Average Mixing Torque, Tensile and Impact Properties, and Thermal Stability of Poly(vinyl chloride)/Sawdust Composites with Different Silane Coupling Agents

N. Sombatsompop, K. Chaochanchaikul

Polymer Processing and Flow (P-PROF) Group, School of Energy and Materials, King Mongkut's University of Technology Thonburi, Bangmod, Thungkru, Bangkok 10140, Thailand

ABSTRACT: Unique explanations are given to describe changes in the mixing torque and tensile and impact properties of poly(vinyl chloride) (PVC)/wood sawdust composites affected by various types and concentrations of silane coupling agents. Concentrations of 0.5–1.0 and 1.5 wt % coupling agent are recommended for the optimization of the tensile and impact properties of the composites, respectively. Changes in the tensile and impact properties of the composites with a low sawdust content were more sensitive to the addition of silane coupling agents than those with a high sawdust content. KBM603 was suitable for improving the tensile properties, whereas KBE603 is recommended for high impact resistance of the composites. The differences in the mechanical and thermal properties of the PVC/sawdust composites were dependent on the characteristics of the functional groups in the silane coupling agents used, such as hydrophilic level, number of functional groups, self-condensation reaction, and effectiveness of the hydrolysis reaction. Silane coupling agents above 1.0 wt % resulted in an increase in polyene sequences in the PVC structure. © 2005 Wiley Periodicals, Inc. J Appl Polym Sci 96: 213–221, 2005

Key words: poly(vinyl chloride) (PVC); surfaces; reinforcement; interfaces; extrusion

INTRODUCTION

The introduction of hydrophilic natural fibers in polymers results in heterogeneous composites whose properties are inferior due to incompatibility between strongly polarized cellulose fibers and hydrophobic polymers, which leads to poor interfacial bonds.[1] Current concepts on methods applied to improve the fiber–matrix interfacial adhesion include molecular chain entanglements, good mechanical contact, the matching of surface tensions, and the formation of chemical and physical bonds through coupling agents.[2,3] *Coupling agents* are primarily defined as materials that enhance the adhesive bonds of dissimilar surfaces.[4] Coupling agents are usually used for eliminating weak boundary layers, developing a highly crosslinked interphase region, improving the wetting between the polymer and fiber substrate, or altering the acidity of the fiber substrate surface. Common coupling agents include tetrafunctional organometallic compounds based on silicon, zirconate, or titanate coupling agents.[1] The mechanism of bonding by coupling agents in polymer–fiber composites appears to be very complex and is still not fully understood, as different polymer systems and different coupling agents tend to show different types of interfacial adhesion.[4]

Composite articles manufactured from blends of thermoplastics and natural fibers have increasingly attracted a number of researchers and manufacturers because of cost savings, good mechanical properties, better dimensional stability, and environment issues.[5] Thermoplastics commonly used include polyethylene,[6,7] polypropylene,[8,9] poly(vinyl chloride) (PVC),[5,10–12] acrylonitrile–butadiene–styrene,[13] and rubbers.[14] Due to incompatibility between the polymers and natural fibers, some surface modifications, usually accomplished by the use of coupling agents, are required for the improvement of the service performances of the composite products. Surface treatment of natural fibers in polymer–natural fiber composites are still one of the most interesting subjects for discussion. Matuana et al.[10] examined the effect of chemical surface treatments on the interfacial properties and mechanical performance of plasticized PVC/cellulosic fiber composites with various coupling agents, including γ-aminopropyltriethoxysilane, dichlorodiethylsilane, phthalic anhydride, and maleated polypropylene. The results show that the nature of the

wood–fiber surface was changed from hydrophilic to hydrophobic with the use of the coupling agents. γ-Aminopropyltriethoxysilane was the most suitable coupling agent as an adhesion promoter for the PVC/wood fiber composites because aminosilane-treated fibers could react with PVC to form chemical bonds, which then significantly improved the tensile strength of the composites.[11] Maldas et al.[12] used poly[methylene(polyphenol–isocyanate)] as a coupling agent in PVC–wood fiber composites and found that the optimum coupling agent concentration was 10 wt %. The molecular weight and chemical composition of silane coupling agents have been found to influence the properties of polymer–fiber composites.[13,14] Chiang and Hu[13] suggested that with high-molecular-weight coupling agents, one could improve the impact and elongation resistance by lowering the glass-transition temperature of the composites. Brinke et al.[14] used different bifunctional silane coupling agents in silica–rubber composites and suggested that changes in the composite temperature during material compounding could be the main factor influencing the reaction of the coupling agents and, thus, the properties of the composite.

