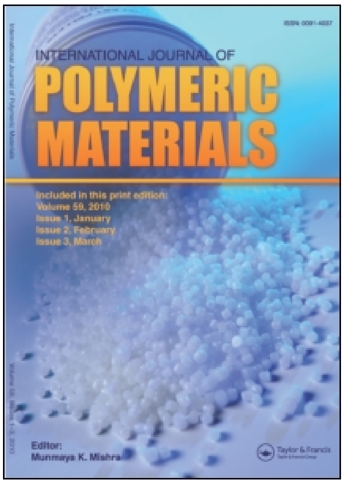

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Properties of Paper Sludge Filled Polypropylene (PP)/Ethylene Propylene Diene Terpolymer (EPDM) Composites: The Effect of Silane-Based Coupling Agent

Salmah[a]; H. Ismail[b]; A. Abu Bakar[b]
[a] School of Materials Engineering, Northern Malaysia College University of Engineering (KUKUM), Jejawi, Perlis, Malaysia [b] School of Materials and Mineral Resources Engineering, Universiti Sains Malaysia, Nibong Tebal, Penang, Malaysia

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Properties of Paper Sludge Filled Polypropylene (PP)/Ethylene Propylene Diene Terpolymer (EPDM) Composites: The Effect of Silane-Based Coupling Agent

Salmah
School of Materials Engineering, Northern Malaysia College
University of Engineering (KUKUM), Jejawi, Perlis, Malaysia

H. Ismail
A. Abu Bakar
School of Materials and Mineral Resources Engineering,
Universiti Sains Malaysia, Nibong Tebal, Penang, Malaysia

Paper sludge was used as a filler in PP/EPDM composites and 3-aminopropyl triethoxysilane (3-APE) was used in this study as a coupling agent. The effects of filler loading and 3-APE on the mechanical properties, water absorption, morphology, and thermal properties of the composites were investigated. It was found that incorporation of a silane coupling agent (3-APE) increased the stabilization (equilibrium) torque, tensile strength, and Young's modulus but decreased the elongation at break and water absorption. Scanning electron microscopy (SEM) study of the tensile fracture surface of the composites indicated that the presence of 3-APE increased the interfacial interaction between paper sludge and PP/EPDM EPDM matrix. The addition of a silane coupling agent also increased the crystallinity of PP and thermal stability of PP/EPDM/PS composites.

Keywords: composites, ethylene propylene diene terpolymer, paper sludge, polypropylene, silane coupling agent

INTRODUCTION

In recent years, the use of coupling agents in the development of particle-filled thermoplastic composites has attracted the attention of polymer researchers. However, the understanding of the role of

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Address correspondence to Professor Hanafi Ismail, Division of Polymer Engineering, School of Materials and Mineral Resources Engineering, University Sains Malaysia, 14300 Nibong Tebal, Penang, Malaysia. E-mail: hanafi@eng.usm.my

coupling agent in enhancing certain mechanical properties of particulate-filled thermoplastic is far from complete. Because of the multiphase nature of particulate-filled thermoplastic composite, the use of coupling agent requires an understanding of the chemical and physical phenomena occurring at all interfaces [1].

Compounding of cellulosic fillers with thermoplastic or thermoplastic elastomer (TPEs) increases the stiffness of the composites but tends to reduce strength [2–4]. The poor strength properties result from a lack of adhesion between the hydrophobic polymer and hydrophilic filler. Coupling agents or compatibilizers have been used to improve dispersion, adhesion, and compatibility for systems containing a hydrophilic cellulose and hydrophobic polymer. The agents modified the interface by interacting with both the fiber and the polymer, thus forming a link between the components [5–6]. Ismail et al. [7] reported that the incorporation of silane coupling agent, 3-aminopropyl triethoxysilane (3-APE), improved the tensile modulus, tensile strength, and stress at yield of white rice husk ash-polypropylene/natural rubber composites. Rozman et al. [8] also found that incorporation of two types of coupling agents, 3-(trimethoxysilyl) propylmethacrylate (TPM) and 3-aminopropyl triethoxysilane (APE), improved the mechanical properties of the rubberwood–high density polyethylene composites.

In recent years, the use of paper sludge as a filler in thermoplastic was increased [9–11]. The main problem of preparation of paper sludge–thermoplastics elastomer composites is the incompatibility of hydrophilic paper sludge and hydrophobic thermoplastics elastomer matrix, which yields composites of poor properties. In the present authors' previous work [12] they reported the effects of compatibilizer (MAPP) and coupling agent (LICA) on the properties of paper sludge–filled polypropylene (PP)/ethylene propylene diene terpolymer composites. The present study investigates the effect of a coupling agent, 3-aminopropyl triethoxysilane and filler loading on the mechanical properties, water absorption, morphology, Fourier Transform Infrared (FTIR), and thermal properties of paper sludge–filled polypropylene (PP)/ethylene propylene diene terpolymer composites.

EXPERIMENTAL

Materials

The polypropylene homopolymer used in this study was of injection molding grade, from Titan PP polymers (M) Sdn Bhd, Johor, Malaysia (code 6331) with MFI value of 14.0 g/10 min at 230°C. Ethylene

TABLE 1 Formulation of PP/EPDM/PS Composites with Different Filler Loading and 3-Aminopropyl Triethoxysilane (3-APE)

	Composites 1	Composites 2
Polypropylene (PP) (wt%)	50	50
Ethylene propylene diene terpolymer (wt%)	50	50
Paper sludge (PS) (wt%)	0, 15, 30, 45, 60	0, 15, 30, 45, 60
3-APE (wt%)	—	3

propylene diene terpolymer grade Mitsui EPT 3072E with tertiary component ENB and 74 Mooney viscosity ($ML_{1+4}100°C$) was obtained from Luxchem Trading Sdn Bhd., Selangor, Malaysia. Paper sludge (PS), a waste product from paper mills process, was obtained from Nibong Tebal Paper Mill Sdn Bhd, Penang, Malaysia. The PS was dried in a vacuum oven at 80°C for 24 h to free it from moisture and then ground to a powder. An Endecotts sieve was used to obtain an average filler size of 63 μm (density, 2.2 g/cm^3). A coupling agent, 3-aminopropyltriethoxysilane (3-APE), was supplied by Bumi Sains, Selangor, Malaysia. The formulation of PP/EPDM/PS composites with and without coupling agent used in this study is shown in Table 1. Table 2 shows the results of semi-quantitative analysis of paper sludge used in this study.

