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IMPACT PROPERTIES OF OIL PALM EMPTY FRUIT BUNCH FILLED IMPACT MODIFIED UNPLASTICISED POLY (VINYL CHLORIDE) COMPOSITES

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Abstract. Oil palm empty fruit bunch (EFB) reinforced composite is an emerging area in polymer technology. EFB is a low cost filler with low density, high specific properties as well as being non-abrasive. This type of composites offers several advantages comparable to those of conventional fiber composites. The main objective of the investigation reported in this paper is to study the effect of EFB filler loading and impact modifiers, namely chlorinated polyethylene (CPE) and acrylic on the mechanical properties of EFB filled-PVC-U composites. In preparing the samples, the PVC-U resins and the additives were initially dry blended using a laboratory blender before being milled into sheets using a two-roll mill. Test specimens were prepared using a hot press and impact tested using a pendulum type machine according to Izod mode. The results from the impact test showed that the unfilled PVC-U samples of both impact modifiers changes from brittle to ductile mode with increasing impact modifier concentration. The incorporation of EFB into unmodified PVC-U and modified PVC-U has resulted in the reduction of impact strength. As the EFB filler content increases from to 10 to 40 phr, impact strength reduction of about 40% and 30% was observed for acrylic-modified PVC-U and CPE-modified PVC-U, respectively. The impact strength reduction was only marginal for unmodified PVC-U composites. The detrimental effect of fillers on the impact performance was due to the volume taken up. Fillers unlike the matrix are incapable of dissipating stress through the mechanisms known as shear yielding prior to fracture. The impact modifier was found to be effective in enhancing the impact strength of EFB-filled PVC-U composites. However, the effectiveness decreases with increasing filler loadings. The impact strength of CPE-modified PVC-U was higher than acrylic-modified PVC-U at filler loading at 20 phr and higher.

Keywords: oil palm empty fruit bunch, poly (vinyl chloride), impact modifier, impact strength, composites

Abstrak. Komposit bertetulang tandan kosong buah sawit (EFB) merupakan bidang yang terkini muncul dalam teknologi polimer. EFB ialah pengisi berkos rendah serta ketumpatan rendah, bersifat tentu yang tinggi dan tidak bersifat menghakis. Oleh itu komposit jenis ini mempunyai pelbagai kebaikan berbanding dengan komposit bergentian. Objektif utama kajian dalam kertas kerja ini adalah untuk mengetahui kesan penambahan pengisi EFB dan pengubahsuai impak, iaitu polietilena berklorin (CPE) dan akrilik terhadap sifat mekanikal komposit PVC-U berpengisi EFB. Semasa penyediaan sampel, PVC-U dan additif pada mulanya telah diadun kering dengan menggunakan pengadun sebelum disebatikan menjadi kepingan dengan menggunakan penyebati dua-pengguling. Alat penekan panas telah digunakan untuk menghasil spesimen untuk ujian impak dengan menggunakan alat jenis pendulum dalam mod Izod. Keputusan daripada ujian impak sampel PVC-U tanpa pengisi untuk kedua-dua pengubahsuai impak telah menunjukkan perubahan dari mod rapuh kepada mod mulur dengan bertambahnya kepekatan pengubahsuai impak. Penambahan pengisi EFB ke dalam

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PVC-U tanpa terubahsuai dan PVC-U terubah suai telah mengakibatkan pengurangan kepada kekuatan impak. Apabila EFB bertambah daripada 10 sehingga 40 phr, pengurangan kekuatan impak adalah 40% untuk PVC-U berpengubahsuai impak akrilik dan 30% untuk PVC-U berpengubahsuai CPE. Hanya sedikit pengurangan kekuatan impak untuk komposit PVC-U tanpa terubah suai. Kesan kekurangan dari pengisi terhadap prestasi impak adalah disebabkan ketidakmampuan pengisi untuk melepaskan tegasan melalui mekanisme yang dikenali sebagai alahan ricihan sebelum patah. Pengubahsuai impak telah didapati berkesan untuk meningkatkan kekuatan impak komposit PVC-U berpengisi EFB. Bagaimanapun, keberkesanannya berkurangan dengan pertambahan pengisi. Kekuatan impak PVC-U berpengubahsuai CPE adalah lebih tinggi dari pada PVC-U berpengubahsuai akrilik pada pengisi muatan 20 phr dan lebih.

