Journal of Industrial Technology 16 (1), 2007, 13-23

EFFECT OF OPEFB ON MECHANICAL PROPERTIES AND PROCESSIBILITY OF CPE-IMPACT MODIFIED PVC-U COMPOSITES

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ABSTRAK : *Kesan pengisi OPEFB terhadap sifat mekanikal dan kebolehprosesan CPE-PVC-U terisi-OPEFB komposit adalah dikaji. Resin poli (vinil klorida) (PVC), terklorin polyethylene (CPE), pengisi tandan kosong kelapa sawit (OPEFB) dan aditif lain pada mulanya diadun kering menggunakan pengisar berskala makmal sebelum dikepingkan dengan pengisar dua pengguling. Spesimen-spesimen ujian kemudiannya ditekan panas dan selepas itu ujian mekanikal ditentukan. Kajian kebolehprosesan adunan-adunan kering dilakukan dengan menggunakan Brabender Torque Rheometer. Keputusan menunjukkan bahawa modulus lenturan meningkat manakala kekuatan hentaman dan kekuatan lenturan komposit tanpa pemplastik poli(vinil klorida) (PVC-U) terisi-OPEFB menurun dengan penambahan OPEFB. Masa lakuran komposit PVC-U terisi-OPEFB terubahsuai hentaman-CPE menurun dengan penambahan 10 dan 20 phr OPEFB tetapi meningkat secara mendadak dengan penambahan seterusnya kepada 30 dan 40 phr.*

ABSTRACT : The effect of OPEFB fillers on the mechanical properties and processibility of CPE-impact modified PVC-U composites were investigated. The poly (vinyl chloride) (PVC) resin, chlorinated polyethylene (CPE), oil palm empty fruit bunch (OPEFB) filler, and other additives were first dry-blended using a laboratory blender before being sheeted with a two-roll mill. Test specimens were then hot pressed, after which the mechanical properties were determined. The processibility studies of the dry-blends were carried out using a Brabender Torque Rheometer. The results showed that the flexural modulus improved while impact and flexural strength of OPEFB-filled composites decreased with the addition of OPEFB. The fusion time of the CPE-impact modified OPEFB-filled unplasticised poly (vinyl chloride) (PVC-U) composites decreased upon the addition of 10 and 20 phr of OPEFB but increased drastically on further increase to 30 and 40 phr.

KEYWORDS : PVC-U, CPE, OPEFB filler, impact strength, flexural properties, fusion time.

INTRODUCTION

Poly (vinyl chloride) (PVC) is the world's third most important thermoplastics in terms of volume sales. The reasons for the success of PVC include its versatility, extraordinary stability to weathering and competitive price (Armitt, 2004). However, PVC has a disadvantage of being prone to occasional brittleness and is notch sensitive.

PVC must undergo fusion in every fabrication in order to achieve its best mechanical properties. It is generally agreed by researchers that the level of fusion has significant influence on the mechanical, physical, and chemical properties of PVC (Covas *et al.,*1988). The properties of PVC are dependent on the degree of fusion, thus additives that control the rate of fusion within the processing equipment become important ingredients in a successful formulation. Many studies have shown that external lubricants delay the fusion time of PVC and internal lubricants promote the fusion time, but the synergistic reaction between them has promoted the fusion time of PVC significantly (Chen *et al.,* 1995; Robinovitch *et al.,* 1984).

Oil palm empty fruit bunch (OPEFB) fibre is one of the oil palm by-products generated from oil palm industries. In order to exploit the abundance of this industrial waste and reduce environmental problems, new applications are required for OPEFB. The incorporation of OPEFB fibre as a reinforcing component in polymers has received much attention (Rozman *et al.,* 2000; Rozman *et al.,* 2001; Abdul Khalil *et al.,* 2001; Sreekala *et al.,* 1996/1997) but the use of OPEFB in PVC has not been studied extensively. Previous researchers on PVC-natural filler composites focused on wood flour, wood sawdust and rice husk (Sombatsompop and Phromchirasuk, 2004; Azman Hassan and Kannan, 2001; Matuana *et al.,* 1998).