The mechanical strength of PVC/fiber composites could be lower than the neat PVC if an appropriate coupling agent is not used due to poor interfacial bonding between natural fibers and PVC, which is caused by a number of things, including homogeneity level,[15] moisture in the fiber,[16] and thermal and mechanical degradations of PVC during processing.[17] According to comprehensive literature,[2–5,10–12] the mechanisms of surface treatments in PVC/cellulosic fiber composites are highly complex, and so far they are not well-understood, especially when different chemical structures of the coupling agents are involved. Previous studies[15,16] have examined the effect of the untreated sawdust content and moisture content on the properties of PVC/sawdust composites, and the results suggest that the changes in the composite properties are dependent on dipole–dipole interactions between PVC and cellulosic sawdust, a reformation of hydrogen bonding between moisture and the fiber, interfacial defects in the PVC matrix, fiber swelling, and/or moisture evaporation during the test.

In this study, which was extended from previous studies,[15,16] we aimed to examine the effect of the molecular structure of silane coupling agents, used as property promoters for PVC/wood sawdust composites, on average mixing torque, tensile and impact strengths, and thermal stability. The fractured surfaces of the composites from mechanical tests were also investigated. In this study, the silane coupling agents used were commercial grade and are widely used in vinyl composite materials. Also, they had different molecular weights and different numbers of func-

tional groups, which allowed us to investigate the effect of the physical and chemical molecular structures of the silane coupling agents on the properties of the PVC/wood sawdust composites. Unexpected results were found, and unique discussions are introduced.

EXPERIMENTAL

Raw materials

PVC was supplied by V. P. Plastics Product Co., Ltd. (Bangkok, Thailand) in the form of compounded PVC powder. There were two types of PVC compounds used in this work, suspension PVC (SIAMVIC 258RB) with a K value of 58 and emulsion PVC (SIAMVIC 374MB) with a K value of 74. The latter PVC compound was used as an impact modifier for the former PVC compound.

Sawdust particles were obtained from carpentry and woodworking processes and supplied by V. P. Plastics Products (1993) Co., Ltd. (Bangkok, Thailand). The average size of the sawdust particles used in this work was in the range 100–300 μm. The contents of wood sawdust particles added into the PVC compound were fixed at 9.1 wt % (for the low-sawdust composite) and 41.2 wt % (for the high-sawdust composite) PVC throughout this study.

Three silane coupling agents were used in this study, N-2(aminoethyl)3-aminopropylmethyl dimethoxysilane (KBM602), N-2(aminoethyl)3-aminopropyl trimethoxysilane (KBM603), and N-2(aminoethyl)3-aminopropyl triethoxysilane (KBE603), whose chemical structures and descriptions are listed in Table I. These coupling agents were supplied by Shin-Etsu Chemical Co., Ltd. (Tokyo). With these coupling agents, we could examine the influences of the chemical structure of the silane coupling agents on the tensile and impact properties of the PVC/sawdust composites.

Other additives [in parts per hundred (pph; SIAMVIC 258RB)] included a PVC (SIAMVIC 374MB, 4 pph) organic complex stabilizer (TS-DBL-Pb-Ba, 3.6 pph), an external lubricant (Finalux G-741, 0.6 pph), calcium carbonate (Omyacarb-2T, 9 pph), calcium stearate (0.3 pph), and acrylic-based processing aids (PA20, 8 pph); the details of these chemicals are found elsewhere.[15]

Surface treatments of the sawdust particles by silane coupling agents

The wood sawdust (SD) particles were carefully dried before use through heat treatment in an oven for 24 h at 80°C until the sawdust weight was constant to minimize the moisture content to a known value; the estimated moisture content was about 1%.[16] A mixture

TABLE I
Chemical Structures and Descriptions of the Silane Coupling Agents

Silane type	Chemical description	M_w	Chemical structure
KBM602	*N*-2(aminoethyl)-3-aminopropylenemethyl dimethoxysilane	206.4	CH_3O—Si—$CH_2CH_2CH_2$—N—$CH_2CH_2NH_2$ with CH_3 on top of Si, $O\|CH_3$ below Si, and H below N
KBM603	*N*-2(aminoethyl)-3-aminopropyltrimethoxysilane	222.4	CH_3O—Si—$CH_2CH_2CH_2$—N—$CH_2CH_2NH_2$ with OCH_3 on top of Si, $O\|CH_3$ below Si, and H below N
KBE603	N-2(aminoethyl)-3-aminopropyltriethoxysilane	264.5	CH_3CH_2O—Si—$CH_2CH_2CH_2$—N—$CH_2CH_2NH_2$ with OCH_2CH_3 on top of Si, $O\|CH_2CH_3$ below Si, and H below N

M_w = weight-average molecular weight.

of deionized water and 1% acetic acid was prepared, and the silane coupling agent was slowly dropped into the mixture until the required concentration was met. The silane concentration was varied from 0.0 to 2.0 wt % wood sawdust. The silane solution was then sprayed onto the sawdust particles, with a better mixing obtained with a high-speed mixer for 5 min. Thereafter, the mixtures were filtered for the removal of the solvent. The treated wood particles were then oven-dried for 6 h at 100°C until a constant weight was achieved.

