

Effects of Acrylic-Based Processing Aids on Processibility, Rheology, Thermal and Structural Stability, and Mechanical Properties of PVC/Wood–Sawdust Composites

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ABSTRACT: Methyl methacrylate and ethylacrylate (MMA-co-EA) and methyl methacrylate and butylacrylate (MMA-co-BA) copolymeric processing aids were introduced into poly(vinyl chloride) (PVC)/33.3 wt % wood–sawdust composites containing 0.6 and 2.4 phr of calcium stearate lubricant. The properties of the composites were monitored in terms of processibility, rheology, thermal and structural stability, and mechanical properties. It was found that the mixing torque, wall shear stress, and extrudate swell ratio increased with increasing processing aid content because of increased PVC entanglement. MMA-co-BA (PA20) was found to be more effective than MMA-co-EA (K120 and K130), this being associated with the flexibility of the processing aids, and the dipole–dipole interactions between sawdust particles and polymeric processing aids. The shark-

skin characteristic of the composite extrudate at high extrusion rate was moderated by the presence of processing aids. Adding the acrylic-based processing aids and lubricant into PVC/sawdust composites improved the thermal and structural stability of the composites, which were evidenced by an increase in glass transition and decomposition temperatures and a decrease in polyene sequences, respectively. The changes in the mechanical properties of the composites involved a composite homogeneity, which was varied by degree of entanglement and the presence of wood sawdust, and un-reacted processing aids left in the composites. © 2004 Wiley Periodicals, Inc. *J Appl Polym Sci* 92: 782–790, 2004

Key words: poly(vinyl chloride); composites; renewable resources; mechanical properties

INTRODUCTION

It is known that the addition of processing aids in small quantities into poly(vinyl chloride) (PVC) compounds can improve the processing characteristics, through an acceleration of the fusion process of PVC, and the rheological and mechanical properties of final PVC products. The term processing aids, sometimes called flow improvers, is often confused with lubricants, as both are added to ease the flowability of the PVC. Processing aids are substances that are usually compatible with PVC molecules and have high molecular weight structure, tending to give wall-adhering PVC compounds, whereas lubricants are low molecular weight structure substances, tending to produce wall-slipping PVC compounds.¹ The mechanism of wall-adhering PVC by the processing aids during processing involves increasing the coefficient of friction

between PVC particles themselves and between the PVC particles and the metal surface of the processing machinery.² These increased frictions cause a formation of shear flow patterns, result in a longer residence time of the melt near the wall, generate higher thermal shearing stresses, and eventually improve the melting characteristic. A better melting of PVC with processing aids is partly caused by an improvement of shear heating during the flow by virtue of increasing frictions between PVC particles. In early days, the commercially promoted processing aids for PVC were of the acrylic type with methyl methacrylate predominant. At present, this type of processing aid remains dominant because of the inherent compatibility between the methacrylate polymers and PVC.² In the case of the addition of the lubricants, the PVC compounds have a tendency to plug flow, which results in nonuniform melting. In general, both processing aids and lubricants are essential when processing PVC compounds, but the intermolecular mechanisms of these two additives with PVC molecules are very complex as these two additives influence each other—the processing aids change the solubility of the lubricants and other additives in the PVC.

A number of articles have been published on the effects of acrylic-based processing aids and lubricants on the properties of neat PVC compounds^{3–7}; very few

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have been for natural fiber-filled PVC composites.⁸⁻⁹ According to literature, experimental data on the properties of PVC/natural fiber composites incorporated with processing aids and lubricants have not been evidenced. It is believed that the molecular interactions between PVC, processing aids, and lubricants will be even more complex in PVC/natural fiber composites because the incorporation of hydrophilic natural fibers into the hydrophobic PVC usually leads to heterogeneous composite systems due to the incompatibility of these two materials.¹⁰ There are some articles related to the improvement of the properties of PVC/natural fiber composites, but they mostly focus on the effects of other additives such as coupling agents, plasticizers, and mechanical-modifying agents into the PVC composites.¹¹⁻¹⁸

At present, data on effects of chemical processing additives such as processing aids and lubricants on the properties of the PVC/natural fiber composites are very demanding. Therefore, research on the effects of incorporating processing aids into PVC/sawdust composites was carried out and reported, not only to complement information in the literature, but also to improve the dispersion of the fibers throughout the PVC matrix and thus enhance the mechanical performance. In this article, PVC/sawdust composites were reformulated by incorporating acrylic-based processing aids with different chemical structures at various concentrations, and a number of properties were then monitored. In this work, processing aids were added into two different PVC/sawdust composite systems, one being composites with low lubricant concentration and the other being those with high lubricant concentration. The processibility and structural changes of PVC/sawdust composites were monitored through average mixing torque, shear stress-shear rate relationship, extrudate swell ratio, and polyene content. The thermomechanical and morphological properties of the composites were assessed by glass transition and decomposition temperatures, tensile and impact properties, and hardness. The results from this work offered an in-depth understanding of the interactions between PVC, acrylic-based processing aids, and a lubricant in such a PVC/wood-sawdust composite system and obtained some criteria for the selection of the most suitable processing aids and their optimum concentration for addition into the composite, as well as the recommended processing conditions.