In our previous paper, the effects of incorporation of OPEFB fibre on the mechanical and thermal properties of EFB-filled PVC-U composites were reported (Abu Bakar *et al.,* 2005). The investigation revealed that an increase of OPEFB fibre content in the PVC-U matrix improved the flexural modulus but decreased the impact and flexural strength. It is known that the addition of impact modifiers into unplasticised poly (vinyl chloride) (PVC-U) compounds can improve the toughness of PVC-U. Chlorinated polyethylene (CPE) impact modifier is amongst the popular impact modifier for PVC. Impact modifiers improve the toughness of a polymer by facilitating the ability of the polymer matrix to shear yield and/or craze (Stevenson, 1995). CPE forms a network enclosing PVC primary particles in order to develop the desired impact strength. However, CPE is dependent on processing conditions whereby the network starts to collapse and toughness decreases with increasing temperature or shear (Robinovic, 1983). In this study, the CPE content in all formulations was fixed at 9 phr. Previous study showed that the optimum properties in terms of impact strength and flexural modulus occurred at 9 phr CPE content (Aznizam, 2006). The influence of OPEFB at various loading level on the mechanical properties and processability of CPE-impact modified PVC-U was investigated.

MATERIALS AND METHODS

Materials

The suspension PVC resin used in this study, with solution viscosity K-value 66, and the trade name MH-66 was supplied by Industrial Resin Malaysia (IRM) Sdn. Bhd. Tampoi, Johor, Malaysia. Its specifications are summarized in Table 1. The additives used in the PVC-U formulations as shown in Table 2 were also supplied by IRM Sdn. Bhd. The OPEFB fibres were purchased from Sabutek Sdn. Bhd., Teluk Intan, Perak, Malaysia. The CPE with the trade name Tyrin 702P was supplied by Dupont Dow Elastomers (Exodus Sdn. Bhd. Malaysia).

Table 1. Specifications of PVC Suspension Resin MH-66

Appearance	White Powder
Degree of polymerisation	1000 ± 50
K-value	66
Specific gravity	1.4
Bulk density (g/cm^3)	0.50 ± 0.05
Volatile Matter (max) (%)	0.5
Foreign Matter (grain/100g)	15
Particle size (retained on 250 μ), (max) (%)	0.3

Table 2. Blend Formulations

Resin	**Part per hundred of PVC resin (phr)**
Poly(vinyl chloride)- PVC	100.0
Additives Tin Stabilizer (Heat Stabilizer)	2.0
Calcium Stearate (Internal Lubricant)	0.5
Stearic Acid (External Lubricant)	0.6
Acrylic Polymer (Processing Aid)	1.5
Titanium Dioxide (Pigment)	4.0
Filler Oil Palm Empty Fruit Bunch (OPEFB)	10, 20, 30, 40
Impact Modifier Chlorinated Polyethylene (Tyrin 702P)	9

Filler Preparation

A Restsch shaker was used to separate the OPEFB fillers into different sizes. The shaking time was 10 min. The size of the OPEFB fillers used in this study was less than 75 μm. The fillers were dried in an oven at 105°C for about 24 h to a constant weight.

Dry Blending Process

The blend formulations of the filled composites are shown in Table 2. The dry blending process for each blend formulation was first done using a laboratory high-speed mixer for 10 min to homogenise the dry blends. All the dry-blended specimens were then used for processibility study and specimen preparation.

Processibility Study

A Brabender Torque Rheometer model PL2200 was used to study the processibility of dry-blended specimens. 54 g of dry-blended specimen was placed into the mixing chamber through a loading chute. Immediately after the dry-blended specimen has been loaded, the piston with 5 kg weight was inserted in place. The piston was pressed gently to force the entire dry-blended specimen into the mixing chamber as quickly as possible (within 20 s) for best reproducibility and comparability of test result. All specimens were run at the constant rotor speed of 50 rpm and at mixer temperature of 180°C. The fusion time of each dry-blended specimen were analysed by observing the changes of the torque curve.

Specimen Preparation

Each dry-blended specimen was first melt-blended and sheeted using a laboratory two roll-mill at 165°C for 10 min. The milled sheets were then placed into a mould with five rectangular cavities and hot pressed at temperature and pressure of 180°C and 1.2 kPa, respectively, for 5 min. The specimens were cooled for 5 min before being removed from the mould cavities.