Blending of PVC and silane-treated sawdust particles

PVC compounds were dry-blended with silane-treated sawdust particles with a high speed mixer for 5 min before they were melt-blended in a twin-screw extruder (Haake Polylab-Rheomex CTW 100P, Germany). The blending temperature profiles on the extruder were 165, 175, 175, and 185°C from hopper to die zones. The screw rotating speed was 40 rpm, and a slit die 18.2 $\times$ 2 x 16 mm^3 was used to produce slit extrudates, which then passed through a water bath for cooling. These composite extrudates were used for the evaluation of the composite properties.

Characterizations

Average mixing torque

The twin-screw extruder used for blending PVC and sawdust was used to generate the average mixing torque during the blending of PVC with the treated wood sawdust particles.

Tensile and impact strengths

The mechanical properties of the PVC/sawdust composites were assessed via tensile and impact properties. We performed the tensile tests on a Shimadzu tensile tester (Tokyo) at a crosshead speed of 5 mm/min, following test procedure ASTM D 638 (1990) Specimen Type I. Izod impact tests [ASTM D 256 (1990)] were performed on a Yasuda impact tester (Osaka, Japan) with the notched side facing the pendulum.

Thermal stability test

Polyene sequences were determined with attenuated total reflection Fourier transform infrared (FTIR) analysis to assess the extent of degradation and the structural changes of the PVC. This was carried out by the measurement of the reflectance difference percentage at the surface of the sample, which was prepared by a twin-screw extruder, as mentioned earlier. The dimensions of the samples were 13 $\times$ 64 $\times$ 2 mm. The tests were performed with a Nicolet (Madison, WI) Nexus 470 FTIR spectrometer to produce spectra of reflectance difference against wave numbers. In this study, the peak for polyene sequences at a wave number of 1584 cm^{-1} was of interest. We expressed the polyene indices as percentage reflectance (ΔR) by subtracting the percentage reflectance value of the peak of interest (%Rc=c) from that of the baseline (%$R_{baseline}$) at 100% reflectance, as shown in eq. (1). For polyene sequences, the weight percentage of PVC had to be taken into account because its content varied with the amount of sawdust wood added:
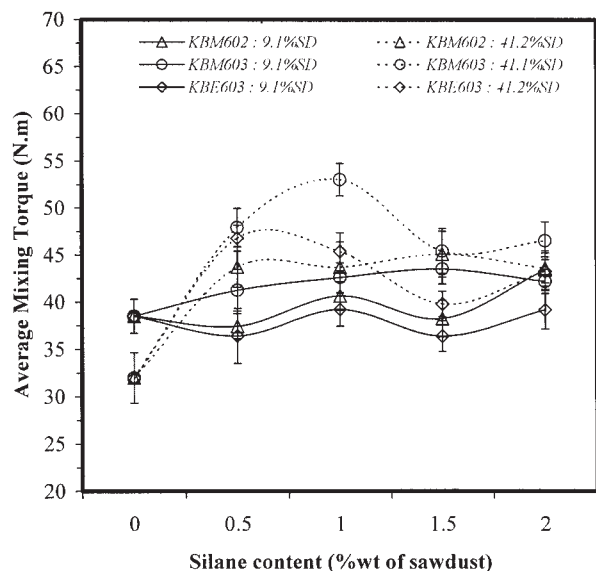
Figure 1 Effect of coupling agent type and content on the average mixing torque of the PVC/wood sawdust composites: (—) 9.1 and (· · ·) 41.2 wt % sawdust.

ΔR of the polyene sequence

$$= \frac{\%R_{baseline} - \%R_{C=C}}{PVC(wt \%)} \times 100 \quad (1)$$

The decomposition temperature was assessed with a TGA Instruments (Perkin-Elmer TGA7, Boston, MA). The degradation temperature results were taken when the onset of weight loss had occurred during the thermogravimetric analysis runs. A temperature range of 100–550°C with a heating rate of 5°C/min was used.

