

Development of HDPE/EPDM-g-TMEVS Nanocomposites with Enhanced Electrical and Flame Properties

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ABSTRACT: Nanoclay reinforced HDPE/silane grafted EPDM composites have been developed using an epoxy functionalized HDPE as compatibilizer. The nanoclay has been varied from 0% to 10% in the composites along with the incorporation of compatibilizer and without compatibilizer in a Brabender plastic order. The dielectric and fire retardant properties of these nanocomposites have been examined. Addition of nanoclay enhanced char formation with increased values of limiting oxygen index. Electrical properties such as volume and surface resistivity improved with addition of nanoclay and compatibilizer. The values of $\tan \delta$ increased with increase in grafted EPDM and silanated nanoclay loading. © 2013 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 130: 352–358, 2013

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INTRODUCTION

The research in polymer materials is being increasingly focused on the development of polymer nanocomposites in the last decade. The objective of flame retarding polymers is to increase the resistance of material to ignition and inhibit flame propagation with minimal degradation of its properties. Addition of flame retardant material to polymer will suppress propagation of flame. These in turn change combustion characteristics of polymeric material which becomes more difficult to ignite. Once ignition takes place flame retardants will cause flame to extinguish or burn slowly which slows down flame propagation. Polyolefin-based nanocomposites are very important due to formation of intercalated structure with layered clay silicates with conventional processing technology.^{1,2} Recently halogen-free, low smoke, flame-retardant composites are becoming increasingly important particularly for polyolefins such as ethylene vinyl acetate copolymer used in cable and wire industry as halogen-type flame retardant cause environmental problems such as toxicity, corrosion, and smoke. Traditional fillers like calcium carbonate, talc, mica, silica, alumina, magnesium hydroxide require high loading to achieve the required performance. However, high loading of these nonhalogenated mineral fillers reduces the mechanical properties due to poor interfacial adhesion. Thus, nanosized fillers are being used as they have large surface area and hence lesser loading of filler is required. Further, if the interfacial adhesion can be improved, the blend

can exhibit both improved fire retardancy as well as enhanced mechanical strength. In addition, thermoplastics such as HDPE (high density polyethylene) and EVA (ethylene vinyl acetate) are commonly used insulation materials with good dielectric properties. Thus, in this study, surface modified nanoclay has been blended with HDPE and EPDM (ethylene propylene diene monomer) elastomer. Chang et al.³ observed that the presence of MMT (montmorillonite) promoted the formation of char layer for (low density polyethylene) LDPE/EPDM nanocomposites. Blending of γ -irradiated HDPE/EPDM composites led to increased char formation as reported by Jia et al.⁴ Similar observations have been reported for (polypropylene) PP/EPDM composites by the addition of melamine phosphate.⁵ Silane crosslinking of PP/EPDM using a coupling agent was found to improve mechanical properties although fire retardancy had to be compromised to a certain extent.⁶ Apart from flame retardant properties a recent report on HDPE/Clay nanocomposites suggested that the electrical properties were significantly improved when the organoclay used has been surface treated with silane.⁷ Triallyl cyanurate has been used for crosslinking and improving the flame retardancy of HDPE/EPDM blends via electron beam irradiation by Shin et al.⁸

Thus, in order to enhance interfacial adhesion, EPDM has been grafted with silane and reinforced with silane treated nanoclay. In this study a small amount of epoxy functionalized HDPE has been used as compatibilizer to anchor the three blend

components together. The developed nanocomposites were examined for both dielectric properties as well as flame retardancy.

MATERIALS AND METHODS

Materials

High Density Polyethylene (HDPE) of 4 g/10 min melt flow index has been purchased from, Saraswathi Plastics, Bangalore, India. EPDM (EP33, Japan Synthetic Rubber Company, Japan) used in the present study is a terpolymer containing ethylene, propylene and 5-ethylidene-2-norbornene (ENB) and their weight percentage composition is 71, 20, and 9, respectively. The tris (2-methoxyethoxy) vinylsilane (TMEVS) and nanoclay (nanomer) is procured from Sigma Aldrich; Switzerland. The analytical grades of dicumyl peroxide (DCP) initiator, Glycidyl methacrylate (GMA) and dibutyl-tin-dilaurate (Merck India) catalyst have been used as received.