Mechanical Tests

The Izod impact strength assessment was carried out on notched specimens with dimensions of 62.5 x 13 x 3 mm^3 at room temperature using an IMPats pendulum impact tester at an impact velocity of 3.0 m s^{-1} and 90° swing angle. The specimens were notched with a notch

cutting apparatus. The notch depth was fixed at 2.6 ± 0.02 mm. Flexural tests were conducted on the Instron Machine Model 5567 according to ASTM D790. The specimens, with dimensions of 125 x 13 x 3 mm^3, were tested at crosshead speed of 3 mm/min at room temperature. The support span for the flexural test was 51 mm. All the reported values for the tests were the average of seven specimens.

RESULTS AND DISCUSSION

Processibility Properties

Table 3 shows that the fusion time of the CPE-impact modified OPEFB-filled PVC-U composites decreased with composites added with 10 and 20 phr of OPEFB, however, the fusion time increased drastically as the OPEFB content increased to 30 and 40 phr. The decrease in fusion time was due to the effect of oil residues in the OPEFB. The presence of oil residues on the OPEFB fillers has also been reported by other researchers (Rozman *et al.,* 2001). OPEFB fillers coated by the oil tend to act as an internal lubricant due to the ester group, which is the main component in the oil. The ester component in the oil residues interacted with calcium stearate, which acts as internal lubricant, to enhance the polarity of calcium stearate and form a good adhesive that binds the PVC resin particles together easily. As a result, the PVC resin particles of OPEFB-filled composites fused at a shorter time. At higher OPEFB content (30 and 40 phr) as shown in Figure 1, there is a higher tendency of OPEFB fillers to agglomerate among themselves that resulted in a greater separation of PVC resin particles in the composites. This outweighs the increase of adhesive property formed by oil residues and calcium stearate. Because of this, the fusion time increased drastically as the filler content increased from 20 to 40 phr.

Table 3. Fusion Time of CPE-Impact Modified OPEFB-Filled Composites

OPEFB filler content (phr) **CPE content (phr)**	**0** **9**	**10** **9**	**20** **9**	**30** **9**	**40** **9**
Fusion time (s)	88	80	82	150	160
Fusion time increment (%)		33	21	150	176

The fusion time of OPEFB-filled composites with 9 phr CPE from the present study was higher than the values reported in our previous papers on unmodified and acrylic-impact modified (Abu Bakar *et al.,* 2005; Abu Bakar *et al.*, 2005). This indicates that CPE impact modifier, which is less compatible with PVC resin particles, reduced the adhesive property of calcium stearate. The ability of CPE in reducing the adhesive property of calcium stearate had also been reported by other researchers (Chen *et al.,* 1995). As an internal lubricant in the filled

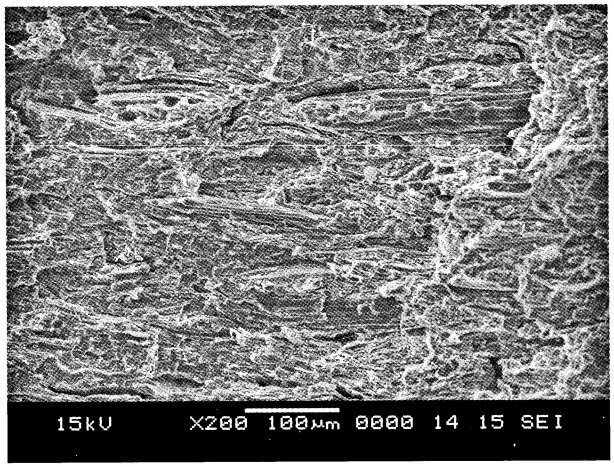

Figure 1. *SEM micrograph of impact fractured surface of composites with filler loading of 40 phr (magnification 200 x)*

composites, the strong polar ends of calcium stearate molecules (CaCOOC-group) are able to perform as adhesive to bind to the adjacent polar PVC resin particles together. In this study, the combined lubricant effect between calcium stearate and CPE was probably insufficient to bind the PVC resin particles together in the filled composites. It means that the CPE and calcium stearate acted as fusion delayer for the filled composites. As a result, PVC resin particles took a longer time to reach a homogeneous melt.

Impact Strength

Figure 2 presents the influence of EFB content upon impact strength of CPE-impact modified filled composites. The result shows that as the fibre content increased from 10 to 40 phr the impact strength of the filled composites was reduced by about 38%. The decrease of impact strength with increasing fibre content is expected and has been reported in our previous papers and other researchers (Woodhams *et al.,* 1984; Zaini *et al.,* 1996; Rozman *et al.,* 1998). Our previous study has also shown that the impact strength of the unmodified OPEFB-filled PVC-U composites decreased with increasing filler content (Abu Bakar *et al.,* 2005).