TABLE 2 Semi Quantitative Analysis of Paper Sludge Using X-Ray Flourescene Spectrometer Rigaku RIX 3000

Component	Wt (%)
Na_2O	0.057
MgO	3.0
Al_2O_3	7.1
SiO_2	10.0
P_2O_5	0.065
SO_3	0.14
Cl_2O	0.19
K_2O	0.035
CaO	20.0
TiO_2	0.11
MnO	0.018
Fe_2O_3	0.19
ZnO	0.017
SrO	0.011
LOI (Organic)	59.0

Filler Treatment

The coupling agent 3-APE was delivered in liquid form. Before application, it was diluted in ethanol to make 20% solution. The amount of 3-APE used was 3% by weight of filler. The filler was charged into a bench-top tumbler mixer and the solution was added slowly to ensure uniform distribution of 3-APE. After completion of the silane addition, the filler was continuously mixed for another 30 min. The treated filer was then dried at 100°C for about 5 h to allow complete evaporation of the ethanol.

Mixing Procedure

Composites were prepared in a Haake Reomix PolyDrive R 600/610. Mixing was done at 180°C and 50 rpm. EPDM was first charged to start the melt mixing. After 3 min the filler and coupling agent were added followed by PP at the fifth minute. Mixing was continued for another 5 min. At the end of 10 min, the composites were taken out and sheeted through a laboratory mill at 2.0 mm nip setting. Sample of composites were compression molded in an electrically heated hydraulic press. Hot-press procedures involved preheating at 180°C for 6 min followed by compressing for 4 min at the same temperature and subsequent cooling under pressure for 4 min.

Measurement of Tensile Properties

Tensile tests were carried out according to ASTM D- 412 on an Instron 3366. 1 mm thick dumb bell specimens were cut from the molded sheets with a Wallace die cutter. A cross head speed of 50 mm/min was used and the test was performed at 25 ± 3°C.

Water Absorption Test

The composite samples were immersed in distilled water at room temperature. The water absorption was determined by weighing the samples at regular intervals. A Mettler balance type AJ150 was used, with a precision of ± 1 mg. The percentage of water absorption, M_t, was calculated by

$$M_t = \frac{W_N - W_d}{W_d} \times 100\% \qquad (1)$$

where W_d and W_N are original dry weight and weight after exposure, respectively.

Morphology Study

Studies on the morphology of the tensile fracture surface of the composites were carried out using a scanning electron microscope (SEM) model Leica Cambridge S-360. The fracture ends of specimens were mounted on aluminium stubs and sputter coated with a thin layer of gold to avoid electrostatic charging during examination.

Fourier Transform Infrared Spectroscopy Analysis

FT-IR spectroscopic analysis of the paper sludge and composites was carried out in a Perkin Elmer Spectrometer 2000 FT-IR. The paper sludge was dispersed in dry KBr powder and land ground to obtain fine particles. Both ATR (Attenuated Total Reflectance) technique and KBr pellet technique were applied. Scanned range was 400–4000 cm^{-1}.

Thermogravimetry Analysis

Thermogravimetry analysis of the composites was carried out with a Perkin Elmer Pyris 6 TGA analyzer. The samples, weighting about 15–25 mg, were scanned from 50 to 600°C using a nitrogen air flow of 50 ml/min and a heating rate of 20°C/min. The sample size was kept nearly the same for all tests.

Differential Scanning Calorimetry

Thermal analysis measurements of selected systems were performed using a Perkin Elmer DSC-7 analyzer. Samples of about 10–25 mg were heated from 20 to 220°C using a nitrogen air flow of 50 ml/min and the heating rate of 20°C/min. The melting and crystallization behavior of selected composites were also determined using a Perkin Elmer DSC-7. The crystallinity (X_{com}) of composites was calculated using the following relationship:

$$X_{com}(\% \text{ crystallinity}) = \frac{\Delta H_f}{\Delta H_f^o} \times 100 \tag{2}$$

where ΔH_f and ΔH_f^o are enthalpy of fusion of the system and enthalpy of fusion of perfectly (100%) crystalline PP, respectively. For ΔH_f^o (PP) a value of 209 J/g was used for 100% crystalline PP homopolymer [9]. X_{com}, which is calculated using this equation, gives only the overall crystallinity of the composites based on the total weight of composites including non-crystalline fractions, and it is not the true crystallinity of the PP phase. The value of crystallinity for PP phase (X_{pp}) of the PP fraction was normalized using Eq. 3 as follows [10]:

$$X_{pp} = \frac{(X_{com})}{Wf_{pp}} \tag{3}$$

where Wf_{pp} is the weight fraction of PP in the composites.

RESULTS AND DISCUSSION

The torque-time curves of PP/EPDM composites for 0, 15, 30, 45, and 60 wt% loading of paper sludge are shown in Figure 1. EPDM was charged into the Haake at the beginning and rotors were started. A sharp increases in torque is obtained because of the resistance exerted by the EPDM against the rotors. The peak starts to decrease as the fusion of EPDM take place. A sharp decrease in the mixing torque was observed immediately after the addition of filler at the third minute due to the lubricant action of finer paper sludge. At the fifth minute, at which cold PP was charged into the mixer, a sharp peak is registered. This abrupt rise in torque represents the loading and fusion peak of PP. As fusion of PP is completed, the torque starts to

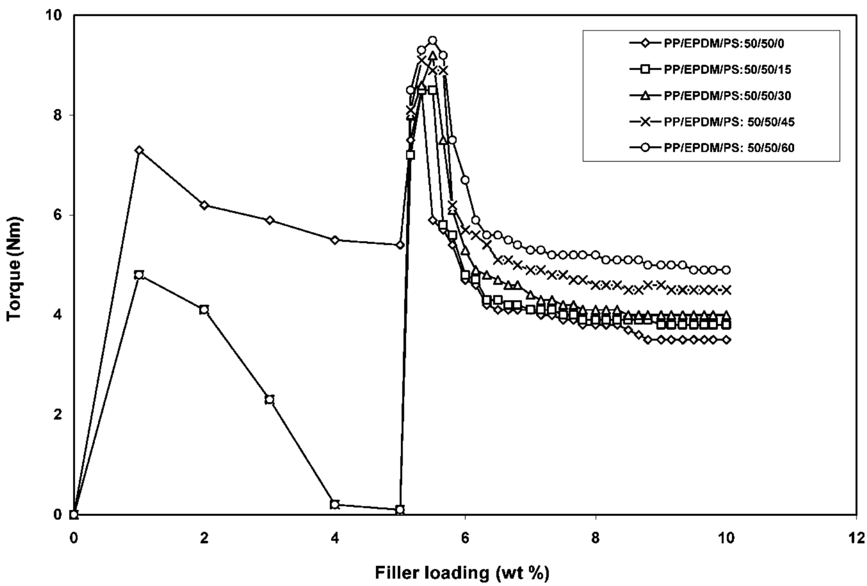

FIGURE 1 Torque versus time of PP/EPDM composites with different filler loading.