Kata kunci: Tandan buah kelapa sawit, poli(vinil klorida), pengubahsuai impak, kekuatan impak, komposit

1.0 INTRODUCTION

Modification of materials by incorporation of fillers has been a popular research activity in the plastics industry since the properties of plastics materials may be significantly changed by the introduction of fillers. Although the use of natural organic fillers is not as popular as inorganic fillers, the awareness of researchers to the advantages of natural organic fillers such as lower density, less abrasiveness to processing equipment, lower cost, no health hazard as well as being an abundant renewable resource, have gradually attracted them using these kind of fillers for plastics modification [1].

Currently, the incorporation of lignocellulosic material as a reinforcing component or filler in the thermoplastic composites has received much intention. One of the lignocellulosic materials of great relevance in Malaysia is the large quantity of biomass generated by oil palm industries. The potential utilization of fillers derived from the oil palm industries, such as empty fruit bunch (EFB), for production of thermoplastics polymer filled composites have been reported by several workers. Zaini *et al.,* [2] has shown that for all filler sizes, the mechanical properties of PP/Oil Palm wood flour composites decreased with increasing filler content. It was also reported that the incorporation of EFB into PP matrix has resulted in the improvement in tensile modulus, but the tensile strength, elongation at break, and impact strength decreased with increasing filler loading [3]. Meanwhile the incorporation of compatibiliser and coupling agent has improved the tensile and impact strength of the composites. In another study, the modification of EFB with anhydride has improved the mechanical properties of EFB /polyester composites [4].

One of the most important aspects in the modification properties of thermoplastics by using fillers is to achieve a good combination of properties and processability at a moderate cost. As far as mechanical properties are concerned, the main target is to strike a balance of stiffness, strength and toughness [5]. The incorporation of filler into thermoplastics results in stiffness enhancement but may reduce the impact strength or toughness. On the contrary, the improvement of toughness is also required. The incorporation of rubbery impact modifiers in thermoplastic will, in general, result in a significant improvement in toughness at the expense of stiffness and strength.

Acrylic has been recommended as an impact modifier especially to modify PVC-U as it gives consistently high toughness under a wide processing temperature and shear. This is so, because the dispersed rubber particles maintain their particle morphology in the continuous PVC phase. The mechanism of impact modification with chlorinated polyethylene is completely different to that in acrylic system. Instead of a distinct dispersed phase, it forms a network enclosing PVC primary particles in order to develop the desired impact strength. However, PVC-U modified with chlorinated polyethylene is dependent on processing conditions when with increased temperature or shear, the network starts to collapse and toughness decreases [6].

Although EFB has been useful as filler in thermoplastics, no study has yet been reported using PVC. PVC is a versatile thermoplastic, has good mechanical, chemical and weathering properties. It is therefore being increasingly used in outdoor applications as a replacement for conventional materials such as wood, aluminum, and steel [6].

The objective of this paper is to investigate the effect of EFB filler loadings, types and various concentrations of impact modifiers including acrylic (KM355P) and chlorinated polyethylene (CPE702P) on the impact strength of the EFB filled impact modified PVC-U composites.

2.0 EXPERIMENTAL PROCEDURE

2.1 Material

The PVC suspension resin (MH-66) with K-value 66 and oil palm EFB filler were purchased from local vendors. The PVC blend formulations are shown in Table 1, which were based upon typical commercial PVC window frame formulation with some modifications.

2.2 Sample Preparation

The Restsch shaker was used to separate the EFB fillers into different sizes. The size of the filler used in this study did not exceed 75 mm. The separated EFB fillers were oven dried for 24 hours at 105°C prior to blending, in order to achieve less than 5% moisture content. The dry blending of PVC-U and additives was done using high-speed mixer for 10 minutes. Then the dry-blended PVC compounds were sheeted using a laboratory two-roll mill at temperature of 165°C for 10 minutes of milling time. The milled sheets were then placed into a mould and hot pressed at the temperature and pressure of 180°C and 120 kg/m^2, respectively, for 5 minutes. The cooling time used was 5 minutes before the specimens were removed from the mould.

