	50
PVC	90	80	70	60	50	90	80	70	60	50
DOP	22.5	20	17.5	15	12.5	22.5	20	17.5	15	12.5
Dibutyltin maleate	2.7	2.4	2.1	1.8	1.5	2.7	2.4	2.1	1.8	1.5
MMA–EA–AA terpolymer	0	0	0	0	0	10	10	10	10	10
Zinc oxide	0.5	1	1.5	2	2.5	0.5	1	1.5	2	2.5
Stearic acid	0.15	0.3	0.45	0.6	0.75	0.15	0.3	0.45	0.6	0.75
Silica	10	10	10	10	10	10	10	10	10	10
Processing oil	5	5	5	5	5	5	5	5	5	5
Dicumyl peroxide	0.3	0.6	0.9	1.2	1.5	0.3	0.6	0.9	1.2	1.5
Cure time at 165°C (min)	5	5	5	5	5	5	5	5	5	5



Figure 10 Weight swelling in the brake fluid of EPDM/ PVC blends with and without the terpolymer compatibilizer versus the EPDM content in the blends.

lations are listed in Table I. The cured blends were subjected to swelling tests in brake fluid. In the absence of the MMA–EA–AA terpolymer, Figure 10 illustrates that the weight swelling in brake fluid shows an irregular pattern with increasing EPDM content in the blend. However, inverse swelling behavior in brake fluid is noticed in the presence of the MMA–EA–AA terpolymer: the weight swelling shows linear behavior with increasing EPDM content in the blend. This may be attributed to the increase in the homogeneity of the EPDM/PVC blends due to the MMA–EA–AA terpolymer. However, the



Figure 11 ε' and ε'' of EPDM/PVC blends with different blend ratios versus the frequency at room temperature.

effect of the MMA–EA–AA terpolymer on the swelling reduction of the cured blends in brake fluid is more pronounced in EPDM/PVC blends (40/60 and 50/50). This could be attributed to the increase in the polarity of EPDM as a result of the incorporation of the MMA–EA–AA terpolymer. Because of its low cost compared to EPDM alone and its low weight swelling in brake fluid, the EPDM/PVC (50/50) blend containing the MMA–EA–AA terpolymer (10 parts by weight) can be recommended for applications in oil seals and hoses subjected to brake fluid.

Dielectric properties

As the incorporation of the MMA–EA–AA terpolymer (10 parts by weight) into the EPDM/PVC blend enhances the mechanical properties of that blend, it was interesting to investigate its effect on the dielectric properties of the investigated blends.

 ε' and ε'' of cured EPDM/PVC blends with different blend ratios (10/90, 20/80, 30/70, 40/60, and 50/50) were determined in the frequency range of 100 Hz to100 kHz. Figures 11 and 12 present the var-



Figure 12 ε' and ε'' of EPDM/PVC blends with different blend ratios in the presence of the terpolymer versus the frequency at room temperature.

4.5

4 N

3.5

2.5

2.0

4.0

τω 3.0





Figure 13 ε' versus the EPDM content in EPDM/PVC blends with and without the terpolymer at 100 Hz and 100 kHz at room temperature.

iation of ε' and ε'' with the applied frequency (f) in the presence and absence of MMA-EA-AA. From these figures, we can see that ε' decreases slightly with *f* increasing, showing an anomalous dispersion. In such a range, ε' has a contribution from the orientation and atomic and electronic polarization.¹⁷ The variation of ε'' with *f* is shown in the same figures. The values of ε'' at low frequencies are higher than those at high frequencies. From the same figures, it is clear that the curves relating ε'' and log *f* are broader than the Debye curve, and this indicates that more than one relaxation process is present. These processes could be attributed to mechanisms related to the main chain and its related motions.¹⁸ One of the expected mechanisms is related to the Maxwell-Wagner effect, which usually occurs at the lower frequency region for heterogeneous systems. The origin of such a process is an alternating current that is in phase with the applied potential due to the difference in the ε' values and resistivities of the blend components. ε' is plotted graphically versus the EPDM content in EPDM/PVC blends in the presence and absence of the MMA-EA-AA terpolymer, at f = 100 Hz and f = 100 KHz, in Figure 13. From this figure, it is clear that the values of ε' show

a curved shape in the absence of the MMA–EA–AA terpolymer. This behavior could indicate that EPDM/PVC blends are incompatible. However, in the presence of the terpolymer, the plot of ε' versus the EPDM content in the EPDM/PVC blends shows a linear relation indicating the homogeneity of such blends. This conclusion is considered to be good support for the results obtained from the mechanical properties of the blend films and from the swelling behavior of the cured blends in brake fluid.

CONCLUSIONS

An MMA–EA–AA terpolymer was synthesized and characterized with GPC, thermal analysis (thermogravimetric analysis and DSC), and ¹H-NMR spectroscopy. The incorporation of the terpolymer into EPDM/PVC blends is beneficial because of the increase in the homogeneity of such blends, as indicated by a microscopy examination of the surface morphology of the blend, DSC traces, mechanical property determination, swelling tests in brake fluid, and dielectric measurements for EPDM/PVC blends.

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