Grafting: Preparation of EPDM-g-TMEVS

EPDM was grafted with tris(2-Methoxyethoxyvinylsilane (TMEVS)) using Dicumyl peroxide (Initiator) in a Brabender plasticorder (Plasticorder, CMEI, 16CMESPL, East Germany). Seventy grams of chipped EPDM granules were coated with 3.5 g of TMEVS containing 0.07 g of dicumylperoxide and 0.007 g of dibutyl-tin-dilaurate catalyst has been put into the plasticorder. The grafting of TMEVS onto EPDM was carried out at 180°C for 4 min at a rotation speed of 60 rpm. The EPDM-g-TMEVS granules obtained were dried at 60°C to constant weight.

Synthesis of Epoxy Grafted HDPE Compatibilizer

The grafting of HDPE-g-GMA was carried out in a plasticorder using GMA with benzoyl peroxide as initiator for 4 min. The product was cooled and chipped into granules. The HDPE-g-GMA granules obtained were dried at 60°C to constant weight.

Blend Preparation: Preparation of HDPE/EPDM-g-TMEVS Blends

The blends of EPDM-g-TMEVS and HDPE were prepared by melt mixing in Brabender (Plasticorder, CMEI, 16CMESPL, East Germany) at 180°C for 5 min by varying percentage (0, 1, 2, 5, 7, and 10%) of nanoclay. During the preparation of HDPE blends, the polymer and compatibilizer were mixed in a molten state for 5 min and nanoclay was added and mixing was carried out continuously at 180°C for another 3 min, followed by addition of dicumyl peroxide over a period of 1 min.

Compression Molding

The blends obtained from Brabender were pressed into sheets in a compression mould (Hot press, Lab Tech) at 15 Mpa pressure and 120°C. The heating time was kept at 10 min and the curing time at 20 minutes.

Transmission Electron Microscopy

The morphology of the blends were observed by Transmission Electron Microscopy (TEM, TECHNAI -T12). The samples were cut to size and trimmed and microtomed at -140°C. Microtomed samples were picked in Cu grid.

Characterization: FTIR Spectroscopy

Fourier transform infrared spectroscopy (FTIR) (Perkin-Elmer spectrum 1000) analysis for HDPE, EPDM, EPDM-g-TMEVS, and blends were performed.

Electrical Properties of Blends

Volume Resistivity. Volume Resistivity was measured as per ASTM D257 standard. Volume Resistance was measured with a high resistance meter from Hewlett-Packard (model 4329A). Compression moulded sheet having diameter 100 mm was inserted into the sample holder (cell) and charged for 1 min at 500 V (direct current). Volume resistance measurements were carried out at 27°C and expressed in ohm centimetres.

Surface Resistivity

Surface resistivity was measured as per ASTM D257 standard. Surface resistance was measured with a high resistance meter from Hewlett-Packard (Make USA, Model 4329A). Compression molded sheet having diameter 100 mm was inserted into the sample holder (cell) and charged for 1 min at 500 V.

Tan Delta

Tan Delta was measured as per IS 6262 using Tettex Bridge (Make Switzerland, Model 2822). Compression molded sheet having diameter 100 mm was inserted into the sample holder (cell) and pressure of 2 N/Cm² was applied and charged for 1 min at 500 V.

Flame Properties of Blends

Oxygen Index Time. Stanton redcroft FTA Flammability Unit was used to determine minimum concentration of oxygen in an oxygen/nitrogen mixture that will support a flame to burn a plastic specimen as per ASTM D2863.

Cone Calorimeter. Material specimen size of 100 x 100 mm in area and 3 mm thick plaques were considered in this study. Specimens were wrapped in single layer of aluminum foil of 0.1 mm thick with shiny side toward specimen, covering sides, bottom and top surface exposed to thermal irradiance in aluminum foil tray of 0.1 mm thick and wrapped specimen was placed on top of bed low density refractory fiber blanket in sample holder. Test specimens were irradiated at a heat flux of 35 kW/m² using a truncated conical heater element to simulate a range of fire intensities. Tests were conducted as per ISO 5660-1 Standard.