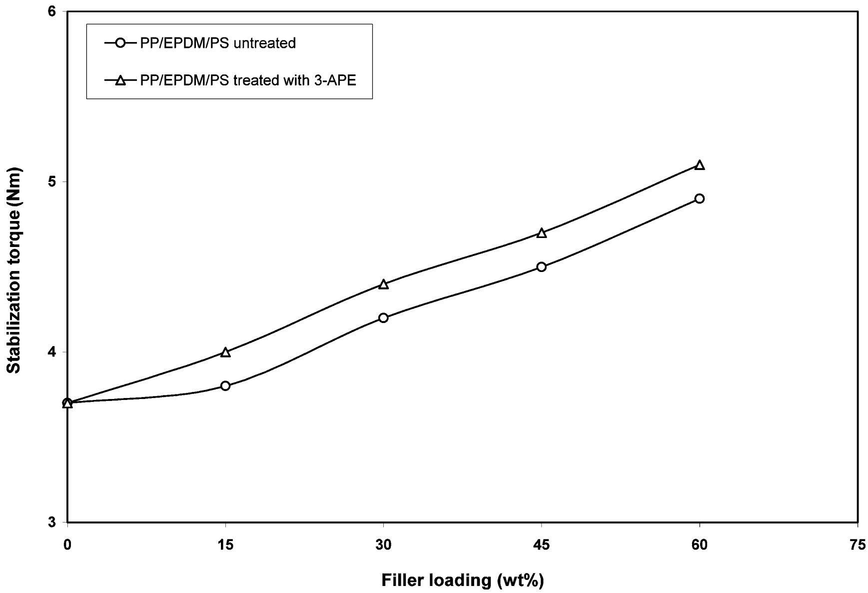

FIGURE 2 Stabilization torque versus filler loading of PP/EPDM/PS composites with and without 3-APE.

decrease gradually, due to a decrease in viscosity, to settle at a more stable value.

Figure 2 shows the relationship between stabilization torque and filler loading for PP/EPDM/PS composites with and without 3-APE. It can be seen that the stabilization torque increases with increasing filler content. This indicates that the increasing content of paper sludge has increased the melt viscosity of the composites. However, at a similar filler loading, PP/EPDM/PS composites with 3-APE exhibit higher stabilization (equilibrium) torque than PP/EPDM/PS composites without 3-APE. The stabilization torque indicates the stable melt viscosity and this results show that the addition of 3-APE in PP/EPDM/PS composites has resulted in higher mixing energy.

Figure 3 shows the effect of filler loading on tensile strength of paper sludge–filled PP/EPDM composites with and without 3-APE. It can be seen that the tensile strength decreases with increasing filler loading. In authors' previous work [11–12], they have reported that paper sludge consists of various shapes and forms such as fibers and particulates. For irregular shape fillers the tensile strength of the composites decreases due to inability of the filler to support the stress transferred from the matrix. It can be seen in Figure 3 that at a

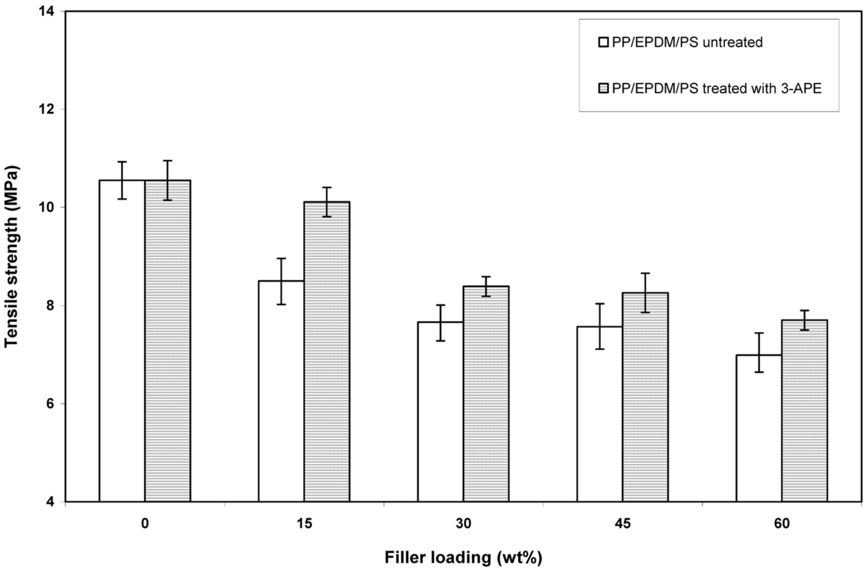

FIGURE 3 Effect of filler loading on tensile strength of PP/EPDM/PS composites with and without 3-APE.

similar filler loading, PP/EPDM/PS composites with 3-APE exhibit higher tensile strength than similar composites but without 3-APE. The use of a coupling agent was proven effective in enhancing the dispersion, adhesion, and compatibility of a system consisting of a hydrophilic filler and hydrophobic matrix. The interaction between the components occurred through modification of the polymer filler interphase [13]. According to Rozman et al. [14] the interaction is brought about by (i) hydrogen bonding between the silanol group and the hydroxyl group of cellulose surface and (ii) a van der walls type of interaction between the remaining chain of silane and PP/EPDM.

Figure 4 shows the effect of filler loading on elongation at break of paper sludge–filled PP/EPDM composites with and without 3-APE. Incorporation of fillers that have poor adhesion to the polymer matrix seems to cause interruption in the alignment process of the polymer chains. When filler loading increased, more weak interfacial regions between the filler surface and the PP/EPDM matrix are formed. Because cracks travel more easily through the weaker interfacial regions, the composites fracture at a lower elongation with increasing filler loading. Figure 4 also shows that composites with 3-APE exhibit lower elongation at break than the composites without 3-APE. The modification with 3-APE has increased the tensile strength of

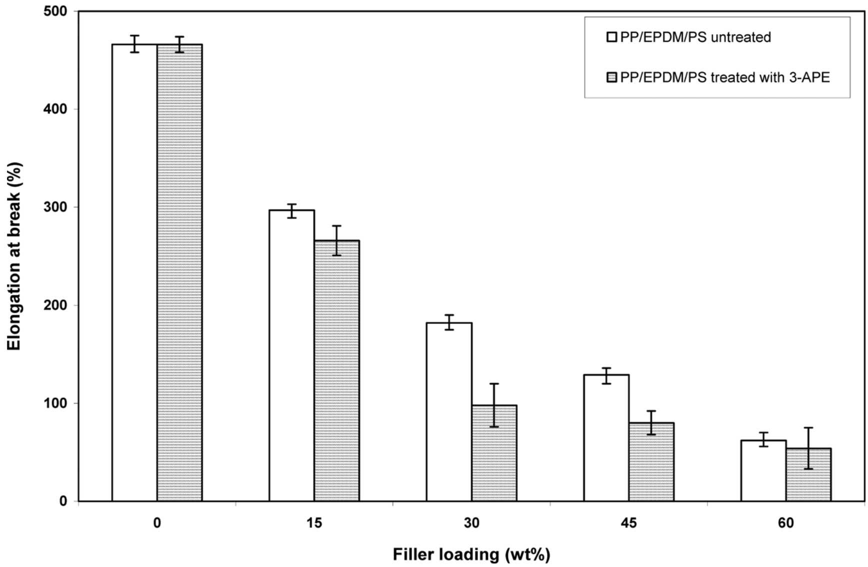

FIGURE 4 Effect filler loading on elongation at break of PP/EPDM/PS composites with and without 3-APE.