Table 1 Blend formulation

Resin	**phr**
Poly(vinyl chloride)- PVC	100.0
Additives	
Tin Stabilizer	2.0
Calcium Stearate	0.5
Stearic Acid	0.6
Acrylic Polymer	1.5
Titanium Oxide	4.0
Filler	**phr**
Oil Palm Empty Fruit Bunch (EFB)	0, 10, 20, 30, 40
Impact Modifier	**phr**
Acrylic	0, 3, 6, 9, 12
Chlorinated Polyethylene (CPE)	0, 3, 6, 9, 12

2.3 Testing

The impact strength assessment was done using IMPats pendulum (Izod) tester. A pendulum hammer with an angle of 90° and velocity of 3.0 m/s was used. Impact specimens were notched (45°) using a Davenport notch cutting apparatus. Test bars were prepared according to ISO 180-1982 (E) and tested at room temperature. All the reported values for the impact test were the average values of seven specimens.

3.0 RESULTS AND DISCUSSION

3.1 Impact Strength

Table 2 shows that for the unfilled samples, the impact strength increases with increasing impact modifiers content from 0 phr (unmodified) to 12 phr. All the samples containing impact modifier 3 phr and less were observed to break completely. There were mixtures in the failure mode for the samples with 6 phr. However at 9 phr, the samples undergone hinge breaks where the samples did not break completely. It shows that the brittle-ductile transition occurs at around 6 phr for both the unfilled CPE and acrylic impact modified PVC-U samples. From the number of unbroken samples at 6 phr and impact strength values at 3 phr, it can be deduced that acrylic impact modifiers are more effective than CPE at lower impact modifier content.

Previous papers have discussed the difference between the toughening mechanism of acrylic and CPE modifiers. Lutz [7] suggested that chemical modifiers operate by somewhat different mechanisms. To explain the toughening mechanism, the impact modifiers are divided into two categories that is predefined elastomer (PDE) and not predefined elastomer (NPDE).

Acrylic is categorized as PDE impact modifier and is sometimes called discrete particles modifiers (DPM). The PDE term is perhaps the most descriptive because it says that the modifier particle has been designed rather than just being discrete. The PDE are usually designed to have a rubbery core surrounded by shell composed of monomer to confer compatibility with PVC. While the rubber is responsible for impact modifying performance, the shell with higher glass temperature is important to enhance isolation as fine free-flowing powder granules and to facilitate the dispersion of the modifier in the PVC. Particle size of acrylic does not change even after incorporation into PVC compound and similar observation has been reported by Robinovic [6]. He observed that the toughness of acrylic modified PVC-U was independent of the processing time and temperature. This is because the dispersed rubber particles maintain their particle morphology in the continuous PVC phase as reported by other researchers [6,7].

CPE is categorised as NPDE and is sometimes referred to as network polymers (NP), a term that defines its operational mode rather than polymeric structure. CPE can be made more compatible with PVC by manipulating the chlorine content and distribution. Deanin [8] found that the blend formulation containing CPE with 36% chlorine was most efficient for processibility, impact and strength. Being incipiently compatible and classically amorphous, CPE has variable morphology in PVC depending on processing temperature, shear and time [6,9]. Impact strength development of CPE/PVC melts is dependent on maintaining PVC primary particles bound by a network of CPE. Around 150°C, this network has not developed and beyond 200°C, melting of PVC primary particles results in the CPE converting into spherical globs dispersed in the PVC matrix resulting in poor impact strength. Therefore, in this study, temperature of 165 and 180°C were used as milling and hot press temperature respectively during specimen preparation in order to obtain the possible good impact strength of unfilled samples as shown in Table 2. However, the impact

Table 2 Effect of impact modifiers concentrations on impact strength of unfilled modified PVC-U

Impact Modifier (phr)	Acrylic		CPE	
	Impact Strength kJ/m^2	Completely Broken (no. of specimen)	Impact Strength kJ/m^2	Completely Broken (no.of specimen)
0	8.04	7	8.04	7
3	19.65	7	18.55	7
6	93.13	3	48.60	5
9	109.53	0	99.60	0
12	109.03	0	119.58	0

strength values of acrylic-modified PVC-U are higher than CPE-modified PVC-U. Similar result on impact has also been found by Deanin.

From Table 3, it can be seen that the impact strength decreases upon the incorporation of the EFB fillers. The impact strength dropped drastically to about one tenth of the original values for the impact modified samples with 9 phr and 12 phr impact modifiers.

