Pertanika J. Sci. & Technol. 20 (2): 415 - 423 (2012)

SCIENCE & TECHNOLOGY

Journal homepage: http://www.pertanika.upm.edu.my/

Effects of Composition Parameters on Tensile and Thermal Properties of Abaca Fibre Reinforced High Impact Polystyrene Composites

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ABSTRACT

The properties of fibre-reinforced composites are dependent not only on the strength of the reinforcement fibre but also on the distribution of fibre strength and the composition of the chemicals or additives addition within the composites. In this study, the tensile properties of abaca fibre reinforced high impact polystyrene (HIPS) composites, which had been produced with the parameters of fibre loading (30,40,50 wt.%), coupling agent maleic anhydride (MAH) (1,2,3 wt%) and impact modifier (4,5,6 wt.%) were measured. The optimum amount of MAH is 3% and the impact modifier is 6% and these give the best tensile properties. Meanwhile, Differential Scanning Calorimetry (DSC) was used to study the thermal behaviour within the optimum conditions of the composites. In this research, glass transitions temperature (Tg) of neat HIPS occurred below the Tg of the optimum condition of composites as the temperature of an amorphous state. The endothermic peak of the composites was in the range of 430-435^{0}C, including neat HIPS. It was observed that enthalpy of the abaca fibre reinforced HIPS composites yielded below the neat HIPS of 748.79 J/g.

Keywords: Tensile properties, Differential Scanning Calorimetry (DSC), abaca fibre, Maelic Anhydride (MAH), impact modifier, High Impact Polystyrene (HIPS)

Article history:
Received: 28 July 2011
Accepted: 13 January 2012

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INTRODUCTION

Generally, some types of polymers have been used as matrices for natural fibre composites

ISSN: 0128-7680

including polyethylene (PE), polystyrene (PS) and polypropylene (PP). These polymers have a different affinity towards the fibre due to the difference in their chemical structures. Joseph *et al*. (1996) reported that sisal/LDPE (low density polypropylene) composites released a better reinforcing effect because of the high matrix ductility and high strength/modulus ratio of sisal as compared to that of the LDPE matrix. The properties of the composites depend on those of the individual components and on their interfacial compatibility. Since thermoplastics such as polyethylene (PE), polystyrene (PS) and polypropylene (PP) are hydrophobic and have poor miscibility, chemical addition was needed to improve and facilitate the interaction between thermoplastics and reinforcing filler. Lee *et al*. (2005) and Lei *et al*. (2006) reported that the miscibility between nanoclay and PE or PP could be improved by the addition of compatibilizers such as MAPE, MAPP or carboxylated PE.

The majority of natural fibres, as a function of cellulose fibres and lignin, have low degradation temperatures (~200°C), making them inadequate for processing temperature above 200°C (Pracella *et al*., 2006). Nair *et al*. (2001) reported that the *Tg* values of polystyrene composites reinforced with short sisal fibres are lower than that of the unreinforced PS and may be attributed to the presence of some residual solvents in the composites. To solve the processing of natural fibre composites, it is necessary to promote polymer modification with polar groups (such as maleic anhydride, stearic acid or glycidyl methacrylate) to enhance the adhesion between the matrix and the composite components. The coupling agent more often used for this application is a polyethylene copolymer grafted with maleic anhydride (Keener *et al*., 2004). In this paper, the investigation of tensile properties resulted in an optimum condition of composites, meanwhile thermal behaviour within the optimum condition from abaca fibre reinforced HIPS composites at glass transition and crystallization processes was clearly observed. Commercial HIPS were used for a comparator to the natural fibre composites. The DSC methods were used in evaluating the basic thermal parameters of the optimum condition of abaca fibre reinforced HIPS composites.

EXPERIMENTAL

Materials

Abaca (*Musa textilis Nee*) fibres are produced by Ridaka Hand Craft, Pekalongan, Central Java, Indonesia. High impact polystyrene (HIPS), Idemitsu PS HT 50, density 1.04 g/cm^3, and melt index of 4.0g/10 min were obtained from Petrochemical (M) Sdn. Bhd., Malaysia. Maleic anhydride (MAH), (polystyrene-block-poly(ethylene-ran-butylene)-block-polystyrene-graft-maleic anhydride) was supplied by Sigma Aldrich Malaysia (M) Sdn. Bhd., Malaysia. Impact modifier, a styrene butadiene styrene (SBS) copolymer rubber (Cyclo resin), was supplied by PT. Wahana Makmur Kencana, Jakarta, Indonesia.