EXPERIMENTAL

Raw materials

All PVC compounds, composites, and ingredients used in this work are listed in Table I.

PVC was supplied by V. P. Plastics Products (1993) Co., Ltd. (Bangkok, Thailand), in the form of com-

TABLE I
The Ingredients of PVC Composites

PVC composite ingredients	Concentrations (pph)
Suspension PVC grade SIAMVIC 258RB (<i>K</i> value = 58)	100.0
Emulsion PVC grade SIAMVIC 374MB (<i>K</i> value = 74)	4.0
Organic complex stabilizer (TS-DBL-Pb-Ba)	2.6
External lubricant (Finalux G-741)	0.3
Internal lubricant (Calcium stearate)	0.6 and 2.4
CaCO ₃ (Omyacarb-2T)	12
Processing aids	
PA20 (MMA- <i>co</i> -butyl-acrylate)	Varied
Paraloid K120 (low <i>M_w</i> MMA- <i>co</i> -ethyl-acrylate)	Varied
Paraloid K130 (high <i>M_w</i> MMA- <i>co</i> -ethyl-acrylate)	Varied
Carpentry sawdust particles	50 (equal to 33.3 wt %)

pounded PVC powder. There were two types of PVC compounds used in this work: one being suspension PVC (SIAMVIC 258RB) (V.P. Plastics Product Co. Ltd, Bangkok, Thailand) with a *K* value of 58, and the other being emulsion PVC (SIAMVIC 374MB) with a *K* value of 74.

Sawdust particles were obtained from carpentry and wood-working processes and supplied by V. P. Plastics Products (1993) Co., Ltd. The average size of the sawdust particles used in this work was in the range of 100–300 μm. The wood-sawdust particles were chemically untreated and the content added into the PVC compound was fixed at 33.3 wt % PVC throughout this work. The sawdust was dried to minimize the moisture content to a known value (5% for this work). Previous work¹⁸ recommended 33.3 wt % sawdust content with a moisture content less than 5% for optimization of properties of the PVC/sawdust composites, the sawdust particles being dried prior to use through a heat treatment in an oven for 24 h at 80°C until the sawdust weight was constant.

The processing aids used in this work were commercial copolymers of methyl methacrylate and ethylacrylate, and methyl methacrylate and butylacrylate, whose chemical structures are shown in Figure 1. These include MMA-*co*-ethylacrylate (MMA-*co*-EA), supplied by Ha Chemicals Co., Ltd. (Singapore), with low (Paraloid K120N) and high (Paraloid K130) molecular weights, and MMA-*co*-butylacrylate (MMA-*co*-BA) (PA20), supplied by Kaneka Corp. Co., Ltd., (Tokyo, Japan). By using these processing aids, the effects of molecular weight and chemical structure of the processing aids on the properties of PVC/sawdust composites could be discussed.

Other chemicals included organic complex stabilizer, external lubricant, calcium carbonate, and calcium stearate, whose ingredients are shown in Table I.

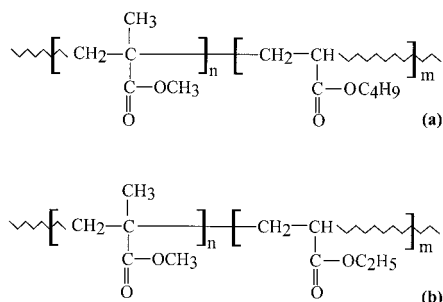


Figure 1 The chemical structures of processing aids (a) MMA-co-butylacrylate (PA20) and (b) MMA-co-ethylacrylate (K120 and K130).

Blending process of PVC and sawdust particles

PVC compounds were dry-blended with sawdust particles by using a high-speed mixer for 5 min before being melt-blended in a twin-screw extruder (Haake PolyLab-Rheomex CTW100P, Germany). The blending temperature profiles on the extruder were 165, 175, 175, and 185°C from hopper to die zones. The screw rotating speed used was 100 rpm and a slit die having width \times height \times length of 18.2 \times 2 \times 16 mm³ was used to produce slit extrudates, which then passed through a water bath for cooling.

Characterizations

Processibility and flow properties

The processibility and flow properties of the composites, containing various amounts of processing aids, were assessed in three different tests as follows.

(1) Average mixing torque values were measured in a twin-screw extruder (Haake PolyLab-Rheomex CTW100P) during melt-blending. The processing temperature profiles on the extruder were used as in the blending process. A single-hole die with 5 mm diameter and 16.4 mm length was located at the end of the extruder.

(2) The flow curves of the PVC/sawdust composites containing different concentrations of processing aids were determined by using an Instron 4467 Extrusion Rheometer (MA, USA) having a barrel diameter of 9.5 mm, fitted with a die 1.25 mm in diameter and 44 mm in length. The test procedure can be found elsewhere.¹⁹ Die entrance pressure was measured by using a photoconductive light pressure sensor.²⁰

(3) The extrudate swell was assessed by determining the ratio of the diameter of the extrudate to that of the die during extrusion in the capillary rheometer.

