

UTILIZATION OF AGRICULTURAL WASTES AS FILLERS IN THERMOPLASTICS COMPOSITES.

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Abstract

The concept of cost reduction in plastics by use of fillers has been known throughout the ages. By the appropriate selection and optimization of such materials, not only the economic but other properties such as processing and mechanical behaviour can be improved. The growing interest in environmentally friendly materials has greatly increased the interest in the use of natural fillers derived from agricultural sources in plastics composites. These natural fillers have the advantages of low densities, very low cost, non-abrasiveness, higher filling levels, recyclability, biodegradability and renewable nature. As the field of plastics develops and expands, a variety of naturally occurring materials are explored such as wood flour, empty fruit bunch (EFB) and rice husk (RH). This paper reports the recent development and current work on the utilization of EFB and RH as fillers in thermoplastics composites. The study has shown that the incorporation of RH and EFB fillers has resulted in the increase of flexural modulus indicating a improvement in stiffness. The flexural modulus increase further with increasing both of RH and EFB loadings. However, impact strength decreases with increasing filler loadings. The impact modifier is effective in enhancing the impact strength of both EFB and RH filled PVC-U composites.

Key words: PVC-U, rice husk, empty fruit bunch, acrylic impact modifier, mechanical properties.

Introduction

The utilization of fillers from various sources into thermoplastics has been an accepted route to achieve enhancement in material properties or cost saving possibilities. Calcium carbonate (CaCO_3), talc and kaolin are examples of mineral fillers which are widely used in plastics industry. However, the use of natural fillers derived from agricultural sources, such as jute, oil palm empty fruit bunch (EFB), wood flour, rice husk (RH) [1,2] has rapidly expanded in recent years. There is a tremendous potential for future growth in this area. These natural fillers have the advantages of low densities, very low cost, non-abrasiveness, higher filling levels, recyclability, biodegradability and renewability [3,4].

In Malaysia, RH is one of the biomass materials, which is a by-product from the rice industry. RH contains cellulose 35%, hemicellulose 25%, lignin 20% and ash 17% (silica 94%), by weight [5]. The Application of RH includes fuel in heat generation for drying rice, use in making cement, and as a fertilizer in agriculture. However, the industrial applications of this material is limited. The rest is burned or used for landfilling. Therefore, more efficient utilization of rice husk is urgently needed. One of the efforts is to produce value-added products such as composite materials from this important bio-sources. Utilization of RH offers some economical and environmental advantages too. There has been considerable effort and interest in the addition of RH to thermoplastics [6,7].

Fuad et al. incorporated two types of fillers namely white rice husk ash (WHRA) and black rice husk ash (BRHA) into PP matrix using twin screw compounder. Of the two fillers, WRHA is rich from silica with approximately 96% (w/w) silica while BRHA contains only about 56% (w/w). Both types of fillers exhibited their capability to strengthen the PP matrix (enhance the tensile modulus) at the expense of some other mechanical properties [1].

EFB filler derived from the palm oil industry has proven its applicability as a reinforcement in polymers. Abdul Khalil has succeeded in utilizing extracted EFB fiber as a reinforcement in

polyester resin and resulting composites have shown excellent mechanical performance [8]. Meanwhile, Rozman had found that the treated EFB fibers enhanced the mechanical properties of Polypropylene. Rozman revealed that the incorporation of compatibilizer and coupling agent produced PP/EFB composites with improved tensile and impact properties [9].

PVC resin is one of the most worldwide used thermoplastics. Nowadays, PVC products are widely used in automobile industry and construction sectors. Besides that, PVC is also popular as electrical insulator and packaging material for the electronics and food industry. The capability of PVC to perform such diverse functions is due to the ability of PVC to incorporate various additives to suit the numerous applications [10]. The other factors responsible for the increase of utilization of PVC is relatively cheap, can be processed by a variety of techniques, good physical, chemical and weathering properties.

However, unmodified PVC-U has the disadvantage of being prone to occasional brittleness and is notch sensitive. Impact modifier is therefore, one of the important additives used to enhance the impact properties of PVC. The impact modifier, which in the rubbery phase, is capable of facilitating the yielding of the PVC matrix before failure when a sudden load is applied. Several types of impact modifiers are currently available which include core-shell acrylic and chlorinated polyethylene (CPE). The core-shell acrylic has been recommended as an impact modifier to modify rigid PVC as it gives consistently high toughness under a wide processing temperature and shear. This is, because the dispersed rubber particles maintain their particle morphology in the continuous PVC phase.

This paper reports the recent development and current work on the utilization of EFB and rice husk as fillers in thermoplastics composites. The aim of the study is to determine the effects of the two natural fillers on stiffness and toughness. One of the most important aspects in the materials development of engineering thermoplastics 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 and toughness.

Material and Methods

The PVC suspension resin (MH-66) with K-value 66 was purchased from Industrial Resin Malaysia (IRM) Berhad, Tampoi Johor. Oil Palm Empty Fruit Bunch filler was purchased from Sabutek Sdn.Bhd, Teluk Intan Perak and Rice Husk filler was supplied by Bernas Perdana Sdn. Bhd, Seberang Perai, Pulau Pinang. The PVC blend formulations are shown in Table 1, which were based upon typical commercial PVC window frame formulation with some modifications.

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 a temperature of 165°C for 10 minutes milling time. The milled sheets were then placed into a mould and hot pressed at a temperature and pressure of 180°C and 120kg/m², respectively, for 5 minutes. A cooling time of 5 minutes was used before the specimens were removed from the mould.