Formulation of the Samples

The composites in this research were formulated with the response surface methodology – Box-Behnken design (BBD). Tensile properties are the responses of three factors to be determined and these three factors are designated as X_1 (abaca fibre), X_2 [maelic anhydride (MAH)] and

X_3 [impact modifier (IM)] within three levels, which were coded as +1, 0, −1 for high, intermediate and low values, respectively, as shown in Table 1.

Table 1: Levels and code of variables for Box Benhken design (BBD)

Variables	Symbol		Coded Levels		
	Uncoded	Coded	-1	0	+1
Abaca	X_1	x_1	30	40	50
MAH	X_2	x_2	1	2	3
IM	X_3	x_3	4	5	6

METHODS

Composite Fabrication

The abaca fibres were dried under the sunlight between 27 and 30^{0}C for four days. The dried abaca fibres were cut into 2 – 3 mm by means of an electronic cutting machine. Based on the proportion of the abaca fibre, maelic anhydride (MAH) and impact modifier were incorporated into the neat HIPS. The processing of the abaca fibre reinforced HIPS composites were accomplished using a rolling machine, as shown in Fig.1. The working temperature of the rolling machine was kept approximately 200^{0}C while the speed was also maintained at the slow rate. The process was continued until all the materials were well-mixed and they produced the sheets of abaca fibre reinforced HIPS composites with an average of 1mm thickness.

Fig.1: Producing abaca fibre reinforced HIPS composites using a rolling machine

Tensile Test

Tensile testing of the specimens was performed according to ASTM D 638-98 on a universal test machine (Instron, model 556) at ambient temperature (27^{0}C). The strain rate was 50 mm/min with a gauge length of 60 mm. The values reported were the average of the three samples tested.

Differential Scanning Calorimetry (DSC)

The characterization of a material requires the use of Differential Scanning Calorimetry analysis. The DSC analysis obtained quantitative and qualitative data concerning the net heat changes as a thermal behaviour. The samples used for the DSC analysis were cut from the sheet of the composite in order to have a weight that ranges from 10-14 mg. A Mettler-Toledo DSC model 822 was used to determine the thermal behaviour. The temperature was programmed for heating from 25°C to 500°C, with a heating rate of 10°C min^{-1} under nitrogen atmosphere.

RESULTS AND DISCUSSION

Tensile Strength

The properties of the matrix and fibres, as well as processing conditions, are very important in achieving good mechanical properties of the composites. The strength parameter is more sensitive to the matrix properties, whereas the modulus is dependent on the fibres' properties. The positive or negative effect of the application of natural fibres reinforced matrix composites is important. Meanwhile, the variables of filler or additive will improve the mechanical performance of the matrix composites. This research on the abaca fibre reinforced high impact polystyrene clearly indicates the relationship between dependent and independent variables of the tensile strength and tensile modulus which were studied in the experimental design. The effects of the response variables are demonstrated using the model in Equations 1 and 2 for tensile strength, as follows:

$$\begin{aligned}\hat{y} = 11.23 - 0.025X_1 + 0.43X_2 + 0.071X_3 - 1.15X_1^2 + \\ 0.35X_2^2 + 1.80X_3^2 + 0.47X_1X_2 + 1.79X_1X_3 + 0.93X_2X_3\end{aligned} \tag{1}$$

and for the tensile modulus, model equation is:

$$\begin{aligned}\hat{y} = 1.25 + 5.345x10^{-3}X_1 + 0.05X_2 + 4.44x10^{-3}X_3 - 0.16X_1^2 - \\ 0.026X_2^2 + 0.12X_3^2 + 0.029X_1X_2 + 0.16X_1X_3 + 0.092X_2X_3\end{aligned} \tag{2}$$

By contour plots, the effects of each variable with their interactions on the abaca fibre reinforced HIPS composites at one fixed level of variable (medium level) impact modifier are shown in Fig.2 and Fig.3.