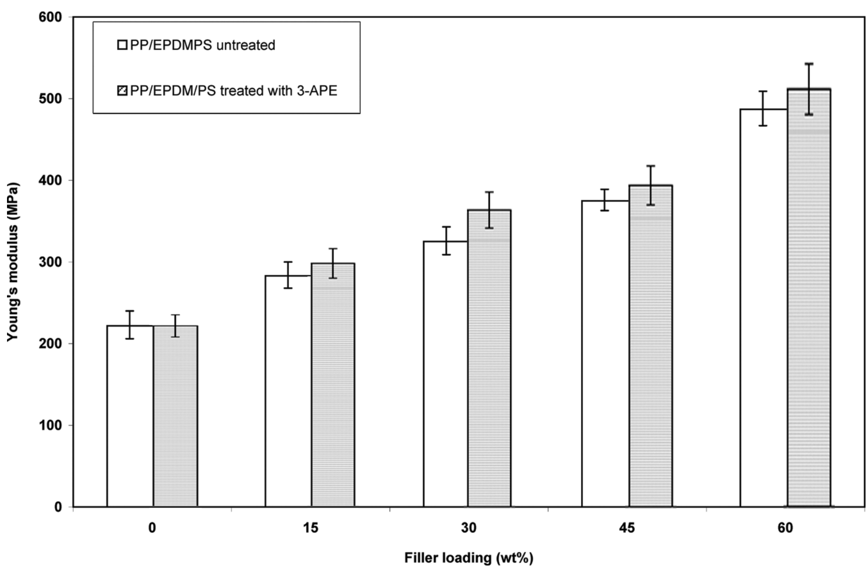

FIGURE 5 Effect filler loading on Young's modulus of PP/EPDM/PS composites with and without 3-APE.

composites with an enhancement in rigidity and reduction of the ductility of composites, which consequently lowered the elongation at break of the composites.

Figure 5 shows the effect of filler loading on Young's modulus of PP/EPDM/PS composites with and without 3-APE: the Young's modulus increases with increasing paper sludge loading. Young's modulus is an indication of the relative stiffness of composites [15]. The increase in Young's modulus with increase in filler loading is expected because the addition of filler increases the stiffness of the composites. At similar filler loadings, the Young's modulus of paper sludge–filled PP/EPDM/PS composites with 3-APE is higher than PP/EPDM/PS composites without 3-APE. This result indicates that the presence of 3-APE has caused a significant improvement in the filler-matrix interfacial bonding.

Figure 6 shows the relationship between water absorption and filler loading of paper sludge–filled PP/EPDM/PS with 3-APE composites. All composites show a similar pattern of water absorption (except for neat PP/EPDM): that is, initial sharp water uptake followed by gradual increase until equilibrium water content was achieved at about 50 days. However, composites with higher sludge content show more water absorption. As shown in Table 2, paper sludge consists 59% of

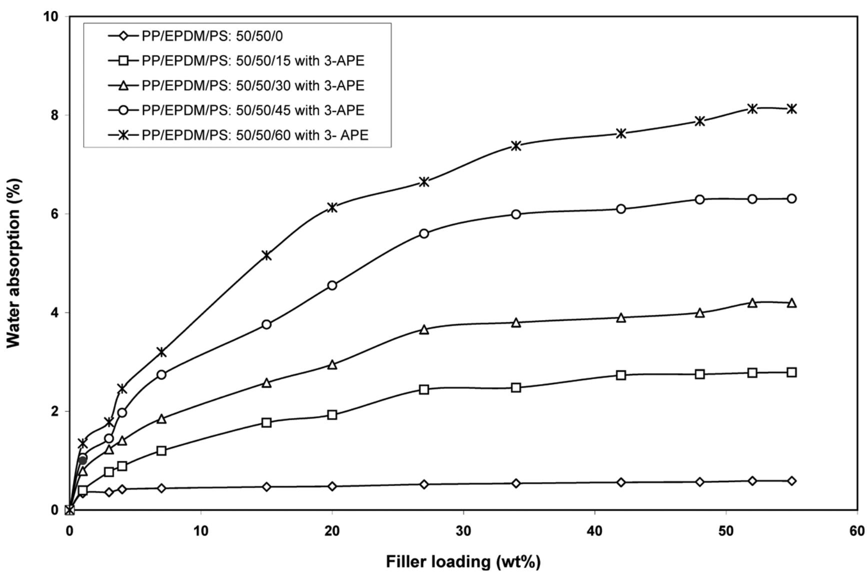

FIGURE 6 Percentage of water absorption versus time of PP/EPDM/PS composites with different filler loading.

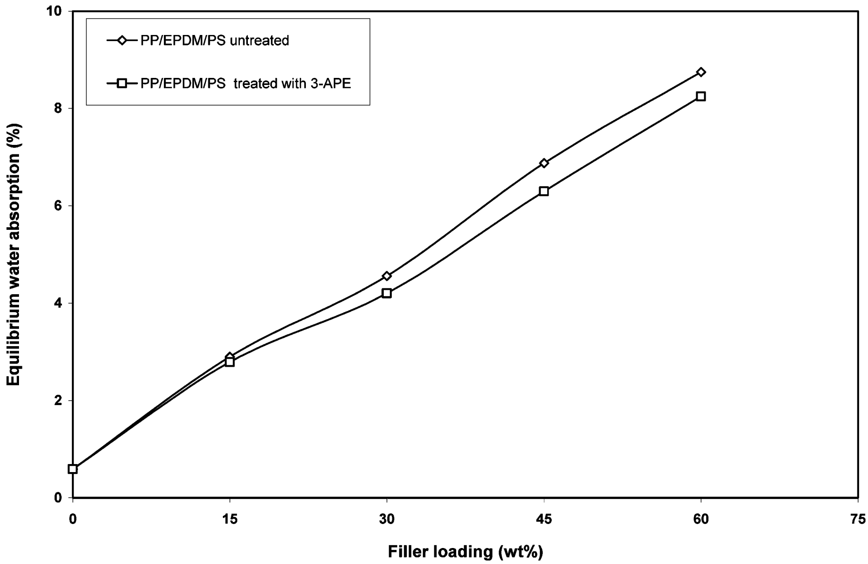

FIGURE 7 Percentage of equilibrium swelling versus filler loading of PP/EPDM/PS composites with and without 3-APE.