The decrease was attributed to two possible reasons. Firstly, the detrimental effect of fibres on the impact performance is due to the volume taken up by the fibres. OPEFB fibre unlike the matrix is incapable to dissipate stress through the mechanism known as a shear yielding prior to fracture. It may also hinder the local chain motions of the PVC molecules that enable them to shear yield (Stevenson, 1995). As a result, the ability of filled composites to absorb energy during fracture propagation was decreased. It has been shown that the surface impact-fracture changed from smooth to a rougher surface as fibre content increased from 0 to 40 phr.

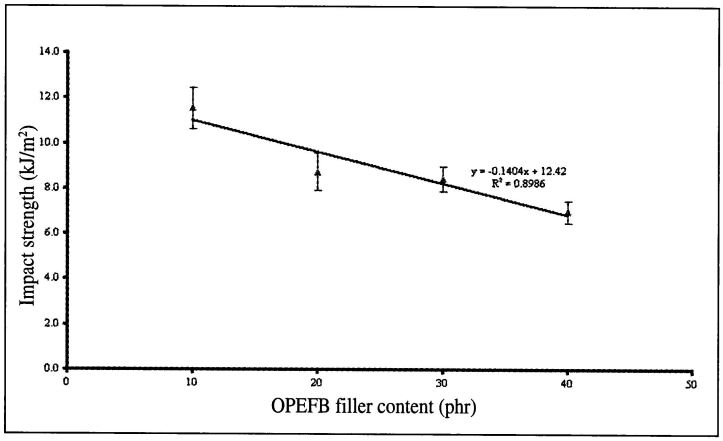

Figure 2. *Effects of OPEFB filler content and 9 phr CPE on the impact strength of filled composites*

(Abu Bakar *et al.* 2005). In other words, the ductile portion contributed by PVC-U matrix automatically reduced, thus the failure mode of impact-fractured surface of PVC-U compound was transformed from a relatively less brittle to a more brittle one as the OPEFB fibre content increased. Secondly, it is known that the composite materials with satisfactory mechanical properties could only be achieved if there is a good dispersion and wetting of the fibres in the matrix that will give rise to a strong interfacial adhesion. However, this is not the case when OPEFB fibres are used in PVC. Although the polarity of OPEFB fibres is able to form a physical interaction with the polar PVC (Woodhams *et al.,* 1984), it is a relatively weak interaction compared to the fibre-fibre interaction caused by hydrogen bondings. Therefore, OPEFB fibres have a greater tendency to agglomerate among themselves into fibre bundles, which, consequently, lowers the area of contact with the matrix.

Our previous study has shown that the impact strength for PVC-U (without any filler and impact modifier) was 8 kJ/m^2 and reduced to 5.7 kJ/m^2 at 40 phr OPEFB filler content (Abu Bakar *et al.,* 2005). Based on the impact strength values given in Figure 2, the addition of 9 phr CPE impact modifier has improved the impact strength of filled composites at all filler content. This result indicates that the CPE has the ability to compensate for the detrimental effects caused by the filler with lowering the yield stress of PVC-U matrix by allowing shear yielding rather than fracture when subjected to the sudden load (Mengeloglu, 2001). Through this mechanism the composites were able to suppress the brittle failure or catastrophic failure. However, with increasing of OPEFB filler content from 10 to 40 phr, the increment of impact strength (Table 4) between the unmodified and CPE-impact modified OPEFB-filled composites became smaller. This is due to the fact that at higher filler content, the efficiency of CPE to improve the impact strength was reduced. The similar trend was also observed in our previous study when 9 phr acrylic was added in the OPEFB-filled PVC-U composites (Abu Bakar *et al.,* 2005).

Table 4. Impact Strength of OPEFB-Filled Composites and CPE-Impact Modified OPEFB-Filled Composites

OPEFB filler content (phr)	Impact strength (kJ/m²)		Impact strength increment (%)
	CPE content (phr)		
	0	9	
0	8.0	99.6	
10	7.1	11.5	62
20	6.8	8.7	28
30	6.1	8.4	38
40	5.7	7.0	23

Flexural Properties

The flexural modulus of unmodified OPEFB-filled PVC-U composites increased as the filler content increased from 0 to 40 phr (Figure 3). However, the flexural strength (Figure 4) was linearly decreased. These results are consistent with the effect of OPEFB filler content on flexural modulus and flexural strength of unmodified PVC composites and acrylic impact-modified PVC composites previously reported (Abu Bakar *et al.,* 2005).