Table 3 Effect of EFB and impact modifier contents on the impact strength of EFB filled-impact modified PVC-U composites

EFB (phr)	Impact Strength (kJ/m^2)									
	Acrylic(phr)					CPE(phr)				
	0	3	6	9	12	0	3	6	9	12
10	7.11	9.00	10.68	10.85	12.66	7.11	7.75	10.97	11.54	12.04
20	6.79	7.05	7.66	8.39	9.27	6.79	7.40	7.79	8.73	9.41
30	6.04	6.53	6.71	7.22	8.03	6.04	7.17	6.95	8.41	9.18
40	5.72	5.74	5.89	6.32	6.54	5.72	6.36	6.85	6.96	7.72

Figures 1 and 2 show that upon increasing of oil palm EFB fillers loading, the impact strength of PVC-U/EFB composites for both the unmodified and impact modified samples decreases linearly. This result is expected for the filled polymer systems and has been reported by other researchers [1,2,10]. As the filler content increases from 10 to 40 phr, impact strength reduction of about 40% and 30% is observed

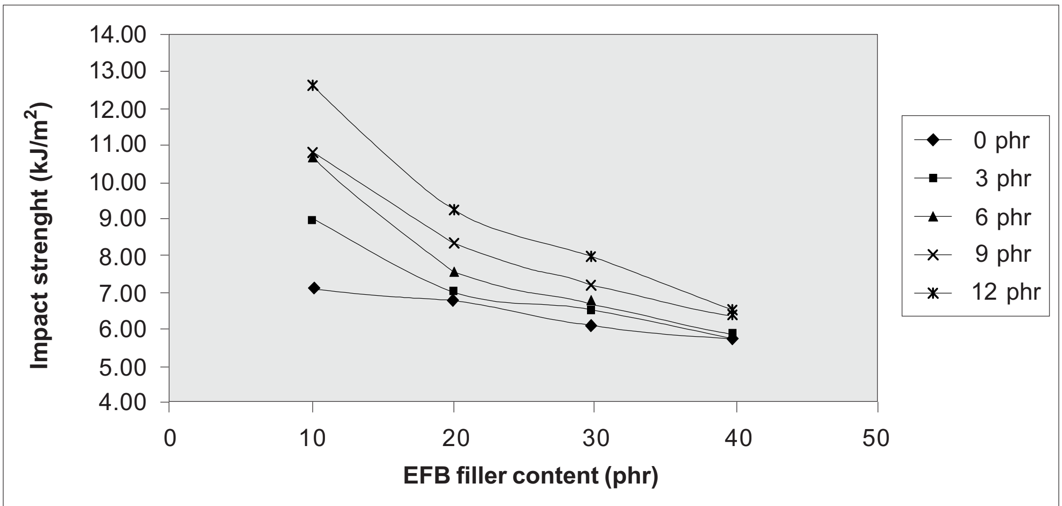

Figure 1 Effect of filler loading and impact modifier concentration on the impact strength of EFB filled-acrylic-modified PVC-U composites

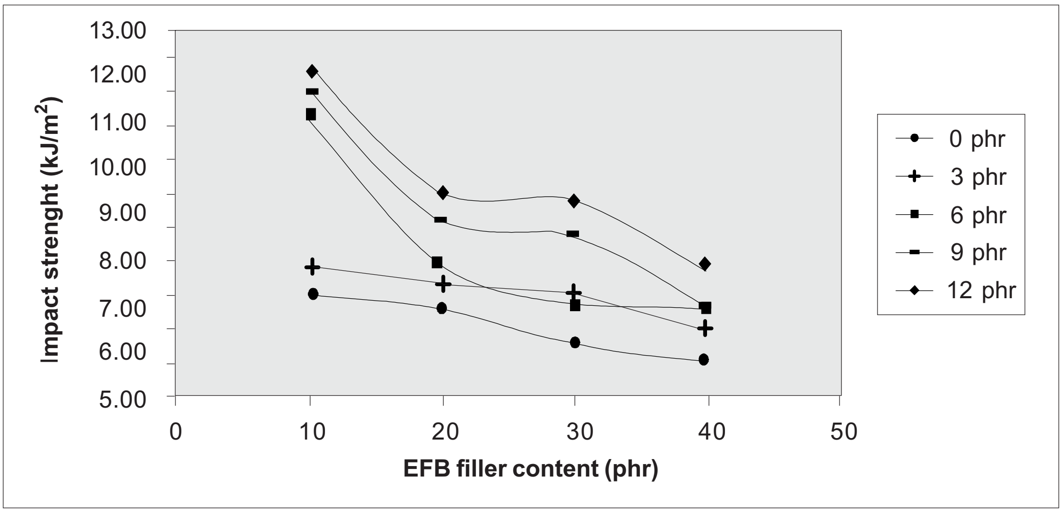

Figure 2 Effect of filler loading and impact modifier concentration the impact strength of EFB filled-CPE-modified PVC-U composites