RESULTS AND DISCUSSION

Fourier Transform Infrared Spectroscopy

Figure 1(a,b) represents FTIR spectrum of HDPE, HDPE-g-GMA, EPDM, and EPDM-g-TMEVS. Figure 1(a) shows the FTIR spectra of pure HDPE- and HDPE-grafted GMA which has been used as a compatibilizer. The characteristic peak at 1730 cm⁻¹ for the carboxyl stretching vibration of GMA and at 721 cm⁻¹ for CH₂-rocking vibration corresponds to polyethylene chain.⁹ Figure 1(b) represents FTIR spectrum of EPDM and EPDM-g-TMEVS, Shows CH₂ wagging vibration at 750 cm⁻¹, may be due to presence of polyethylene chain. The unsaturation band (>C=CH-) at 820 cm⁻¹ is due to the presence of 2-ethylidene-5-norbornene content. IR spectrum of EPDM-g-TMEVS, indicates -CH₂ rocking vibration at 1350 cm⁻¹, -CH₃ symmetric bending vibration of 1407 cm⁻¹ and C-H stretching vibration at 2921 cm⁻¹. However, -CH₂ wagging vibration of Si-CH=CH₂ at 875 cm⁻¹, C-C stretching at 1629 cm⁻¹, and C-H stretching vibration (vinyl) at 3027 cm⁻¹ disappeared because of grafting of TMEVS with unsaturation present in side chains of ENB monomer at 820 cm⁻¹ without affecting the Si-O stretching vibration at 1025 cm⁻¹. The absence of peaks

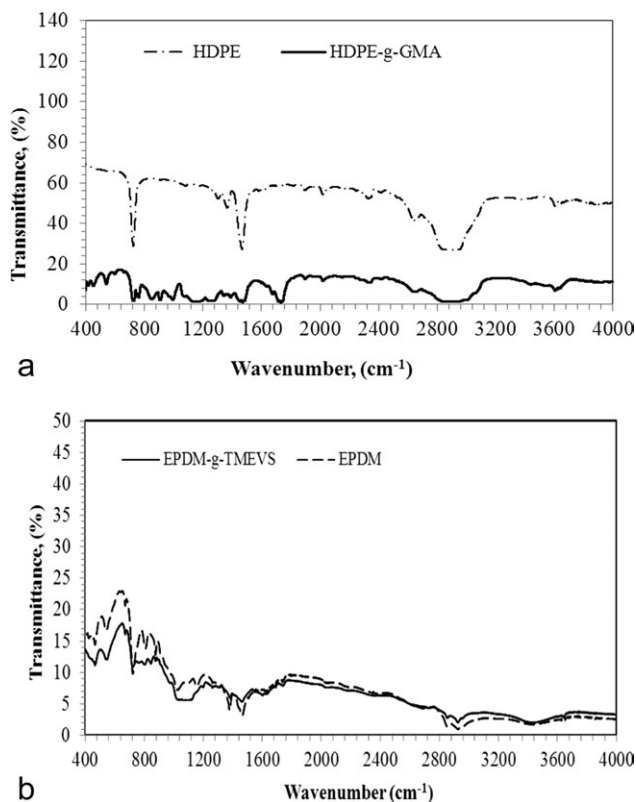


Figure 1. (a) FTIR spectra of HDPE and HDPE-g-GMA. (b) FTIR spectra of EPDM and EPDM-g-TMEVS.

at 820, 875, and at 3027 cm⁻¹ indicates that $>C=CH-$ has been utilized in the reactive blending.¹⁰

Thermogravimetric Analysis

Samples are heated from ambient temperature to 800°C at a heating rate of 10 deg/min in nitrogen atmosphere. Figure 2(a,b) shows TGA thermograms for HDPE/EPDM-g-TMEVS blends with varying nanoclay loadings. The values of inception temperature, peak temperature, final decomposition temperature, and weight loss are presented in Table I. Pure HDPE and blends with varying EPDM-g-TMEVS without nanoclay and compatibilizer are also shown in figure for comparison. Pure HDPE is thermally stable and has single stage degradation at 546°C owing to decomposition of $-C-C-$ backbone of polyethylene chain. It is noticed with increase in EPDM-g-TMEVS there is an increase in inception, peak and final decomposition temperature of blends. It is ascertained there is single stage degradation due to scission of conjugated polythene chains and further this supports fact that blends have single chemical entity as reported by Alagar et al.¹¹

Figure 2(b) shows 10 % EPDM blends with 10% nanoclay char formation of about 7.86%. Similarly for 20% and 30% EPDM blends with 10% nanoclay loadings showed char formation of about 8.58% and 8.85%, respectively. Thus, this indicates that addition of nanoclay led to char formation which in turn slows down flame propagation.

Limited Oxygen Index

Table II shows the limiting oxygen indexes of HDPE/EPDM-g-TMEVS blends. Addition of silane-grafted EPDM slightly improved LOI (Limiting oxygen Index) values. However, addition of compatibilizer and nanoclay (NC) increased the LOI value to 23.5 for 10% EPDM-g-TMEVS loading. For 20% grafted EPDM and 10% silane-treated nanoclay the LOI value increased to 25.8. For 30% EPDM-g-TMEVS and 10% NC along with compatibilizer exhibited an LOI value of 28.7 as compared with 19.8 for neat HDPE. In all cases, compatibilization did not significantly effect the LOI value. Addition of cross-linked polyethylene helps formation of char.¹² Further, addition of nanoclay also helps in char formation due to increase in barrier properties as reported Ozkaraca et al.¹³

Cone Calorimeter Studies

Figure 3(a) and Table III gives the analysis of cone calorimetry for HDPE/EPDM-g-TMEVS blends. As per the LOI tests, the blend with 30% silane crosslinked EPDM showed the maximum value. Hence, cone calorimetry was carried out for this composition.