Thermal and structural stabilities

The thermal properties of the PVC/sawdust composites were determined in terms of glass transition and

decomposition temperatures. Glass transition temperature of PVC/sawdust composites was determined by using a Dynamic Mechanical Analyzer (DMA; Model-2980, TA Instruments Ltd., MA, USA). The test samples were rectangular in shape, having 17.5 mm length, 15 mm width, and 2 mm thickness. The dual cantilever mode of deformation was used under test temperatures from 0 to +140°C with a heating rate of 5°C min⁻¹. The test amplitude and frequency were 15 μm and 1 Hz, respectively. The cooling process was achieved through liquid nitrogen. To determine the decomposition temperature (T_d), a TA Instruments thermal gravimetric analyzer (TGA-2950) (Massachusetts) was used. A temperature range of 50–600°C, with a heating rate of 10°C/min, was used.

Polyene sequences present in PVC/sawdust composites were measured to assess the structural changes of PVC molecules in PVC/sawdust composites by using Fourier transform infrared (ATR-FTIR) analysis.²¹ This was carried out by measuring the percentage reflectance difference at the surface of the sample. The tests were performed by using a Nicolet Nexus 470-FTIR spectrometer (WI, USA) to produce spectra of reflectance difference against wave number. In this work, the peak of polyene sequences at a wave number of 1584 cm⁻¹ was of interest. The polyene sequences were calculated in terms of percentage reflectance (ΔR), this being carried out by subtracting the percentage reflectance (%R) of the peak of interest, from that of the baseline, as shown in eq. (1). It should be noted that the percentage weight of PVC (66.7 wt %) had to be taken into account when determining the polyene sequences

$$\Delta R \text{ of Polyene sequence} = \frac{\%R_{\text{Baseline}} - \%R_{\text{c=c}}}{66.7} \times 100 \quad (1)$$

Mechanical performance

Tensile, impact, and hardness properties of PVC/sawdust composites were measured to assess the mechanical performance of the composites containing different types and levels of processing aids. The tensile test was performed on a Hounsfield QMAT 1.23 tensile testing machine (Surrey, UK) at a cross-head speed of 5 mm min⁻¹, the tensile test procedure following the ASTM D638 (1990) Specimen Type I. Izod impact tests were carried out, according to ASTM D256 (1990), on a Yasuda Impact Tester (Osaka, Japan) with the notched side facing the pendulum. Hardness was evaluated by using a Durometer Shore D (Model 409) supplied by PTC Instruments Co. Ltd. (CA, USA), the test procedure being specified by ASTM D2240 (1997).

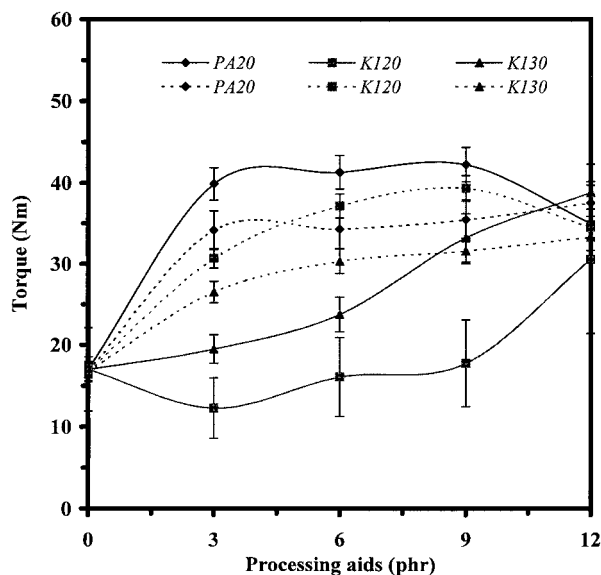


Figure 2 Variations of average torque value of PVC/sawdust composites for various processing aids at 0.6 phr (solid line) and 2.4 phr (dashed line) lubricant contents.

RESULTS AND DISCUSSION

Processibility and flow properties

Figure 2 shows the relationship between average torque value and the additional level of processing aids at 0.6 and 2.4 phr lubricant contents. Generally, it was found that the average torque increased with increasing processing aids. This was expected because the addition of processing aids increases interparticle frictions and the fusion process of PVC molecules.²⁻³ The details of the results can be explained in two different cases, these being low- and high-lubricant contents.