Scanning electron microscopy (SEM) investigations

Failure mechanisms were investigated with a Jeol (JSM-6301F) (Peabody, MA) SEM machine at a 15-kV accelerating voltage. The composite fracture surfaces for examination were obtained after 2-min immersion in liquid nitrogen. The details of the experimental procedure and sample preparations are described elsewhere.[15]

RESULTS AND DISCUSSION

Figure 1 shows the effect of types and concentrations of silane coupling agents on the average mixing torque of the PVC/sawdust composites. As shown, the composites with treated wood sawdust seemed to have higher average torque values than untreated ones. The average torque for the composites with 9.1 wt % sawdust content slightly increased with the addition of the silane coupling agent, whereas a pronounced change in the torque values was observed for the composites with 41.2 wt % sawdust particles. This was expected

because the greater the sawdust content was, the higher the amount of silane coupling agent was required and, thus, the more interfacial interactions there were between the sawdust and the coupling agents. For the composites with 41.2 wt % sawdust, the torque values increased to a maximum around 0.5–1.0 wt % coupling agent and then decreased for higher silane loadings. It seemed inappropriate at this (mixing) stage to thoroughly examine the differences in the torque values of the composites with different coupling agents because the chemical reactions between these coupling agents and the composites dynamically changed during mixing.

Figure 2 shows the effect of the silane coupling agents on the tensile modulus of the PVC/sawdust composites. In general, the tensile modulus increased to a maximum value around a silane coupling agent content of 0.5–1.0 wt % sawdust particles, depending on the type of silane coupling agent used. After that, the tensile modulus tended to decrease with coupling agent content. Similar behaviors were found for the tensile strength of the composites, whose results are shown in Figure 3. The optimum silane coupling agents around 0.5–1.0% to be added in the composites seemed to agree with a number of related works on vinyl composites.[18,19] It also seemed that the changing trends in the tensile modulus and tensile strength as a function of the concentration of silane coupling agents and the optimum silane concentration to be added into the composites were in line with those in the average mixing torque, as shown in Figure 1. The
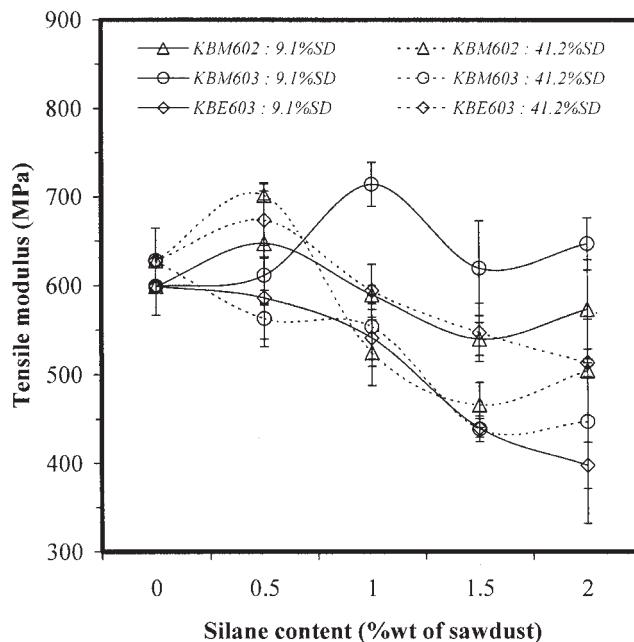

Figure 2 Effect of coupling agent type and content on the tensile modulus of the PVC/wood sawdust composites: (—) 9.1 and (· · ·) 41.2 wt % sawdust.
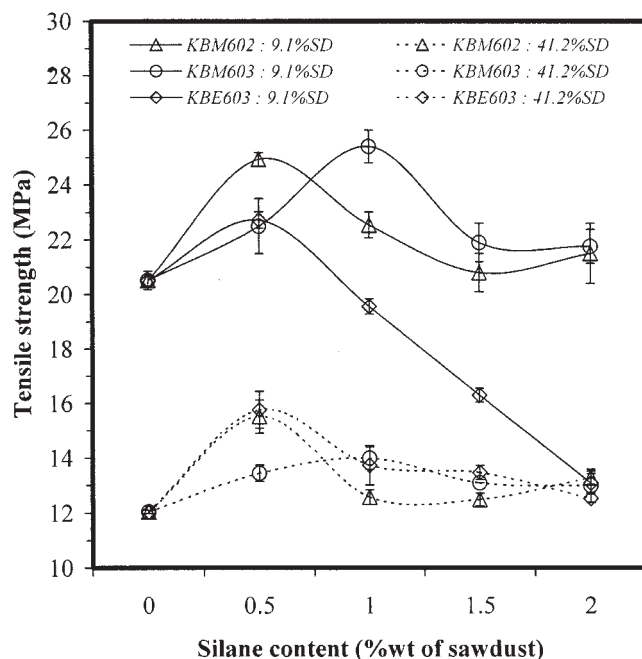
Figure 3 Effect of coupling agent type and content on the tensile strength of the PVC/wood sawdust composites: (—) 9.1 and (· · ·) 41.2 wt % sawdust.