The test were performed on blends without compatibilizer and 0% nanoclay and blends of HDPE: EPDM-g-TMEVS: 70:30 for 2% and 7% nanoclay loading with and without compatibilizer. Results for the test have been summarized in Table III. Here

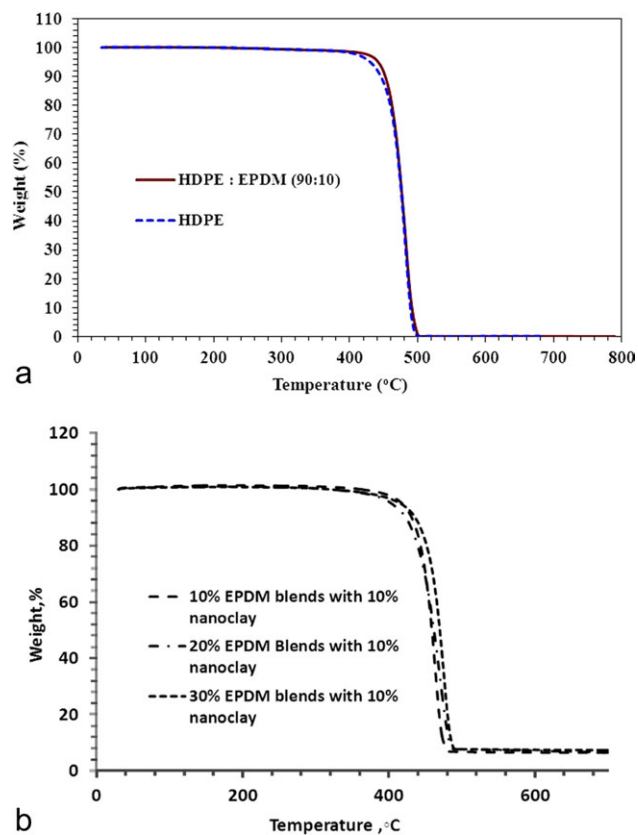


Figure 2. (a) TGA curve of HDPE and HDPE:EPDM-g-TMEVS (90:10) blends. (b) TGA curve of 10, 20, and 30% EPDM blends with 10% nanoclay. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table I. Temperature and Weight Loss (%) for Blends

| Blend composition | | | | Inception | Peak | Final |
|-------------------|--------------|------------------|------------|--------------------------------|------|-------|
| HDPE | EPDM-g-TMEVS | % Compatibilizer | % Nanoclay | | | |
| | | | | Decomposition temperature (°C) | | |
| 90 | 10 | 0 | 0 | 459 | 482 | 510 |
| 80 | 20 | 0 | 0 | 457 | 481 | 500 |
| 70 | 30 | 0 | 0 | 456 | 478 | 498 |
| 90 | 10 | 0 | 1 | 437 | 469 | 490 |
| 90 | 10 | 0.6 | 1 | 425 | 466 | 485 |
| 90 | 10 | 0 | 5 | 431 | 460 | 480 |
| 90 | 10 | 0.6 | 5 | 444 | 467 | 475 |
| 80 | 20 | 0 | 1 | 442 | 476 | 495 |
| 80 | 20 | 1.2 | 1 | 445 | 474 | 492 |
| 80 | 20 | 0 | 5 | 441 | 475 | 490 |
| 80 | 20 | 1.2 | 5 | 440 | 469 | 485 |
| 70 | 30 | 0 | 1 | 444 | 475 | 495 |
| 70 | 30 | 1.8 | 1 | 448 | 476 | 495 |
| 70 | 30 | 0 | 5 | 442 | 473 | 490 |
| 70 | 30 | 1.8 | 5 | 441 | 467 | 490 |

combustion parameters of composites containing 2% nanoclay and 7% nanoclay without compatibilizer were compared with that of the composites with compatibilizer.