The impact strength assessment was done using a pendulum tester model ATA FAAR for the EFB filled PVC composites and Toyo Seiki for RH. The flexural properties assessment was done using a universal testing machine model Instron for the EFB filled PVC composites and Lloyds for RH.

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
EFB or RH	0, 10, 20, 30, 40
Impact Modifier	phr
Acrylic	0, 3, 6, 9, 12 0, 4, 8, 12 [#]

For EFB filled PVC composites.

[#] For RH filled PVC composites.

RESULT AND DISCUSSION

Table 2 shows the effect of RH and EFB loadings on flexural modulus of unmodified and impact modified PVC. The result shows that the incorporation of RH and EFB has resulted in an increase of flexural modulus indicating an improvement in stiffness. This trend is consistent with another study on the effect of rice husk ash on the modulus of PVC-U composites [11].

Table 2: Effect of Fillers on Flexural Modulus of Unmodified and Impact Modified PVC

Filler Content (phr)	EFB		RH	
	Unmodified (MPa)	Impact Modified (9 phr) (MPa)	Unmodified (MPa)	Impact Modified (8 phr) (MPa)
0	3065	2653	3357	2574
10	3588	2880	3774	2949
20	3768	3104	3949	3525
30	3864	3347	4467	3897
40	4299	3496	4907	4098

It is found that there is a considerable increase in flexural modulus with increasing RH content whereby 40 phr RH is 46%, higher compared to unfilled unmodified PVC. A similar trend was obtained for the effect of RH loading on flexural modulus of impact modified PVC. For the similar increase in RH content, flexural modulus increase by nearly 60. The overall results shows that impact modified samples have lower values compared to the unmodified. It is interesting to observe that at 20phr RH of impact modified PVC composites, the flexural modulus values is slightly higher than the blank samples (unfilled unmodified PVC). For the impact modified samples, the addition of 40 phr RH increased the flexural modulus by 59 % higher compared to unfilled impact modified PVC.

Addition of EFB filler into unmodified and modified PVC-U increased the flexural modulus as expected. For the unmodified samples PVC-U composites, the increment of modulus is about 40% with the incorporation of 40 phr. For a similar increase in filler loading, the modulus increase only by 32 % for the impact modified.

Table 3 shows the effect of RH and EFB loadings on impact strength of unmodified and impact modified PVC. The result shows that the incorporation of RH and EFB has resulted in a decrease of impact strength for both samples. This result is expected for the filled polymer systems and has been reported by other researchers [9,11,12].

Table 3: Effect of Fillers on Impact Strength of Unmodified and Impact Modified PVC

Filler Content (phr)	EFB		RH	
	Unmodified (kJ/m ²)	Impact Modified (9 phr) (kJ/m ²)	Unmodified (kJ/m ²)	Impact Modified (8 phr) (kJ/m ²)
0	8.8	10.9	4.9	11.1
10	7.1	10.9	3.7	5.5
20	6.8	8.4	3.6	5.1
30	6.1	7.2	3.1	3.8
40	5.7	6.3	3.0	3.3

There is a relatively sharp decrease in impact strength for unmodified PVC upon the addition of 10 phr RH followed by a gradual decrease upon increasing filler loading. A similar trend of impact strength result is observed upon addition of 10 phr RH of impact modified PVC. Impact strength decrease about 50% at 10 phr RH and decrease with further increase in filler loading. It is interesting to observe that the addition of impact modifier has enhanced the impact strength of PVC-U samples at all levels of RH content. The effectiveness of the impact modifier, however, decreases with increasing RH content.

The impact strength of PVC-U composites for both the unmodified and impact modified samples decreased upon increasing of EFB fillers content. For EFB filled PVC-U composites, as the filler content increased from 0 to 40 phr, impact strength reduction of about 30 % was observed.

The reduction of impact strength is due to two factors. The first factor is due to the detrimental effect of filler. This effect on the impact performance is due to the volume they take up. Fillers unlike the matrix are incapable of dissipating stress through the mechanisms known as a shear yielding prior to fracture. Therefore, the total ability of the material to absorb energy is decreased.

Secondly, the 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 that, poor wetting of the fillers by PVC-U matrix might cause the impact strength to decrease because of poor interfacial adhesion between filler and polymer matrix. 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. Increasing the filler content increases the agglomerations of filler and the interfacial regions that exaggerates the weakening of the resulting composites to crack propagation. The polarity of EFB filler, due to hydrogen bonds, these fillers have greater tendency to agglomerate among themselves into filler bundles.

The addition of acrylic impact modifier improved the impact strength of PVC-U composites. The acrylic has the capability to compensate for the detrimental effect 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. Through this mechanism, the composites are able to suppress brittle or catastrophic failures; therefore, the improvement of impact strength with the addition of acrylic impact modifier is expected. As the filler content increased, the impact strength values tends to come close to the values of unmodified composites due to the predominant of fillers in the PVC-U matrix. Thus the difference between the impact strength values of both composites gets smaller.

Conclusion

The main objective of this investigation was to study the effect of RH and EFB filler loadings on the flexural and impact properties of PVC-U. The results from the flexural test showed an increase in flexural modulus upon addition of RH and EFB fillers. The incorporation of RH and EFB into unmodified PVC-U and impact modified PVC-U has resulted in the reduction in the impact strength. The acrylic impact modifier is effective in enhancing the impact strength of the filled PVC-U composites. However the effectiveness decrease with increasing filler loadings.

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