optimum silane concentration (ca. 0.5–1.0 wt % sawdust), which gave the maximum mixing torque and tensile properties in the composites, indicated high interfacial interactions between the sawdust and the coupling agents. The increase in the tensile modulus and tensile strength at small coupling agent concentrations (0.0–1.0 wt %) was associated with ether linkages formed by the chemical reactions of the coupling agent and sawdust particles and N—C linkages between the amino groups in the coupling agent and the PVC; the descriptions of this mechanism can be found elsewhere.[10–12,18] The decrease in the tensile modulus and tensile strength at higher coupling agent concentrations may have been caused by a self-condensation reaction of the hydrolyzed or partially hydrolyzed coupling agents on the surfaces of the wood sawdust. The main product of the self-condensation reaction formed flexible polysiloxynol molecules (due to the presence of ether linkages) on the wood surfaces, which could have acted as a plasticizer between the wood sawdust and the PVC molecules.[13] This may eventually have caused a reduction in the tensile modulus and tensile strength of the composites because the tensile strength of a polymer is widely known to involve the ability for the polymer molecular chains to slip past each other; the easier the slippage of the molecular chains is, the lower the tensile strength will be. This explanation can be substantiated by the SEM micrographs in Figure 4, which show the fracture surfaces of the PVC/wood sawdust composites (9.1 wt %) at high [Fig. 4(a) for 0.5 wt % KBM602] and low

[Fig. 4(b) for 1.0 wt % KBM602] tensile strengths. As shown, the composite with KBM602 at 1.0 wt % exhibited some interfacial debondings (and voids) between the wood sawdust particles and the PVC matrix, which caused a reduction in the tensile strength.

Interestingly, the content of coupling agent that gave the maximum tensile modulus for each silane coupling agent was different; the composite with KBM603 had a relatively higher coupling agent content compared to KBM602 and KBE603. The following should be noted:

- At a low coupling agent content (0.5 wt %), both the tensile modulus and tensile strength of the composites with KBM603 and KBE603 were lower than those with KBM602.
- When KBM603 with KBM602 were compared at coupling agent contents higher than 0.5 wt %, the tensile modulus and tensile strength for the composite with KBM602 were lower than those with KBM603. The differences in the tensile properties of the composites with KBM602 and KBM603 were caused by the differences in the number of methoxy groups (see Table I). Because KBM602 had two hydroxyl groups, the possibility of the formation of ether linkages was more remote than for KBM603, as the hydroxyl groups may have been used up for the self-condensation reaction. For KBM603, because it had three hydroxyl groups, ether linkages between the coupling agent and the wood sawdust were still possible during the self-condensation reaction. This could have been why the tensile properties of the composites with KBM603 were higher.
- When KBM603 with KBE603 were compared at coupling agent contents higher than 0.5 wt %, we could see that the tensile modulus and tensile strength of the composites with KBM603 were much higher than those with KBE603; the properties of the composite with KBE603 progressively dropped with increasing coupling agent content. Because the chemical structures of these two coupling agents after the hydrolysis reaction were the same, the differences in the tensile properties of the composites with these two coupling agents had to be explained logically in terms of the effectiveness of the hydrolysis reactions of KBM603 and KBE603, with this referred to as the possibility of the elimination of methoxy and ethoxy groups in KBM603 and KBE603, respectively. That is, KBE603, which contained ethoxy functional groups, was more hydrophobic than KBM603, which contained methoxy functional groups.[20] Therefore, KBE603 was more difficult to hydrolyze to form hydroxyl groups; this view was also supported by Park et al.[21] This means that the coupling reaction between the sawdust and the PVC by KBE603 was not as complete as that by KBM603. In this respect, the unhydrolyzed KBE603 acted as a
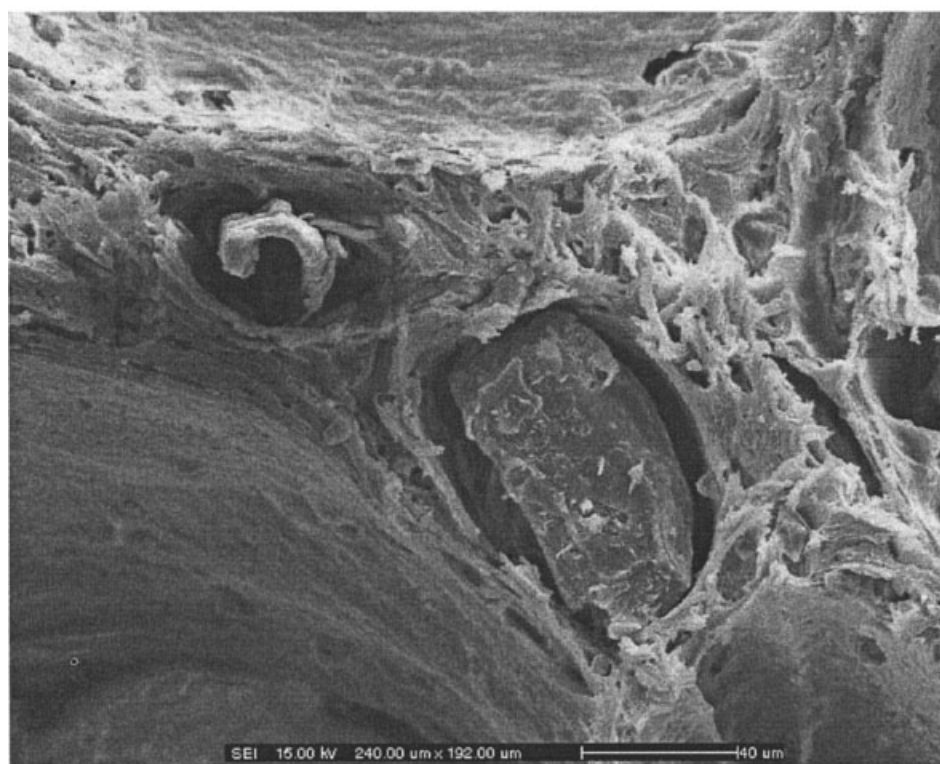
Figure 4 SEM micrographs of the PVC/wood sawdust composites: KBM602 at (a) 0.5 and (b) 1.0 wt %.