Figure 3(a) and Table III shows the heat release rate for the HDPE/silane-grafted EPDM blend. It can be seen, time of ignition reduces for compatibilized blends owing to improved dispersion of the blend components. However, an increase in nanoclay loading increases the ignition time. The heat release rate (HRR) and the peak heat release rate (PHRR) reduces as the content of nanoclay increases. Compatibilization further lowers the HRR and PHRR values as seen in Table III and Figure 3(a). Similar observations for functionalized clay nanocomposites have been reported by Wang et al.¹⁴

Ozkaraca et al.¹³ studied the effect of nanoclays on flame retardancy characteristics for ABS and it was suggested that the condensed phase flame retardancy mechanism is the predominant mode. In addition the reduction of PHRR value is attributed to exfoliation of nanoclay which acts as a barrier for heat flow and flammable gases. This also becomes clear from the plot of HRR versus time as shown in Figure 3(a). However, there is no improvement in smoke emission as shown in Figure 3(b) and Table III. Hence there is a raise in SEA (Specific extinction area) values. A similar observation has been reported for HDPE/EPDM/Silicon elastomer blends by Jia et al.⁴ It was suggested that increase in the carbon atoms fraction also increases the smoke emission. The addition of nanoclay did not affect CO emission although a slight reduction on CO₂ emissions could be noted.

Electrical properties

Volume Resistivity. Figure 4(a–c) shows the volume resistivity of HDPE/EPDM-g-TMEVS composites. The volume resistivity values improved by the increase in nanoclay. Addition of

Table II. Limited Oxygen Index of HDPE/EPDM-g-TMEVS Blends

| Blend composition | | | | |
|-------------------|--------------|------------------|------------|----------|
| HDPE | EPDM-g-TMEVS | % Compatibilizer | % Nanoclay | % Oxygen |
| 100 | 0 | 0 | 0 | 19.8 |
| 90 | 10 | 0 | 0 | 20 |
| 80 | 20 | 0 | 0 | 21 |
| 70 | 30 | 0 | 0 | 22 |
| 90 | 10 | 0 | 1 | 21 |
| 90 | 10 | 0.6 | 1 | 21.5 |
| 90 | 10 | 0 | 5 | 22 |
| 90 | 10 | 0.6 | 5 | 22.5 |
| 90 | 10 | 0 | 10 | 23 |
| 90 | 10 | 0.6 | 10 | 23.5 |
| 80 | 20 | 0 | 1 | 23 |
| 80 | 20 | 1.2 | 1 | 23.5 |
| 80 | 20 | 0 | 5 | 24 |
| 80 | 20 | 1.2 | 5 | 24.6 |
| 80 | 20 | 0 | 10 | 25 |
| 80 | 20 | 1.2 | 10 | 25.8 |
| 70 | 30 | 0 | 1 | 26 |
| 70 | 30 | 1.8 | 1 | 26.5 |
| 70 | 30 | 0 | 5 | 27 |
| 70 | 30 | 1.8 | 5 | 27.5 |
| 70 | 30 | 0 | 10 | 28 |
| 70 | 30 | 1.8 | 10 | 28.7 |

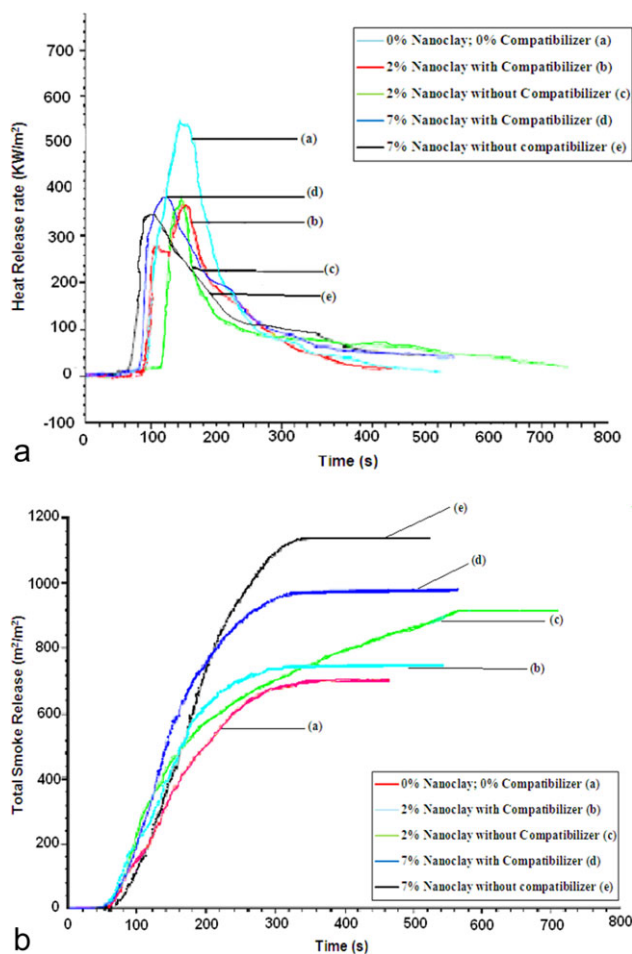


Figure 3. (a) Plot of Heat release rate v/s time for HDPE: EPDM blends with 2 and 7% nanoclay. (b) Plot of total smoke released vs. time for HDPE: EPDM blends with 2 and 7% nanoclay. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

compatibilizer has also further improved the volume resistivity values. However, increased addition of grafted EPDM reduced volume resistivity values.