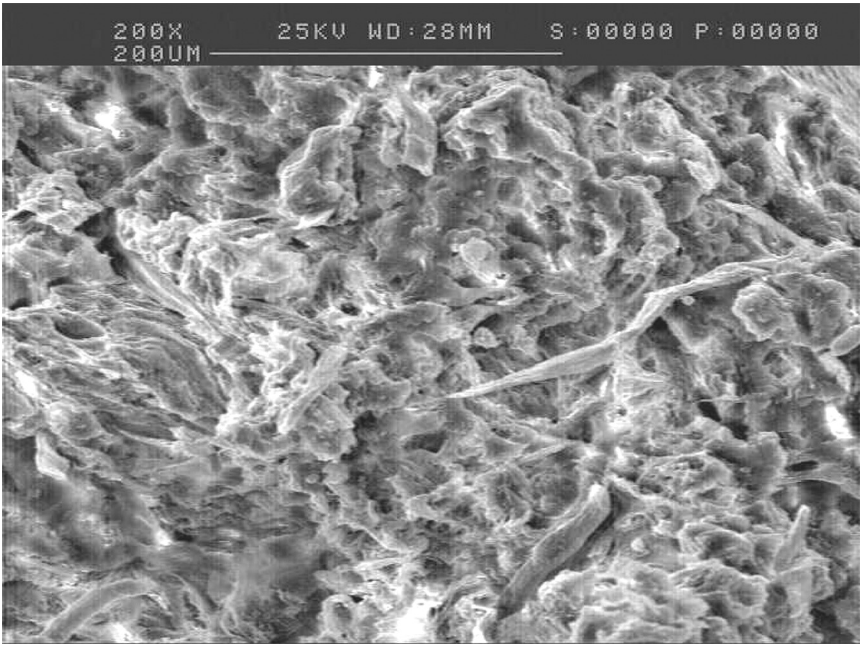

FIGURE 8 Scanning electron micrograph of tensile fracture surface of PP/EPDM/PS composite (30 wt%) at magnification 200×.

organic components (cellulose, hemicellulose, and lignin) and 10% of silica. As filler content increases the number of hydrogen bonds between organic components and silica with water molecules increases.

Figure 7 shows the variation of equilibrium swelling at 55 days immersion in water with filler loading of paper sludge filled PP/EPDM composites with and without 3-APE. It can be seen that at a similar filler loading, incorporation of 3-APE has reduced the amount of water absorption of the composites. The lower water absorption of the paper sludge–filled PP/EPDM composite treated with 3-APE may be attributed to the 3-APE consummating some of the hydroxyl groups of the PS.

Figures 8 and 9 show the tensile fracture surface of paper sludge–filled PP/EPDM composites at 30% and 60% weight of filler. It can be seen that the polar characteristic of paper sludge is not capable of forming a good filler-matrix interaction with non-polar PP/EPDM composites. However, in the presence of 3-APE, there is an evidence of improvement in interfacial bonding between the filler and

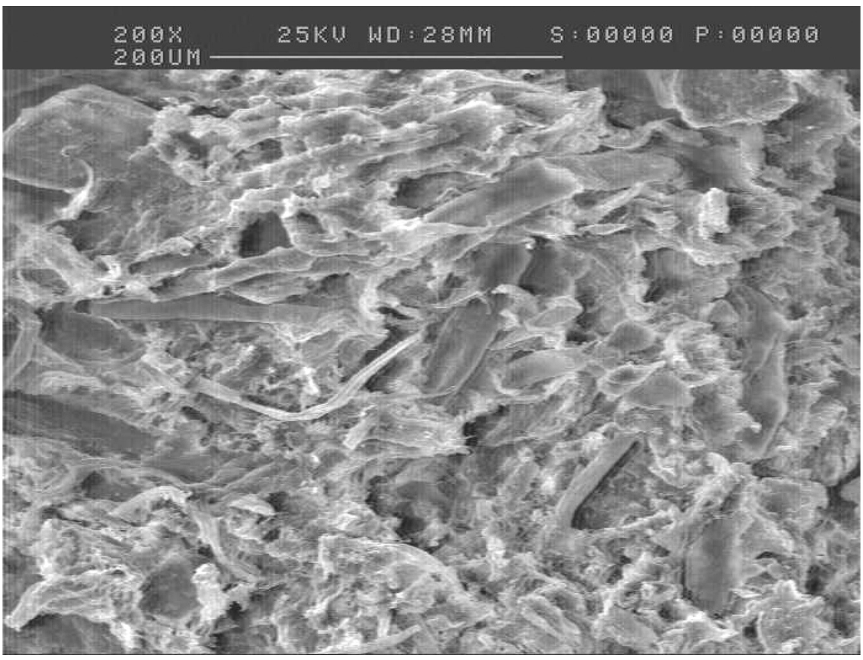

FIGURE 9 Scanning electron micrograph of tensile fracture surface of PP/EPDM/PS composite (60 wt%) at magnification 200×.

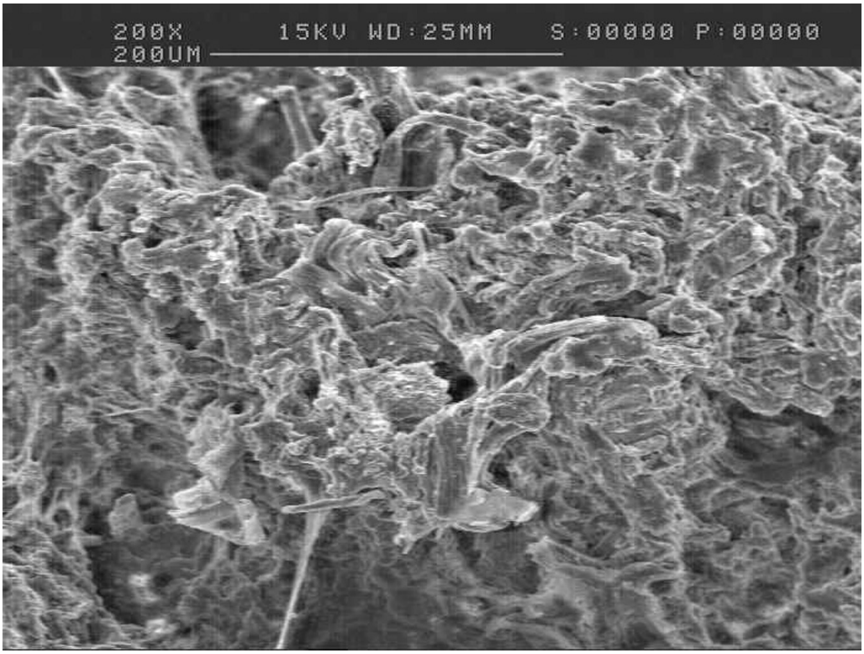

FIGURE 10 Scanning electron micrograph of tensile fracture surface of PP/EPDM/PS composite with 3-APE (30 wt%) at magnification 200×.