minor polymeric phase in the PVC/wood saw-dust composites. This could have been why the tensile properties of the composites with KBE603 dropped considerably, especially when the content of the KBE603 coupling agent was increased to 1.0–2.0 wt %.
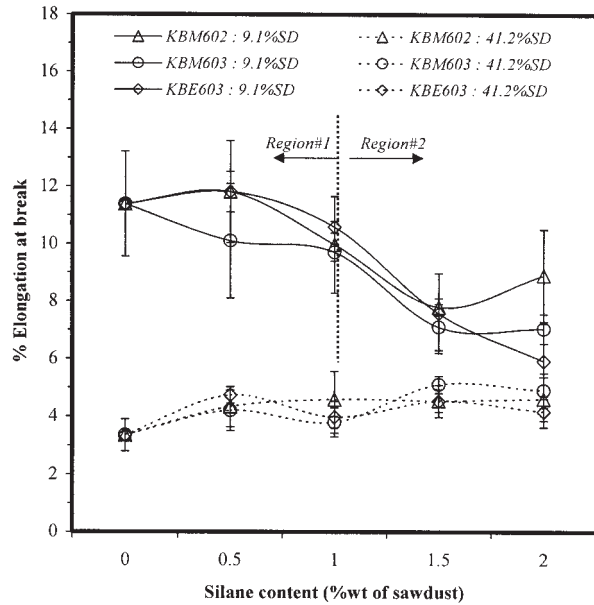
Figure 5 Influence of coupling agent type and concentration on the percentage elongation at break of the PVC/wood sawdust composites: (—) 9.1 and (· · ·) 41.2 wt % sawdust.

When we considered the effect of sawdust content, we found, as expected, that the composites with a higher sawdust content (41.2 wt %) exhibited a lower tensile strength than those with a lower sawdust content (9.1 wt %). This was caused by two things: (1) composites with higher sawdust content were likely to contain a greater moisture content because the wood sawdust was hydrophilic and the water molecules on the fiber surface could promote or accelerate the self-condensation reaction of the coupling agent,[18] and (2) increasing the wood sawdust content automatically increased the interfacial defects in the PVC matrix, which resulted in a decrease in composite homogeneity and, thus, caused poor stress transfer between the polymer matrix and the fiber.[15]

Figure 5 shows the effect of the coupling agent type and concentration on the elongation percentage at break of PVC/wood sawdust composites with 9.1 and 41.2 wt % wood sawdust. For the 9.1 wt % silane content, the elongation at break of the composites was stable at silane contents up to 1.0 wt % (Region 1) and then dropped at higher coupling agent loadings of 1.5–2.0 wt % (Region 2). The explanation for the decrease in the elongation at break is similar to those given for the tensile strength of the composites, as discussed earlier. For a silane content of 41.2 wt %, the changes in the percentage elongation at break of the composites were rather small, the differences being within the experimental errors (±5%).