The maximum value of volume resistivity has been observed for 10% nanoclay at 9.2419 ohm_{cm} for compatibilized blends

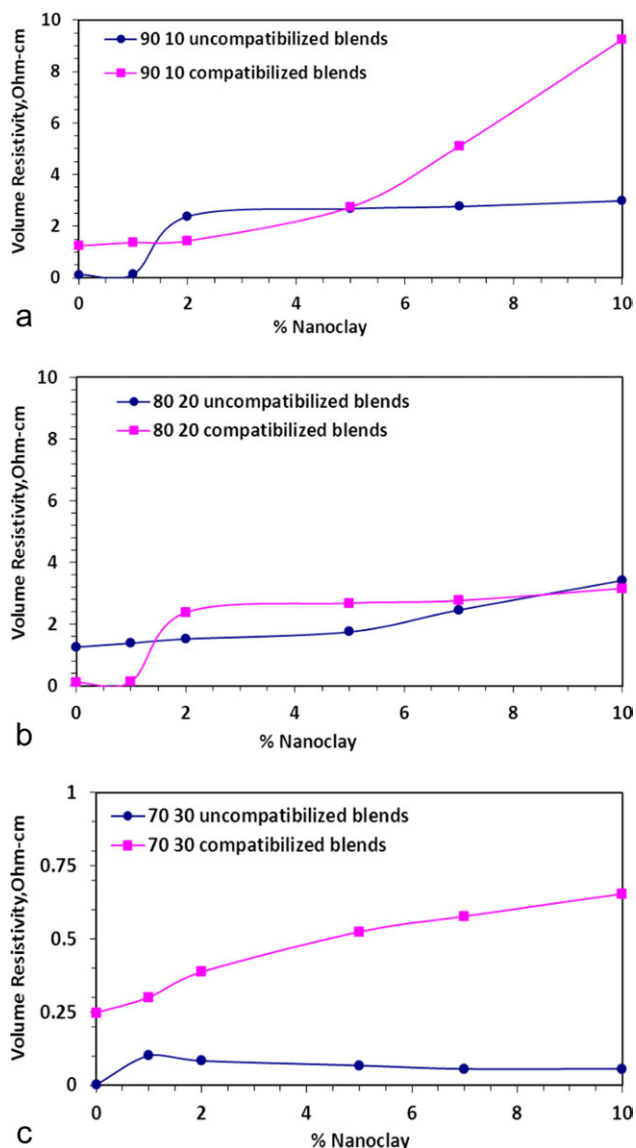


Figure 4. (a) Volume resistivity versus nanoclay for 10% grafted EPDM blends. (b) Volume resistivity versus nanoclay for 20% grafted EPDM blends. (c) Volume resistivity versus nanoclay for 30% grafted EPDM blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table III. Cone Calorimeter Results for HDPE: EPDM 70:30 Blends with and without Compatibilizer

| Sample | Blank | WOC | WC | WOC | WC |
|--|--------|--------|--------|--------|--------|
| Nanomer | 0% NC | 2%NC | 2%NC | 7% NC | 7% NC |
| Heat flux kW/m ² | 35 | 35 | 35 | 35 | 35 |
| Time to ignition, s | 87 | 82 | 76 | 83 | 77 |
| Burning time, s | 549 | 529 | 477 | 525 | 475 |
| Heat release rate, kW/m ² | 337.91 | 185.25 | 127.71 | 181.63 | 124.76 |
| Peak Heat release rate, kW/m ² | 587.66 | 383.05 | 370.11 | 352.42 | 340.47 |
| Specific extinction area(SEA) (m ² /kg) | 316.91 | 286.83 | 350.04 | 407.58 | 471.21 |
| Total smoke release m ² /m ² | 590.5 | 621.1 | 613.1 | 711.1 | 703.2 |
| Carbon monoxide yield (kg/kg) | 0.0418 | 0.0363 | 0.0359 | 0.0505 | 0.0498 |
| Carbon dioxide yield (kg/kg) | 2.57 | 1.95 | 2.10 | 2.15 | 2.30 |