for acrylic-modified PVC-U and CPE-modified PVC-U, respectively. However, for the unmodified samples, the percentage reduction in impact strength is slightly less.

This detrimental effect of fillers on the impact performance is due to the volume they take up. Fillers unlike the matrix are incapable to dissipate stress through the mechanism known as a shear yielding prior to fracture. Therefore, the total ability of the material to absorb energy is decreased. Incorporation of fillers may also hinder the local chain motions of the polymer molecules that enable them to shear yield, thereby lowering the ability of composites to absorb energy during fracture propagation. Besides poor wetting of the fillers by PVC-U might cause the impact strength to decrease because of poor interfacial adhesion between filler and polymer matrix. Figure 3 clearly shows the occurence of filler pullout and debonding. This indicates the lack of adhesion between fillers and PVC-U matrix, a major reason for the poor impact strength. During the impact test, this adhesion in the interfacial regions becomes the potential sites for crack growth as inability of the fillers to support the stress transfers to the polymer matrix. The increase of filler content increases the agglomeration of filler and the interfacial regions, which exaggerates the weakening of the resulting composites to crack propagation. These filler agglomerates can be observed by SEM (Figure 4) to be distributed unevenly throughout the matrix. The polarity of EFB filler, due to hydrogen bondings, these fillers have greater tendency to agglomerate among themselves into filler bundles.

The results from Table 3 also show that the impact strength of the modified composites improved with increasing impact modifier concentrations. The impact modifier has the capability to compensate for the detrimental effect caused by the filler with lowering the yield stress of PVC-U by allowing shear yielding rather than

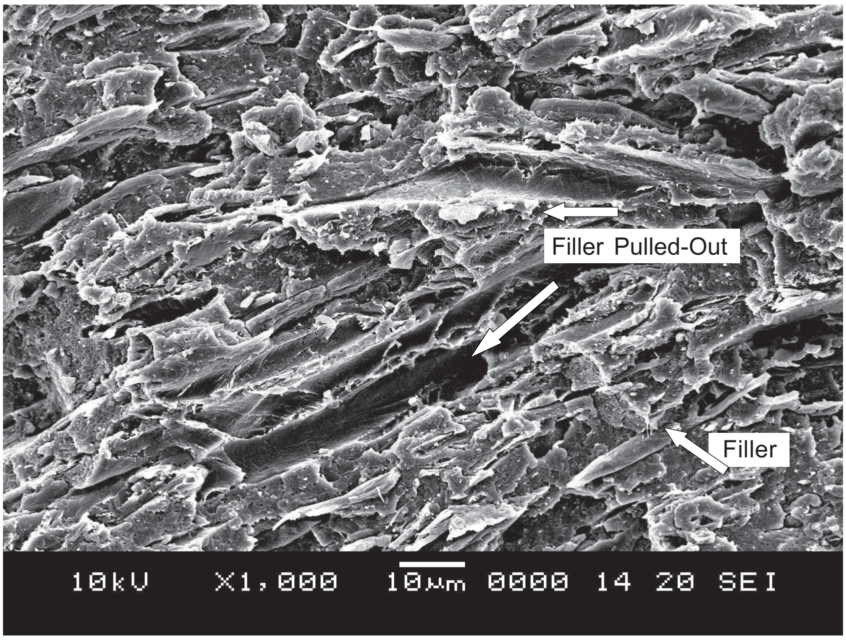

Figure 3 SEM micrographs of the impact fracture surface of EFB-filled – unmodified PVC-U composite with filler loading of 30 phr (magnification 1000X)