PP/EPDM composites, as can be seen in Figures 10 and 11. Figures 12 and 13 show the fillers were coated with PP/EPDM matrix and better adhesion occurred between filler and PP/EPDM matrix.

Figure 14 shows the FTIR spectra comparison of PP/EPDM/PS composites with and without 3-APE at 30 wt% of paper sludge. It can be seen that the PP/EPDM/PS composites with 3-APE exhibit no stretching vibration of –OH at 3439 cm^{-1}, compared to PP/EPDM/PS composites without 3-APE, which is evidence for the interaction between the hydroxyl group in paper sludge and the polar group of the silane coupling agent. Both curves exhibit the olefinic C–H stretching frequency around 2900–3000 cm^{-1}. The introduction of 3-aminopropyl triethoxysilane into PP/EPDM/PS leads to the appearance of two new absorptions at 1730 cm^{-1} and 1635 cm^{-1}, corresponding to the vibration of the carbonyl group and double bond of 3-aminopropyl triethoxysilane, respectively. The characteristic band of PP/EPDM/PS composites with 3-APE at 1635 cm^{-1} is dye to the existence of carbonyl group (C=O). In other work [2], a band at 1635 cm^{-1} has been attributed

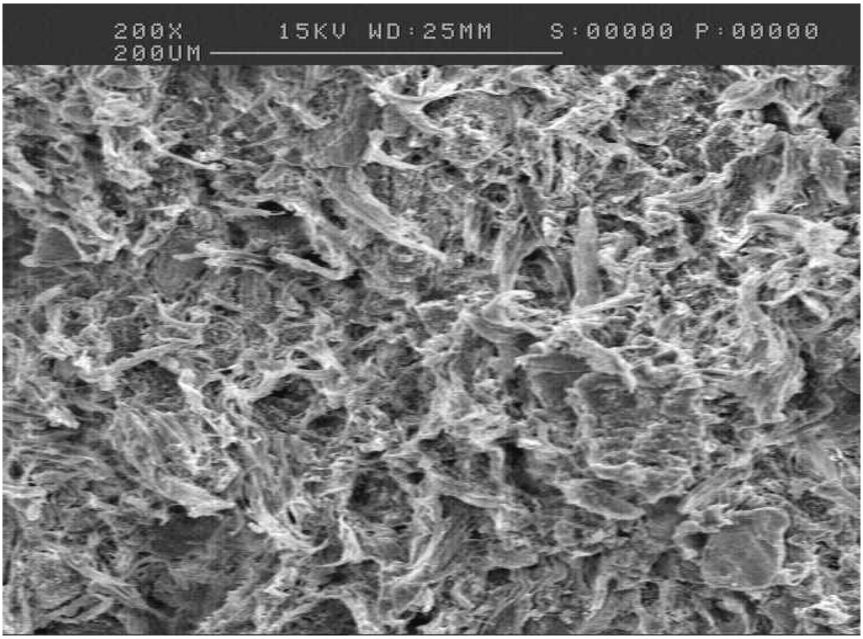

FIGURE 11 Scanning electron micrograph of tensile fracture surface of PP/EPDM/PS composite with 3-APE (60 wt%) at magnification 200× .

to hydrogen bonding between carboxyl groups created during the processing of PP/EPDM/PS composites that can react with the amino functionality of 3-aminopropyl triethoxysilane (Figure 15 (1)). The double band at 1730 cm^{-1} can be related to the symmetric and antisymmetric C=O vibration of amide. This hints at the possibility that the secondary amide enters into a reaction with another acid group forming imides (Figure 15 (2)). The formation of hydrogen bonding (Figures 15 (1) and (2)) in PP/EPDM/PS composites with 3-APE improved the mechanical properties due to grafting of 3-APE with PP/EPDM/PS in the composites. The schematic reaction between paper sludge with 3-APE in PP/EPDM/PS composites is shown in Figure 16.

Thermal degradation curves of PP/EPDM, PP/EPDM/PS composites with and without 3-APE at different filler loading are shown in Figure 17. It can be seen that the initial decomposition temperature of composites reduces with increasing filler loading. Table 3 shows the percentage weight loss of PP/EPDM, PP/EPDM/PS composites with and without 3-APE at different temperatures and filler loading. It can be seen that the total weight loss of PP/EPDM/PS composites

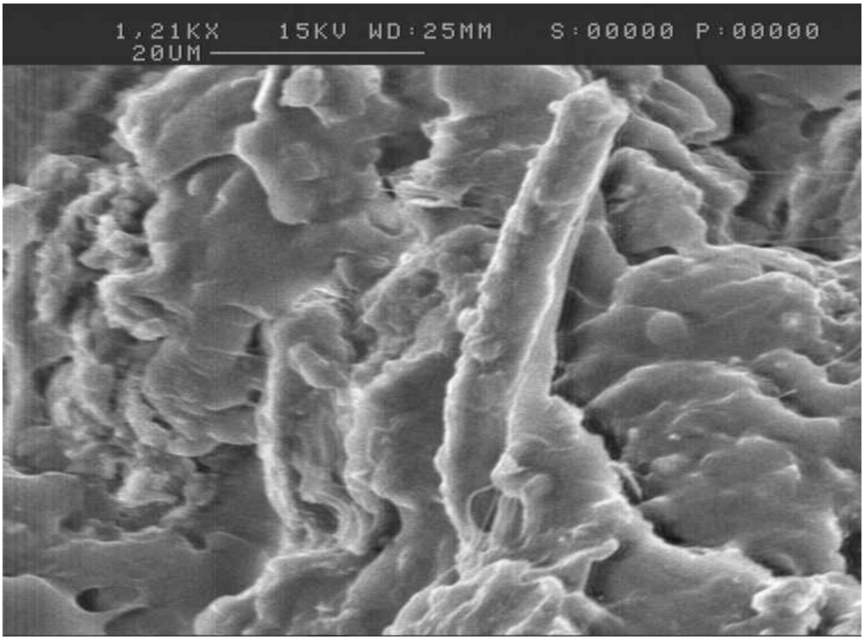

FIGURE 12 Scanning electron micrograph of tensile fracture surface of PP/EPDM/PS composite with 3-APE (30 wt%) at magnification 200×.

with 3-APE is lower than PP/EPDM/PS composites without 3-APE. This indicates that the presence of 3-APE has increased the thermal stability of PP/EPDM/PS composites.