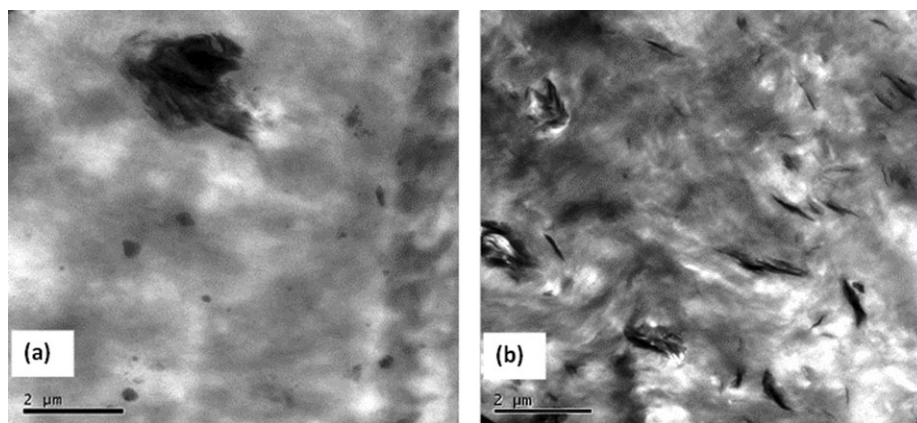


Figure 5. (a) TEM image of HDPE/EPDM composites with 1% nanoclay. (b) TEM image of HDPE/ EPDM composites with 7% nanoclay.

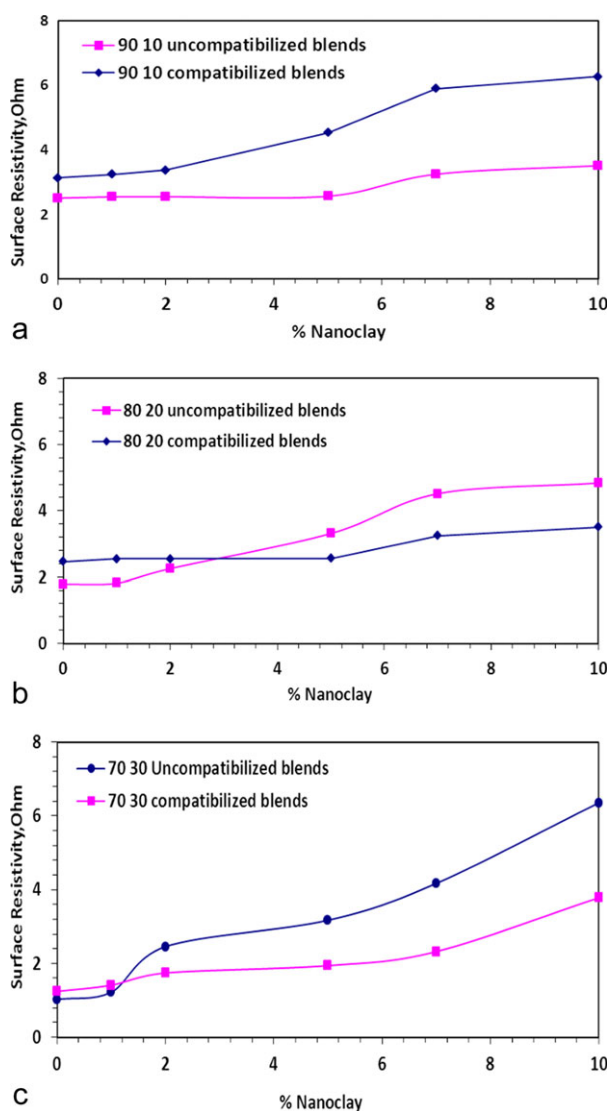


Figure 6. (a) Surface resistivity versus nanoclay for 10% grafted EPDM blends. (b) Surface resistivity versus nanoclay for 20% grafted EPDM blends. (c) Surface resistivity versus nanoclay for 30% grafted EPDM blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