At low lubricant level (0.6 phr)

The average torque value for PVC/sawdust composites with PA20 was greater than that with K130 and K120, the average torque value, for a given processing aid content, decreasing in the order of PA20 > K130 > K120. This was associated with the differences in molecular structures of the processing aids used. This suggested that the fusion of PA20/PVC composites was greater than that of K130/PVC and K120/PVC composites. This may be caused by two possible reasons. First, PA20 is a copolymer of MMA-co-BA, whose molecular chains are more flexible and have lower glass transition temperature than K130 and K120 (MMA-co-EA). During thermal processing, the molecular chains of PA20 are more easily and more likely to become softened and then react with PVC molecules through polar-polar attractions, and thus, increase in interparticle friction and degree of PVC

fusion. The second reason is associated with dipole-dipole interactions between processing aids and wood-sawdust particles. Having considered the chemical structures of processing aids and wood-sawdust, it is thought that the sawdust particles preferred to react with K120 and K130, which also had greater polarities than PA20. Therefore, the effectiveness of K120 and K130 to induce entanglement in PVC/sawdust composites decreased. To yield a higher average torque level (entanglement level), the amounts of K120 and K130 had to be increased to about 12 phr. Therefore, it could be said that PA20 was more effective, in terms of assisting in PVC entanglement, than both K120 and K130. Considering the effect of the molecular weight of the processing aids (K120 and K130), PVC composites with the higher molecular weight processing aid (K130) appeared to show greater torque value (higher entanglement level) due to the fact that greater molecular weight tended to have a greater chance to react with PVC molecules during processing.

At high-lubricant level (2.4 phr)

It was clearly seen that when the amount of the lubricant was increased from 0.6 to 2.4 phr the effect of the type of processing aid was suppressed by the lubricating effects. Work by Grossman²² indicated that the main function of a lubricant in PVC was to facilitate the internal rotations of polymer molecules (when miscible in the polymer) and to increase the polarity of the polymers (both PVC and polymeric processing aids in this case). It was postulated that the increase in the polarities of the polymers resulted in two simultaneous effects: (1) promoting the entanglement level due to increased dipole-dipole interactions between PVC particles and processing aids,² and (2) facilitating (or lubricating) the molecular mobility of the polymers.²² The former effect resulted in an increase in average torque value, which was the case for composites with K120 and K130 because of their abilities to increase the entanglement level. The latter case resulted in a decrease in the average torque value, which was the case for composites with PA20.

Table II lists the values of shear stress and extrudate swell ratio of the composites with various types and concentrations of processing aids at two different shear rates (27.8 and 55.6 s⁻¹). It can be seen that the composites with PA20 had a greater shear stress and extrudate swell ratio than those with K130 and K120. The differences in the shear stress and extrudate swell values were closely associated with the degree of entanglement induced by each processing aid used. Previous work²³ found that the greater the entanglement the greater the shear stress and the swelling ratio of the PVC extrudate. The shear stress and extrudate swell ratio of the composites at a shear rate of 55.6 s⁻¹

TABLE II
Changes in Shear Stress and Extrudate Swell Ratio of PVC/Sawdust Composites for Different Processing Aids at 0.6 and 2.4 phr Lubricant Contents

		Rheological properties											
		Shear stress $\times 10^4$ (N m $^{-2}$)						Extrudate swell ratio					
Chemical content (phr)		PA20		K120		K130		PA20		K120		K130	
Lubricant	Processing aids	Shear rate (s $^{-1}$)		Shear rate (s $^{-1}$)		Shear rate (s $^{-1}$)		Shear rate (s $^{-1}$)		Shear rate (s $^{-1}$)		Shear rate (s $^{-1}$)	
		27.8	55.6	27.8	55.6	27.8	55.6	27.8	55.6	27.8	55.6	27.8	55.6
0.6	0	5.1	5.3	5.1	5.3	5.1	5.3	1.02	*	1.02	*	1.02	*
	3	23.9	*	7.9	8.3	12.0	17.6	1.08	*	1.06	*	1.08	1.11
	6	28.9	*	14.3	24.0	12.4	22.5	1.08	*	1.10	*	1.11	1.14
	9	12.8	23.3	11.3	13.1	7.5	8.3	1.15	1.19	1.11	1.12	1.05	0.98
	12	17.3	25.5	13.9	23.3	8.3	8.6	1.19	1.18	1.15	1.17	1.12	1.11
2.4	0	7.1	7.9	7.1	7.9	7.1	7.9	1.05	*	1.05	*	1.05	*
	3	13.9	15.4	6.4	6.8	6.0	8.3	1.05	*	1.07	*	1.07	*
	6	15.0	26.3	7.9	12.0	12.4	15.4	1.10	1.14	1.09	*	1.09	*
	9	21.4	28.2	9.4	18.8	6.0	13.5	1.14	1.18	1.12	1.14	1.14	*
	12	25.5	30.4	11.5	22.4	7.9	13.9	1.17	1.22	1.14	1.13	1.14	*

*indicates extrudate distortions during extrusion.

were also found to be greater than those at 27.8 s $^{-1}$, this being caused by the fact that the entanglement of PVC was promoted by the increasing rate of deformation.^{1,23} For a given processing aid, the shear stress and extrudate swell ratio were observed to increase with increasing processing aid content, the results being in good agreement with the torque results, as shown in Figure 1. The asterisk in Table II indicates a sharkskin surface (from rough to fracture surface) of the composite extrudates during extrusion (this was observed during the experiment, thus the results are not shown). During the experiment, there were no distortions of the extrudate observed under a shear rate of 27.8 s $^{-1}$, but such distortions started to appear when the shear rate was increased to 55.6 s $^{-1}$. It is interesting to note from the results in Table II that the sharkskin character of the composite extrudate at high shear rate was moderated by the addition of processing aids. The addition level depended on the type of processing aids used, preferably at concentrations of around 6–9 phr for the processing aids used in this work. The improved extrudate texture surfaces resulted from an increased melt strength of the extrudates by the presence of suitable amounts of processing aids.²