Figure 6 illustrates the impact strength of the PVC/wood sawdust composites with 9.1 and 41.2 wt % sawdust. Generally, KBE603 was the most suitable for

obtaining composites with high impact resistance compared to KBM603 and KBM602. The optimum addition of KBM603 in the composites was 1.5 wt % for the optimization of the impact strength. In general, the impact strength of a polymeric fiber composite can be enhanced by two mechanisms: improving the interfacial bondings between phases of the polymer and the fiber and introducing a flexible and elastic material that can act as an impact modifier.[22] In relation to this study, the unhydrolyzed KBE603 in the composites, which contained ether linkages, acted as an impact modifier that improved the impact strength of the composites.[22] When we compared KBM602 and KBM603, we could see that the composites with KBM603 had a slightly higher impact strength than those with KBM602. This could be explained by the number of hydroxyl groups in the coupling agents. That is, KBM603 had more hydroxyl groups to form ether linkages with the sawdust, which led to improved interfacial bondings between the PVC and the wood sawdust. The composites with a higher sawdust content had a lower impact strength than those with a lower sawdust content, as expected.[15]

The extent of degradation of the PVC in the PVC/sawdust composites with different types and concentrations of silane coupling agents is shown in Figure 7. There were two regions to be considered in terms of the changes in conjugated double bond (polyene) contents in the PVC as a result of loading the coupling agents. At 0–1.0 wt % coupling agent (Region 1), the polyene content for the composites with KBM603 and
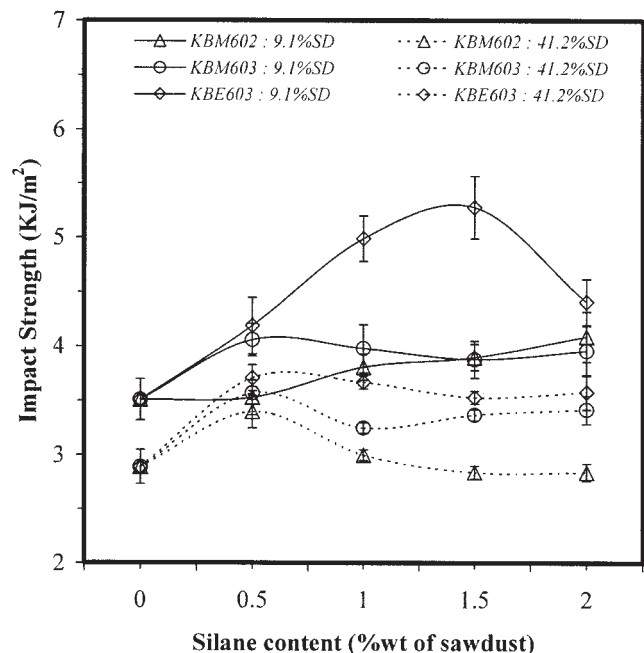

Figure 6 Plots of impact strength against the silane coupling agent content for the PVC/wood sawdust composites: (—) 9.1 and (· · ·) 41.2 wt % sawdust.
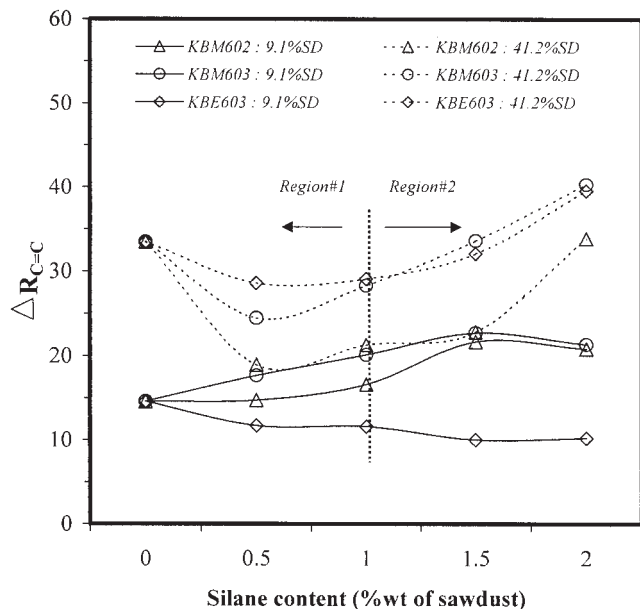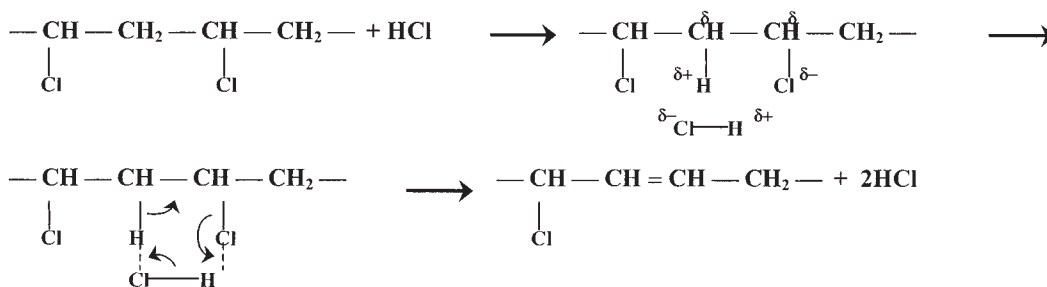
Figure 7 Changes in the polyene content of PVC in the PVC/wood sawdust composites for various types and concentrations of silane coupling agents.