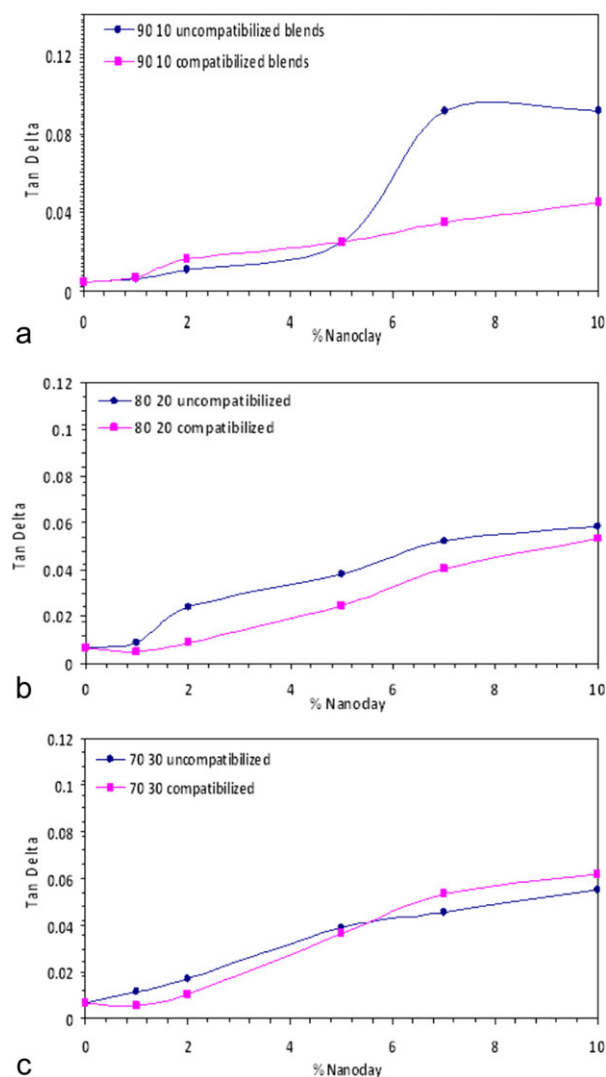


Figure 7. (a) Tan delta versus nanoclay for 10% grafted EPDM blends. (b) Tan delta versus nanoclay for 20% grafted EPDM blends. (c) Tan delta versus nanoclay for 30% grafted EPDM blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

loaded with 10% grafted EPDM. Similar trends have been noted for blends with 20% and 30% grafted EPDM. However, the optimal results can be seen for 10% loading of grafted EPDM. A similar observation has been reported by Shah et al.¹⁵ for HDPE/Clay nanocomposites along with a coupling agent. It was suggested exfoliation and better dispersion of nanoclay along with coupling agent led to improved interfacial adhesion between the blend components. Further, clay particles led to treeing effect as they act as breakdown barrier.

Figure 5(a,b) shows the TEM micrographs of the nanocomposites loaded with 1 and 2% nanoclay. It can be seen from the figures that the nanoclay has exfoliated although pockets of agglomerated nanoparticles can be seen. The amine groups of silanated nanoclay along with functionalized EPDM interacts efficiently with the epoxy group of the compatibilizer thereby anchoring the components of the nanoblend.

Surface Resistivity. Surface resistivity of HDPE/EPDM-g-TMEVS blends improve as nanomer content increases as shown in Figure 6(a–c). Surface resistivity followed a similar trend as volume resistivity. Silane linkages help reduce the surface roughness and have an anchoring effect between the blend components.¹⁵ The maximum values of surface resistivity has been attained with blends reinforced with 10% nanoclay along with epoxy grafted compatibilizer.

Dissipation Factor or Loss Tangent ($\tan \delta$). Figure 7(a–c) shows the plots of nanomer content versus $\tan \delta$. The $\tan \delta$ values increase as the grafted EPDM content increases (even without nanoclay or compatibilizer). Further addition of nanoclay also increase the $\tan \delta$ value. As both EPDM and nanoclay are modified by silane, it imparts polarity to the blends and polar molecules exhibit dielectric power losses as compared to nonpolar polymers. Similar observation for treated carbon nanofiber composites has been reported by Yang et al.¹⁶

CONCLUSIONS

HDPE has been blended with silane-grafted EPDM along with nanoclay using epoxy functionalized HDPE as interfacial modifier. TGA studies suggested char formation due to nanoclay addition. Limited Oxygen Index exhibited an LOI value of 28.7 as compared with pure HDPE 19.8. Cone calorimetric studies revealed that the time of ignition reduced for compatibilized

blends owing to improved dispersion of blend components while increase in nanoclay loading increased the ignition time. The heat release rate and peak heat release rate reduced as the nanoclay content increases. The volume resistivity values improved with increase in nanoclay content which were further enhanced with the addition of compatibilizer. Surface resistivity followed a similar trend as volume resistivity. The $\tan \delta$ values increased as the grafted EPDM content increases. Nanoclay content also increased the $\tan \delta$ value.

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