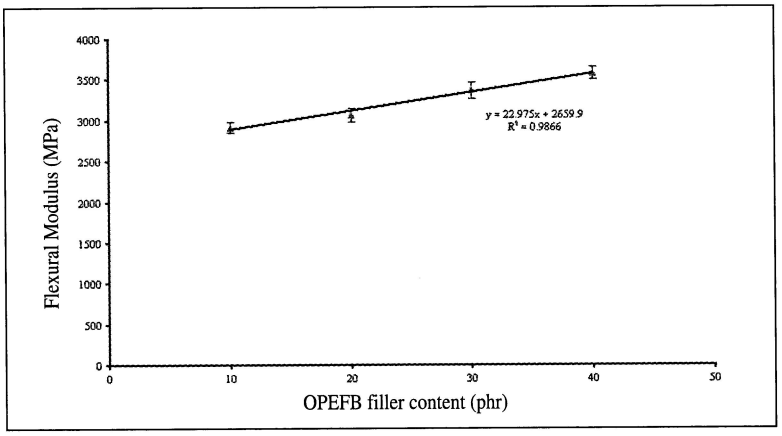

Figure 3. *Effects of OPEFB filler content and 9 phr CPE on the flexural modulus of filled composites*

The ability of OPEFB fillers to restrict the local chain motions of the PVC chains by reducing the free volume of PVC increased the flexural modulus of filled composites. Meanwhile, the agglomeration of OPEFB fillers into filler bundles decreased the flexural strength. This was due to the inability of OPEFB filler bundles to support the stresses transferred from the PVC-U matrix.

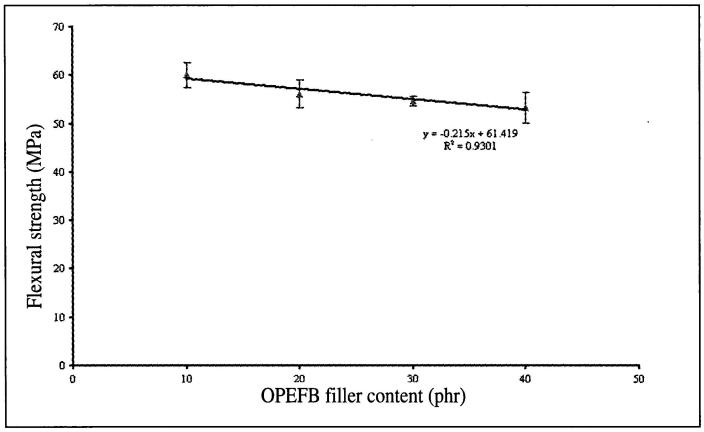

Figure 4. *Effects of OPEFB filler content and 9 phr CPE on the flexural strength of filled composites*

The flexural modulus and flexural strength of the CPE-impact modified OPEFB-filled composites (Figure 3 and Figure 4) are lower than OPEFB-filled composites reported earlier (Abu Bakar *et al.,* 2005). The softening effect generated by the rubbery phase of CPE, which lowered the yield stress, thereby increasing the energy absorption, had consequently lowered the stiffness and strength of PVC-U matrix. Similar result was also reported by other researcher (Lutz, 1993) who found that the flexural properties of PVC diminished almost linearly with the incorporation of the impact modifier.

CONCLUSION

The flexural modulus improved while impact and flexural strength of OPEFB-filled composites decreased with the addition of OPEFB. The fusion time of the CPE modified OPEFB-filled PVC-U composites decreased upon the addition of 10 and 20 phr of OPEFB but increased drastically on further increase to 30 and 40 phr.

ACKNOWLEDGEMENTS

The authors would like to express their thanks to the Ministry of Science, Technology and Innovation of Malaysia for funding this work under an IRPA project (Vot 72197). The authors would also like to thank Industrial Resin Malaysia (IRM), Sdn. Bhd., Sabutek Sdn. Bhd. and Dupont Dow Elastomers (Exodus Sdn. Bhd. Malaysia) for materials support throughout this work.

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