Thermal and structural stability

Figure 3 shows the changes in glass transition temperature of PVC/sawdust composites having different types and addition levels of processing aids at 0.6 and 2.4 phr lubricant contents. It was generally found that the variations of T_g were in the ranges of 91.5 to 95.5°C. At 0.6 phr lubricant content, the T_g of composites with PA20 increased with processing aid content

and leveled off around 9–12 phr processing aid contents; this behavior was supported by Petrich and Lutz.² The increase in T_g was associated with an increase in entanglement level due to the presence of the processing aids. As the degree of entanglement increases, the molecular chains become more difficult to mobilize.²⁴ It was also found that the T_g value of PVC/sawdust composites with PA20 was higher than with K120 and K130 for a given processing aid content due to the higher effectiveness of PA20 as stated. The changes in the T_g value of the composites at 2.4 phr

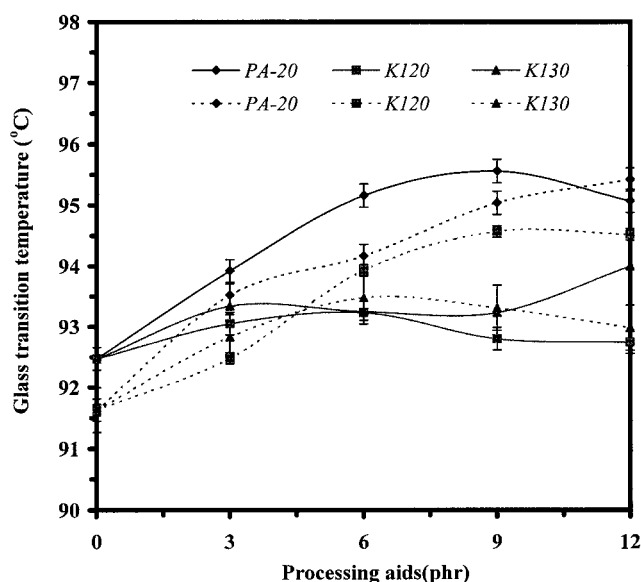


Figure 3 Effect of processing aid contents on glass transition temperature of PVC/sawdust composites at 0.6 phr (solid line) and 2.4 phr (dashed line) lubricant contents.

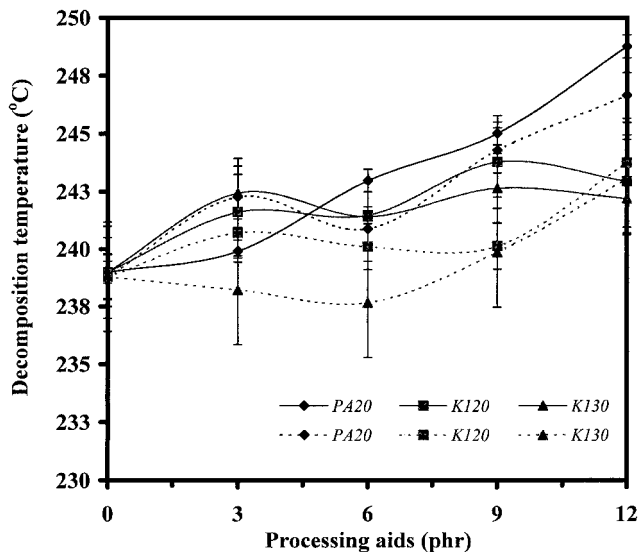


Figure 4 Variations in decomposition temperature with processing aid contents at 0.6 phr (solid line) and 2.4 phr (dashed line) lubricant contents.

lubricant content can be explained in connection with the changes in torque value in Figure 2. As the content of the lubricant was increased from 0.6 to 2.4 phr, the entanglement level of the composites with K120 and K130 arose because of the increased polarity of the PVC and polymeric processing aids. This may therefore have resulted in the increase in T_g values of the composites with K120 and K130 at 2.4 phr lubricant. The decrease in the T_g value of composites with PA20 was due to the lubrication effect. Similar behavior was also found with the changes in decomposition temperature (T_d), as illustrated in Figure 4, showing the T_d value of the composites increasing with increasing processing aids.

Figure 5 represents structural stability of PVC in PVC/sawdust composites in terms of conjugated double-bond (polyene) sequences with varying processing aid contents. It can be seen that, at both lubricant contents, the composites with PA20 had fewer polyene sequences than those with K120 and K130 for any given processing aid contents. This suggested that the composites with PA20 were more stabilized (more difficult to degrade) than those with K120 and K130, the results being very coherent with the T_g and T_d results. Previous work¹⁸ has indicated the Cl atoms from PVC could be cleaved due to a re-formation of hydrogen bonds in sawdust particles, resulting in a production of polyene units in PVC. In relation to this work, the wood-sawdust particles preferred to react with the added processing aids through hydrogen bonds rather than with PVC. Therefore, the cleavage of Cl atoms in PVC decreased, resulting in increased molecular stability. It should also be noted that the changes in polyene sequences were independent of the content of the processing aids.