Figure 18 shows the DSC curve of PP/EPDM/PS composites with and without 3-APE at different filler loading. Table 4 indicates the value of melting temperature (Tm), heat of fusion of composites ($\Delta H_{f(com)}$), crystallinity of composites (X_{com}), and crystallinity of PP (X_{pp}) for PP/EPDM/PS composites with and without 3-APE. The results show that the percentage of crystallinity of both composites changes with filler loading. It can be seen that the value of $\Delta H_{f(com)}$ and X_{com} decrease with increasing filler loading. This is due to decreasing PP contents at higher filler loading. The addition of filler results in an increase in X_{pp}. This observation was due to the nucleating ability of fillers for the crystallization of PP. From Table 4, at similar filler loading PP/EPDM/PS composites with 3-APE exhibit higher X_{com} and X_{pp} than PP/EPDM/PS without 3-APE. This might be due to the better nucleation effect of paper sludge in the presence of 3-APE. However, the presence 3-APE has reduced the melting temperature of the composites.

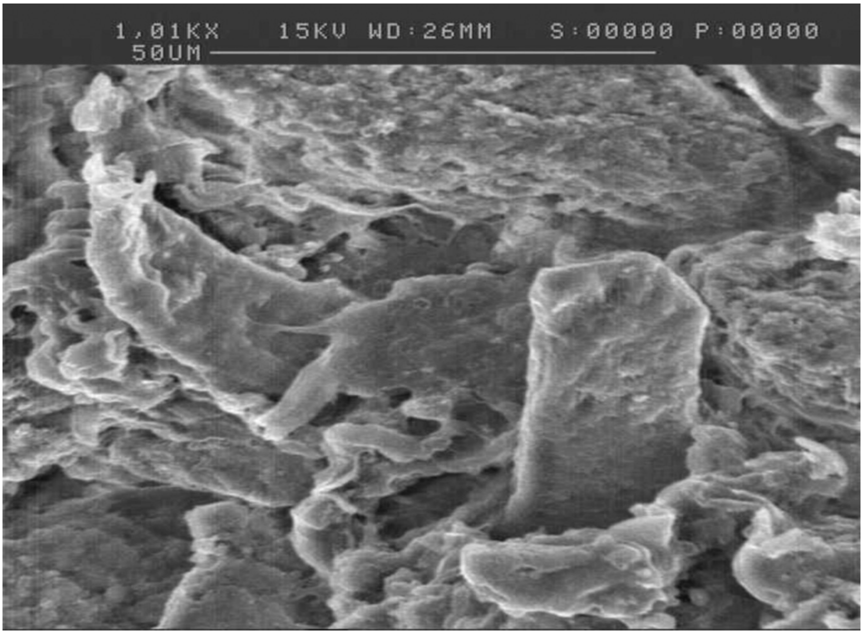

FIGURE 13 Scanning electron micrograph of tensile fracture surface of PP/EPDM/PS composite with 3-APE (60 wt%) at magnification 200× .

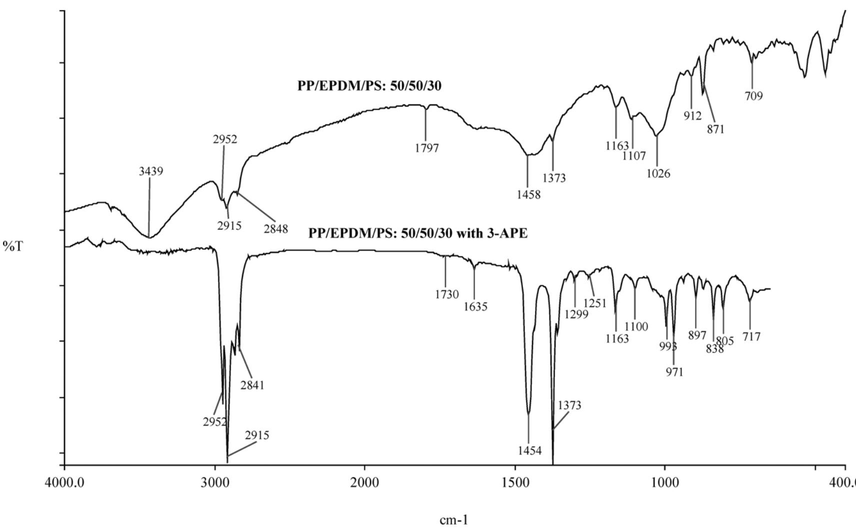

FIGURE 14 FTIR spectrum of PP/EPDM/PS composites with and without 3-APE.

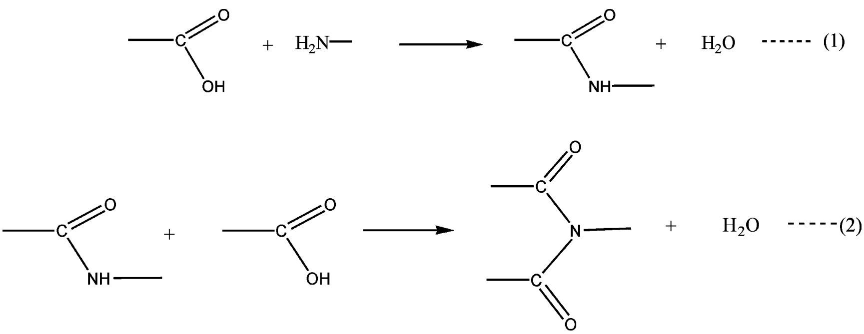

FIGURE 15 Schematic reaction of 3-APE with PP/EPDM/PS composites.