The plots described in Fig.2 indicate that the tensile strength of abaca fibre reinforced HIPS composites to the saddle point curve and that two high values (the maximum of up to 12.28 MPa) at the area of abaca loading which are close to 45 wt% and 30 wt%. The graph of the tensile strength starts to decrease but then increases when the loading of the abaca fibre is close to 45 wt% and 30 wt%. In this condition, the impact modifier deals with the area close to 4.5 wt% and 5.5 wt%. This response exposes the minimum point as the effect of the impact modifier (IM) and Maelic Anhydride (MAH), while the abaca fibre is loading in level (40 wt. %).

The case of tensile modulus was studied based on the interaction effects; the abaca fibre and impact modifier also reflected the saddle point graph effect illustrated in Fig.3. The plot describes that the minimum yield of tensile modulus reflects the interaction between the impact modifier (IM) and maelic anhydride that is close to area 1.16 GPa. Upon studying the coefficient values of the tensile strength and tensile modulus, the variable X_1 (β_1 = 0.1837 and 0.0312) was found to be higher than the variables X_1 and X_3, indicating that it contributed the most in predicting the properties of the abaca fibre reinforced HIPS composites. Meanwhile,

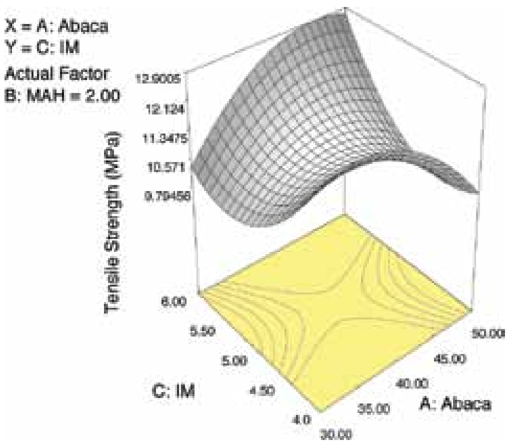

Fig.2: Response surface 3D plots showing the effect of abaca fibres and Impact Modifier for tensile strength (GPa)

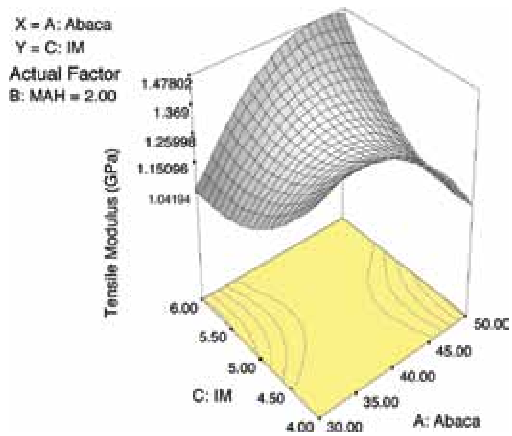

Fig.3: Response surface 3D plots showing the effect of MAH and Impact Modifier for Tensile modulus (GPa)

the interaction between X_1 and X_3 provides a more real impact for the yields. It means that abaca fibres are highly influenced by impact modifier (IM).

The Optimum Condition of the Composites and the DSC Analysis

Optimum conditions were solutions within the applications due to tensile and thermal properties of the abaca fibre reinforced HIPS composites. Discussion on the account of this particular situation was necessary to identify a compromise zone where the experimental responses satisfied the specifications to achieve the proposed aims. In order to choose the best coordinates of an acceptable compromise, desirability function was taken on. The acceptable values of desirability function were the values closed to one (100%). In this research, the tensile properties of the abaca fibre reinforced HIPS composite compromised on the parameters of the abaca fibre (36.76 wt.%), maelic anhydride (3 wt%) and impact modifier (4 wt.%), and this condition dealt with 77% desirability. Table 2 shows the predicted optimization experiments of the tensile properties as prepared by BBD that referred to the actual parameters of composites.

The reviewed optimization of the composites presented in Table 3 explains the condition thermal properties, tensile properties and desirability function. This research predicted the parameter of abaca fibre, maleic anhydride and impact modifier compiled optimum of the tensile properties within desirability function and summarized by composites A, B and C.