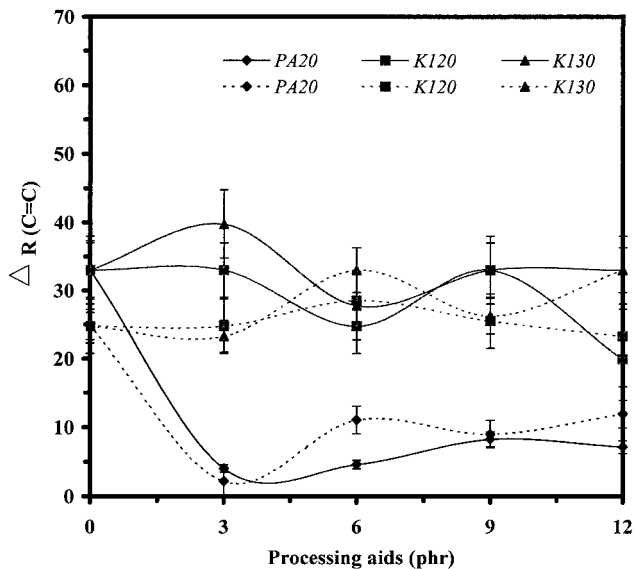


Figure 5 Changes in polyene content of PVC for different processing aids at 0.6 phr (solid line) and 2.4 phr (dashed line) lubricant contents.

Mechanical properties

Figure 6 shows the effect of processing aid content on the tensile modulus of PVC/sawdust composites for 0.6 and 2.4 phr lubricant contents. It was found that the tensile modulus of the composites was independent of the PA20 content, but progressively decreased with K120 and K130 contents, the latter effect being more pronounced as the lubricant content was increased to 2.4 phr. The composites with PA20 had a

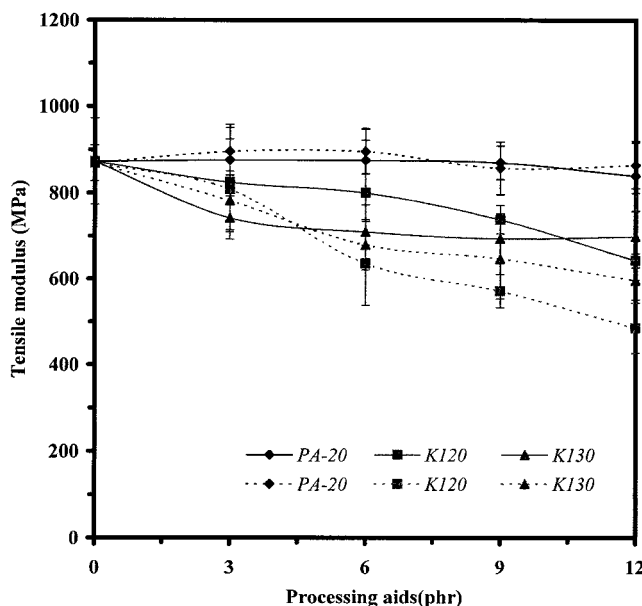


Figure 6 Plots of tensile modulus of PVC/sawdust composites at different processing aid contents at 0.6 phr (solid line) and 2.4 phr (dashed line) lubricant contents.

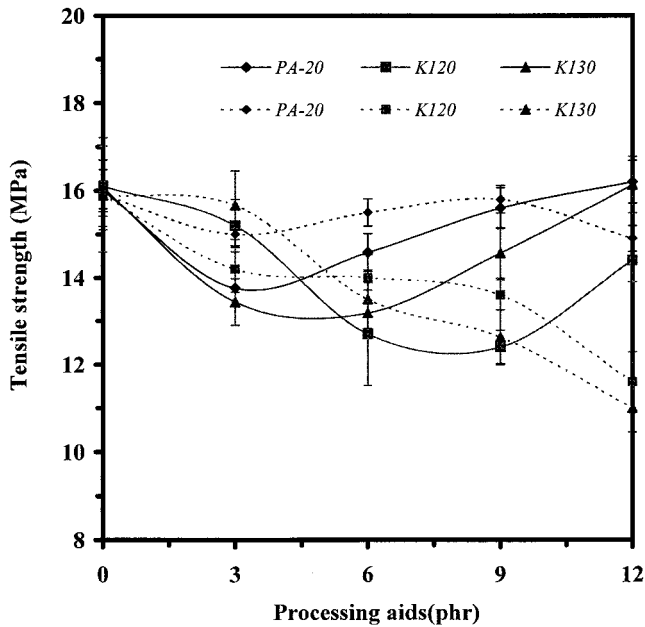


Figure 7 Effect of processing aids on elongation at break of PVC/sawdust composites with 0.6 phr (solid line) and 2.4 phr (dashed line) lubricant contents.

greater tensile modulus than those with K120 and K130 because of a higher gelation level. The unchanged tensile modulus of the composites with PA20 was probably due to the leveling-off of the entanglement when adding PA20. The progressive decrease in tensile modulus of the composites with K120 and K130 may be explained in relation to the effectiveness of K120 and K130. From the torque results in Figure 2, K120 and K130 had less ability to induce or accelerate the entanglement of PVC than PA20. During processing, certain amounts of K120 and K130 required for the acceleration of entanglement had probably not been used and the leftover polymeric processing aids then acted as polymer blends (with PVC) in the composite system. Because the processing aids used in this work were of a more flexible material as compared with PVC, the tensile modulus of the composites with K120 and K130 therefore decreased with increasing concentrations of K120 and K130.