KBE603 appeared to increase approximately 30%, whereas that with KBM602 remained more or less constant for the composites with 9.1 wt % sawdust. The polyene content decreased for the composites with 41.2 wt % sawdust. During thermal processing in the twin screw extruder, PVC encountered a dehydrochlorination reaction, which resulted in a release of HCl molecules, and the amino groups in the coupling agents may have reacted with the macromolecules of dehydrochlorinated PVC, and N—C linkages would have been formed, and this reaction would have prevented or delayed the formation of conjugated double bonds in the PVC. This explanation suggests that the silane coupling agent acted as a stabilizer for the PVC. However, when the silane coupling agent was increased to greater than 1.0 wt % (Region 2), a self-condensation reaction of the coupling agent on the surfaces of the sawdust occurred. A separation between the PVC and the treated wood sawdust proba-

bly occurred, with the PVC no longer reacting with the coupling agent. In this case, the PVC then solely underwent a dehydrochlorination reaction, which produced polyene sequences. Interestingly, the rate of polyene production increased when a high silane content was used. The increased production rate of polyene sequences in the composites involved a reaction between released HCl molecules and the PVC main chains. Hjertberg and Sorvik[23] suggested that HCl could be a catalyst for the polyene propagation through dehydrochlorination reaction. In this study, the HCl produced at this initial stage could then act as a catalyst in the PVC/sawdust composite, which accelerated the rate of the dehydrochlorination reaction and produced additional polyene sequences to the PVC molecules; this mechanism is shown in Scheme 1. However, the change in the polyene sequences in the PVC did not seem to affect the onset of the degradation temperature of PVC in the PVC/sawdust composites, whose results are shown in Figure 8. This may have been because the changes in polyene contents in the PVC composites by varying type and amount of silane coupling agents were not sufficiently significant.

CONCLUSIONS

Three silane coupling agents with different molecular structures were used for the surface treatment of wood sawdust particles that were composited with PVC. The PVC/sawdust composites with the addition of all of the silane coupling agents used in this study enhanced the mechanical properties compared with untreated PVC/sawdust composites from previous studies.[15,16] Concentrations of 0.5–1.0 and 1.5 wt % coupling agents are recommended for the optimization of tensile and impact properties of the composites, respectively. Variations in the tensile and impact properties for the composites with low wood sawdust contents were more sensitive to the addition of silane coupling agents than those with high sawdust contents. The use of KBM603 is recommended for the improvement of the tensile properties, whereas

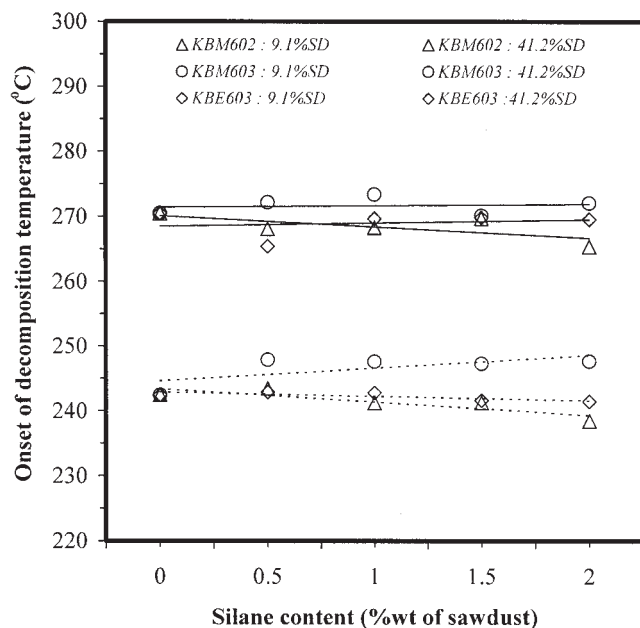**Scheme 1** Influence of HCl on the polyene sequence generation.

Figure 8 Variations in the onset of decomposition temperature of PVC in the PVC/wood sawdust composites for various types and concentrations of silane coupling agents.

KBE603 was more suitable for high impact resistance. The differences in the mechanical properties of the PVC/sawdust composites were dependent on the types and number of functional groups in the coupling agents, the self-condensation reaction of the coupling agent itself, and the effectiveness of the hydrolysis reaction. Thermal degradations of PVC in the PVC/sawdust composites were noted by the occurrence of polyene sequences at silane coupling agent concentrations above 1.0 wt %, and the decomposition temperature did not change with coupling agent concentration.

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