Fig.4 shows the DSC scan of the abaca fibre reinforced HIPS composites. The compositions of the abaca fibre reinforced HIPS composites (A, B, and C) may influence the energetic terms correlated with the difference of *Tg*. The glass transition (Tg) occurs over a wide temperature range (Thirta et al., 2005) as shown in Table 3 and depicted in Fig.4; Tg is determined by the region of the onset and midpoint temperature. The specimen of the abaca fibre reinforced HIPS composites with the differences of composition at 1 wt.% and 3 wt.% of maleic anhydride, respectively, resulted in a difference of *Tg*. The difference of *Tg* was ~ 34^{0}C, and thus, it summarized that maleic anhydride should play a dominant role in controlling the amorphous

Table 2: Predicted optimization experiments of that tensile strength as prepared by Box-Behnken design (BBD) referred to the actual composition of composites

No	Abaca	MAH	IM	Tensile Strength	Tensile Modulus	Desirability
	wt%	wt%	wt%	MPa	GPa	
1	40	3.00	4.00	12.288	1.314	0.77
2	40	3.00	6.00	12.876	1.512	0.77
3	40	3.00	4.00	12.201	1.306	0.76
4	40	1.00	6.00	12.840	1.264	0.64
5	40	1.00	6.00	12.842	1.270	0.64
6	40	1.00	4.00	13.306	1.431	0.61
7	40	1.00	4.00	13.268	1.426	0.61
8	40	1.00	4.00	13.263	1.425	0.61

MAH: Maleic Anhydride; IM: Impact Modifier

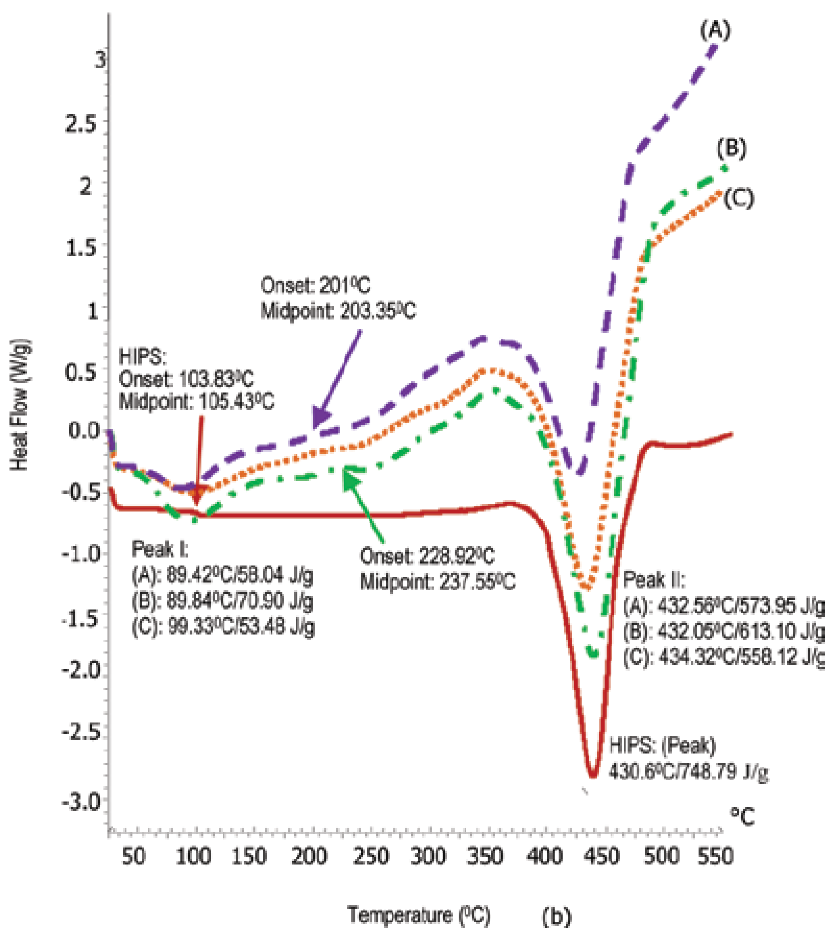

Fig.4: The DSC scan of the optimum condition of abaca fibre reinforced HIPS composites (compositions A, B, and C)

Table 3: Results of the tensile and thermal (DSC analysis) properties for the abaca fibre reinforced HIPS composites (formulations A, B and C) within the desirability function