Tensile strength is widely associated with an ability for polymer molecular chains to slip past each other; the more difficult the slippage of the molecular chains, the greater the tensile strength. Figure 7 shows the changes in tensile strength of PVC/sawdust composites having different types and levels of processing aids at 0.6 and 2.4 phr lubricant contents. At low lubricant content (0.6 phr), the tensile strength decreased with increasing processing aids to give a minimum and then started to increase with further processing aid contents. The decrease in tensile strength was associated with a nonhomogeneity of the composite system (PVC compounds, wood sawdust, and pro-

cessing aids). The addition of a small amount of processing aids probably gave a partial entanglement of PVC particles; the degree of entanglement for a given amount of processing aids depending on the effectiveness of the processing aids used. The partial entanglement of PVC particles led to nonhomogenous phases (gelled and ungelled phases), which acted as defects in the PVC composites, the nonhomogeneity being more pronounced with the presence of wood-sawdust particles, thus decreasing tensile strength. It is interesting to note that the position and the value of minimum tensile strength for each processing aid was different; the greater the effectiveness of the processing aids, the higher the value of the minimum, and the lower the processing aids amount required before improving the tensile strength. The increase in tensile strength at higher processing aid contents was caused by a decrease in the nonhomogeneity of the composites as the degree of entanglement had increased. These results corresponded very well with the torque results. However, upon application of the high lubricant content (2.4 phr), the tensile strength for composites with relatively low entanglement (K120 and K130) decreased with processing aid content, whereas that with PA20 did not change significantly.

Figure 8 illustrates the effects of processing aid content on the elongation at break of PVC/sawdust composites. The changes in elongation at break of the PVC/sawdust composites at 0.6 phr lubricant mirrored the tensile strength results. The decrease in elongation at low processing aid content was caused by the nonhomogenous phases as a result of partial en-

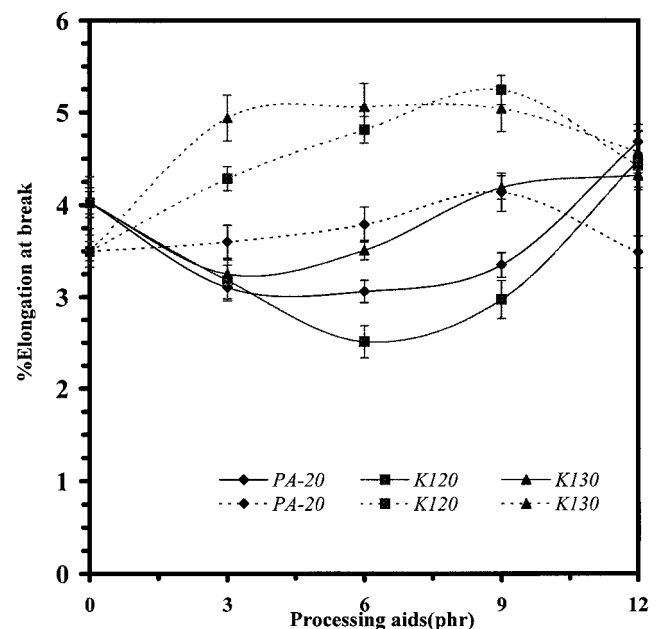


Figure 8 Variations of tensile strength as a function of processing aid contents at 0.6 phr (solid line) and 2.4 phr (dashed line) lubricant contents.

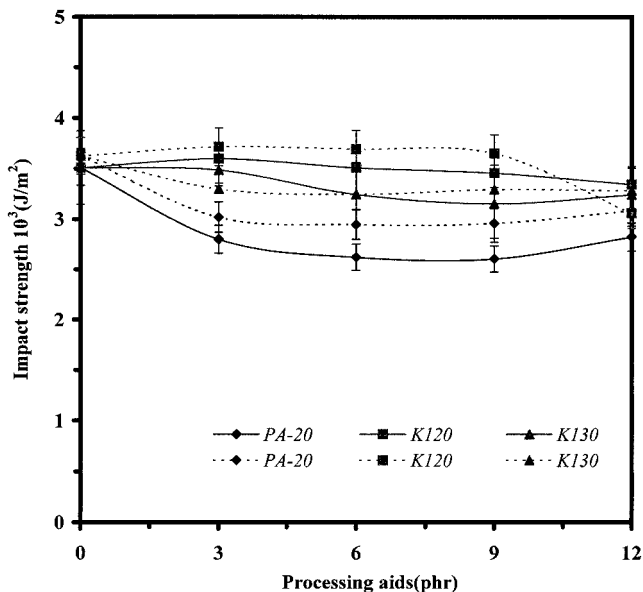


Figure 9 Effect of processing aids on impact strength of PVC/sawdust composites with 0.6 phr (solid line) and 2.4 phr (dashed line) lubricant contents.

tanglement, whereas a progressive increase in elongation at break after the minimum was caused by increased entanglement which led to more homogeneity of the composite system. The entanglement effect became less pronounced as the lubricant content was increased from 0.6 to 2.4 phr because the lubricant was also responsible for slippage of PVC molecules, which increased the elongation at break.