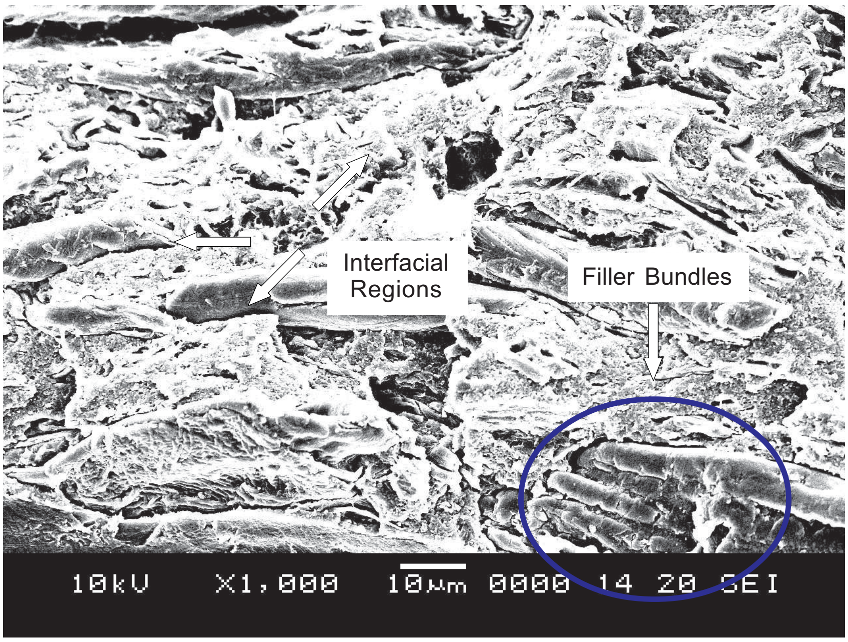

Figure 4 SEM micrographs of the impact fracture surface of EFB-filled –Acrylic modified PVC-U composite with filler loading of 30 phr (magnification 1000X)

fracture when subjected to the sudden load. Through this mechanism, the composites are able to suppress brittle or catastrophic failures; therefore, the improvement of impact strength with increase in the concentration of impact modifier is expected. It is interesting to observe the effectiveness of the impact modifier in enhancing, whilst the impact strength decreases with the increasing filler content. For the acrylic modified samples with filler loading of 10 phr, an increase of acrylic impact modifier from 0 to 12 phr resulted in an increase of 40% in impact strength. However, for the acrylic modified samples with filler loading of 40 phr, similar increase of acrylic impact modifier resulted only in 10% increase.

The impact strength of CPE-modified PVC-U is higher than acrylic-modified PVC-U at filler loadings of 20 phr and higher. This shows that the CPE is the preferred impact modifier at higher filler loading for more effective impact strength enhancement. Gerlach [11] also reported similar advantage of CPE. The plausible reason for this observation is the adhesion between EFB filler and CPE-modified PVC-U is better than between EFB filler and acrylic-modified PVC-U. It can be elucidated in terms of polarity between hydroxyl (OH) groups on the EFB filler surface and chlorine (Cl) atoms of PVC-U. The addition of CPE modifier, which contains 36% of Cl atoms, into the PVC-U increases the Cl content and polarity of CPE-modified PVC-U. Because of that, physical interaction between CPE-modified PVC-U with OH groups on the EFB filler surface increases and thus produces better wetting of filler surface. This, results in improvement of filler-matrix adhesion.

4.0 CONCLUSION

The main objective of this investigation is to study the effect of filler and impact modifier loadings on the impact property of EFB filled-modified PVC-U composites. The results from the impact test showed that the unfilled PVC-U samples of both impact modifiers changes from brittle to ductile mode with increasing impact modifier concentration. Acrylic is more effective than CPE at lower loading that is at 3 and 6 phr. The incorporation of EFB into unmodified PVC-U and modified PVC-U has resulted in the reduction in the impact strength. The impact strength reduces further with increasing EFB content. The detrimental effect of fillers on the impact performance was due to the volume taken up. Fillers unlike the matrix are incapable to dissipate stress through the mechanism known as shear yielding prior to fracture. The impact modifier is effective in enhancing the impact strength of EFB-filled PVC-U composites. However the effectiveness decreases with increasing filler loadings. The impact strength of CPE-modified PVC-U is higher than acrylic-modified PVC-U at filler loading at 20 phr and higher. This shows that the CPE is the preferred impact modifier at higher filler loading for more effective impact strength enhancement.

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