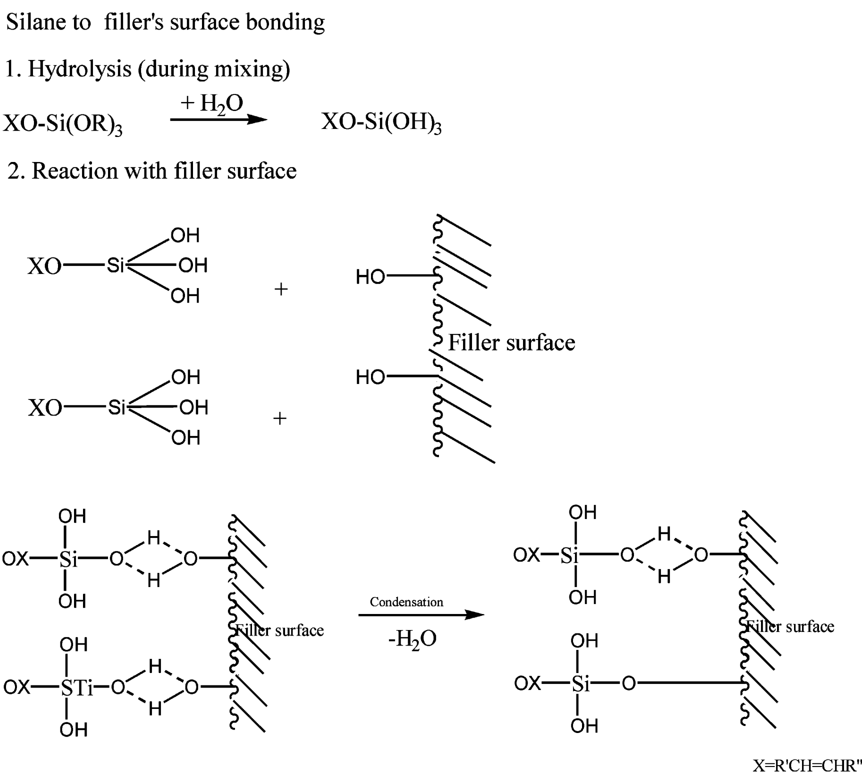

FIGURE 16 Schematic reaction between paper sludge with 3-APE in PP/EPDM/PS composites.

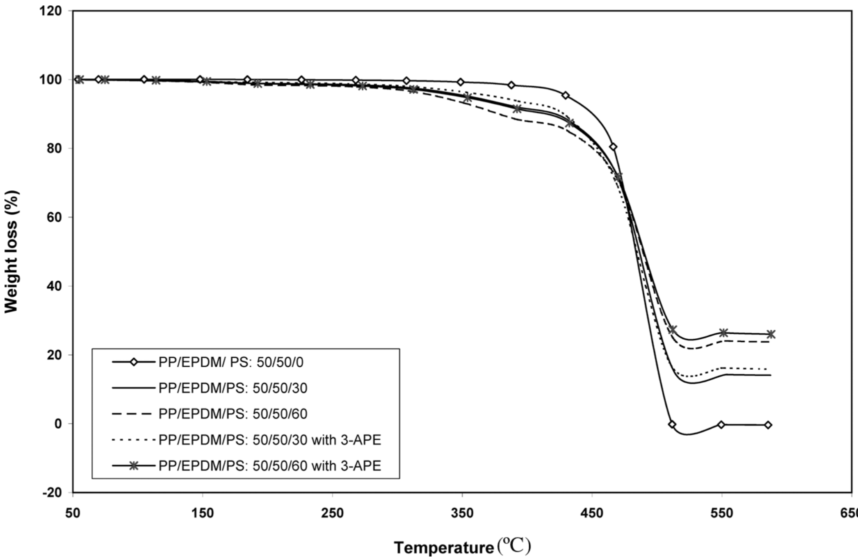

FIGURE 17 Comparison of thermogravimetric analysis curves of PP/EPDM/PS composites with and without 3-APE.

TABLE 3 Percentage Weight Loss of PP/EPDM and PP/EPDM/PS Composites with and without 3-APE

	Weight loss (%)				
Temperature (°C)	PP/EPDM: 50/50	PP/EPDM/ PS: 50/50/30	PP/EPDM/ PS: 50/50/60	PP/EPDM/ PS: 50/50/30 with 3-APE	PP/EPDM/PS: 50/50/60 with 3-APE
100	0.05	0.18	0.25	0.05	0.10
150	0.031	0.41	0.47	0.35	0.40
200	0.052	0.47	0.81	0.31	0.69
250	0.093	0.22	0.3	0.25	0.35
300	0.188	0.73	1.07	0.66	0.86
350	0.450	2.43	3.7	1.70	2.48
400	1.283	3.82	5.36	2.13	4.06
450	7.341	8.22	7.09	9.61	7.68
500	82.94	60.08	49.78	62.62	52.16
550	7.57	6.01	6.37	3.82	4.73
Total weight loss	100	82.60	75.20	81.50	73.51

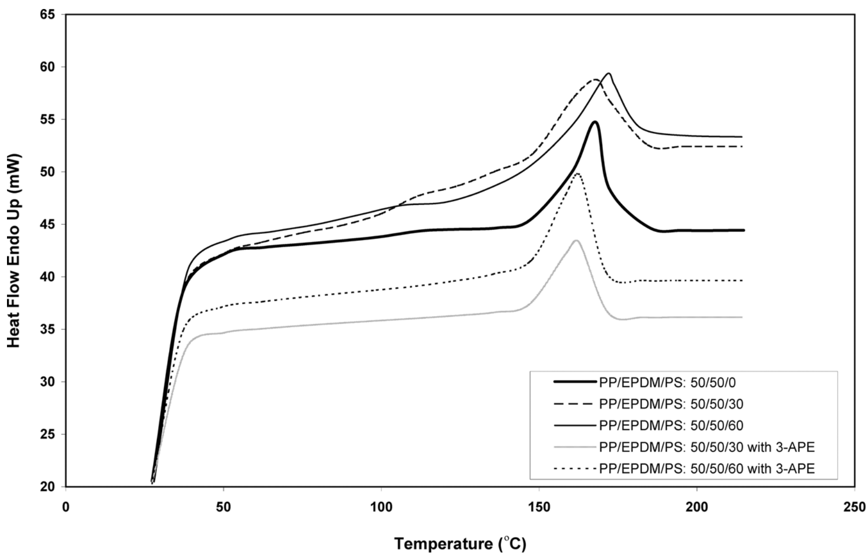

FIGURE 18 Comparison of differential scanning calorimetric curve of PP/EPDM/PS composites with and without 3-APE.

TABLE 4 The Thermal Parameter DSC of PP/EPDM and PP/EPDM/PS Composites with and without 3-APE

Composites	Melting temperature T_m (°C)	$\Delta H_{f(com)}$ J/g	X_{com} (% crystallinity)	X_{pp} (%)
PP/EPDM: 50/50	167.1	40.07	19.2	38.4
PP/EPDM/PS: 50/50/30	167.3	36.45	17.4	45.1
PP/EPDM/PS: 50/50/60	167.5	35.35	16.9	54.1
PP/EPDM/PS: 50/50/30 with 3-APE	161.4	40.50	19.3	50.1
PP/EPDM/PS: 50/50/60 with 3-APE	161.7	39.17	18.7	59.8

CONCLUSION

The incorporation of 3-aminopropyl triethoxysilane (3-APE) has increased the stabilization (equilibrium) torque, tensile strength, and Young's modulus but reduced the elongation at break and water absorption. Scanning electron microscope studies indicate that the interfacial adhesion between paper sludge and PP/EPDM matrix is improved in the presence of 3-APE. The PP/EPDM/PS composites with 3-APE have higher thermal stability and crystallinity than PP/EPDM/PS composites without 3-APE.

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