Composition of	THERMAL PROPERTIES						MECHANICAL PROPERTIES		
	Glass Transition Tg (°C)		Endoterm Temp (°C)		Entalphy (J/g)		Tensile Strength	Tensile Modulus	Desirability
	Onset Temp	Mid-point Temp	Peak I	Peak II	ΔH Peak I	ΔH Peak II	MPa	GPa	
Abaca reinforced HIPS composites									
(A) with composition							13.306	1.431	0.61
Abaca (40 wt.%)	201.00	203.35	89.42	432.56	58.04	573.95	13.268	1.426	0.61
Maleic anhydride (1 wt.%)							13.263	1.425	0.61
Impact modifier (4 wt.%)									
Abaca reinforced HIPS composites									
(B) with composition							12.288	1.314	0.77
Abaca (40 wt.%)	228.92	237.55	89.84	432.05	70.90	613.10	12.201	1.306	0.76
Maleic anhydride (3 wt.%)									
Impact modifier (4 wt.%)									
Abaca reinforced HIPS									
composites (C) with composition									
Abaca (40 wt.%)	-	-	99.33	434.32	53.48	558.12	12.786	1.512	0.77
Maleic anhydride (3 wt.%)									
Impact modifier (6 wt.%)									
High Impact Polystyrene	103.83	105.43	-	430.60	-	748.79	-	-	-

or glass transition state of the abaca fibre reinforced HIPS composites. The composition (C) of the abaca fibre reinforced HIPS composites was less free and the molecules underwent crosslinking. When compared with neat HIPS, the *Tg* of the abaca fibre reinforced HIPS composites (A and B) increased by 97.92^{0}C and 132.12^{0}C, respectively.

The heating thermograms of the abaca fibre reinforced HIPS composites (A, B, and C) represent the one peak correlated with the glass transition state and another peak in the endothermic due to crystallization state. The first peak of the abaca fibre reinforced HIPS composites were on 89.42^{0}C (A), 89.84^{0}C (B), and 99.33^{0}C (C), respectively, as shown in Table 3. This peak normally attributed to the release of the absorbed moisture related to humidity on the surface from the fibres. As the natural fibres are hydrophilic, a water desorption peak was observed around 100^{0}C (Huda & Drzal, 2008). In this study, the highest enthalpy of the abaca fibre reinforced HIPS composites (B) at the transition temperature was 70.90 J/g. Based on the highest enthalpy, the enthalpy increased by 12.86 J/g by adding 2 wt% of the maleic anhydride but decreased by 17.48 J/g by adding 2 wt% impact modifier.

The second endothermic peak of the abaca fibre reinforced HIPS composites in the range of 430-435^{0}C in the DSC curves (Fig.4) determined the dehydration of the composites. An endothermic of HIPS was also in the same range. It gave the indication that the heating rates (polymer degradation) of the composites and neat HIPS felt within the same range of temperatures. Taking composition B as a reference, with enthalpy (613.10 J/g) of abaca fibre reinforced HIPS composites, the enthalpy of (A) increased by 39.15 J/g and (C) and decreased by 54.98 J/g, respectively.

CONCLUSION

By utilizing the Box-Benhken design of experiments, the MAH and impact modifier have been shown to improve the tensile strength and tensile modulus properties of the composites by enhancing the adhesion between the fibres and HIPS. In this study, the predicted parameters of the abaca fibres were ~40 wt%, MAH (~3 wt.%) and Impact Modifier (~6 wt%) represented for the optimum tensile properties due to desirability function. Through DSC scan, the first peak of the composites (A, B, C) revealed that the absorbed moisture release to humidity on the surface of the fibres to be below ~100^{0}C, whereas the *Tg* of optimum conditions (A, B, and C) of the abaca fibre reinforced HIPS composites were above of neat HIPS. Scanning by DSC also resulted in the peak temperature of endothermic in the range between 430-435^{0}C, including neat HIPS. The second enthalpy of the abaca fibre reinforced HIPS composites yielded less neat HIPS of 748.79 J/g.

ACKNOWLEDGEMENTS

The authors are thankful to Universitas Malahayati, Lampung, Indonesia for the financial support. The authors also wish to acknowledgement the assistance from PT. Tara Plastic Indonesia and Radiation Technology, and Malaysian Nuclear Agency, Bangi, Malaysia for providing and testing materials for this study.

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