Impact strength of PVC/sawdust composites as a function of processing aids is shown in Figure 9. The results are best explained in connection with tensile property results. At 0.6 phr lubricant content, the decrease in impact strength was due to the nonhomogeneity of the composite system which then led to a difficulty for stress transfer under impact load, the effect being less pronounced as the lubricant content was increased. The greater impact strength of the composites with K120 and K130, as compared with that with PA20, was probably due to some residual unreacted processing aids present in the case of composites with K120 and K130, as discussed earlier. These residual processing aids were flexible materials as compared to PVC, thus increased impact strength of the composites.

Figure 10 shows the changing hardness property for PVC/sawdust composites with different concentrations of PA20, K120, and K130 at 0.6 and 2.4 phr lubricant contents. It should be noted that it was very difficult to quantitatively evaluate the changing hardness property because of varying contents of processing aids and lubricant because the hardness measurement had been carried out only on the surface of the composite samples, which may only be slightly af-

ected by the additions of the processing aids and lubricant, as the surface properties were also dependent on processing conditions such as cooling and shear rate.¹⁸ For qualitative analysis, the hardness of the composites reduced with increasing processing aids, but the effect was less when increasing the concentration of the lubricant.

In summary, it seems that introduction of the acrylic-based processing aids and lubricant did not improve the mechanical properties of PVC/sawdust composites. However, the processing aids were found to clearly assist in causing increases in the thermal and structural stability, and the ability to process the composites, which were being evidenced by increases in average mixing torque, glass transition temperature, and decomposition temperatures, accompanied by a decrease in polyene sequences (especially for the PA20 case).

CONCLUSION

MMA-co-EA (K120 and K130) and MMA-co-BA (PA20) were added into PVC/33.3 wt % sawdust composites containing 0.6 and 2.4 phr of calcium stearate. The following were noted.

1. The mixing torque, shear stress, and extrudate swell ratio increased with increasing processing aid content because of the increased PVC entanglement. PA20 was more effective in terms of accelerating the entanglement than both K120 and K130. The sharkskin characteristic of the composite extrudate at high extrusion rate was moderated by the presence of pro-

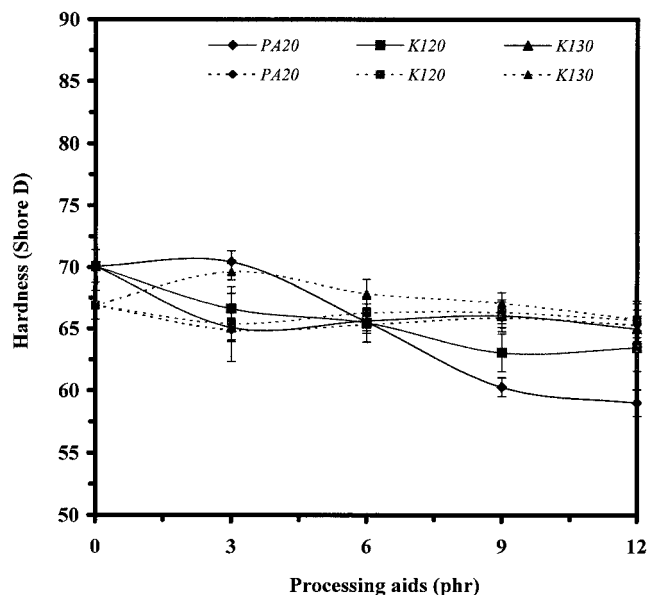


Figure 10 Effect of processing aids on hardness of PVC/sawdust composites with 0.6 phr (solid line) and 2.4 phr (dashed line) lubricant contents.

cessing aids, preferably at concentrations of around 6–9 phr.

2. The glass transition and decomposition temperatures of PVC/sawdust composites were found to increase with processing aid content, the effect being reduced as the amount of calcium stearate lubricant was increased.

3. The number of polyene sequences of PVC in PVC/sawdust composites did not change significantly with the addition of K120 and K130, but was greatly reduced with increasing PA20 content.

4. Tensile modulus was noticeably reduced with K120 and K130, but did not change with PA20 for all concentrations. The changes in tensile strength and elongation at the break were dependent on the homogeneity of the composites.

5. The differences in impact strength of the composites as a result of various processing aids were related to the composite homogeneity and the residue of the un-reacted processing aids.

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