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Full Length Research Paper

Study on abaca (*Musa textilis* Nee) fibre reinforced high impact polystyrene (HIPS) composites by thermogravimetric analysis (TGA)

E. H. Agung[1,4]*, S. M. Sapuan[1], M. M. Hamdan[2], H. M. D. K. Zaman[3] and U. Mustofa[4]

[1]Department of Mechanical and Manufacturing Engineering, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia.
[2]Faculty of Engineering, Universiti Pertahanan Nasional Malaysia, Kem Sungai Besi 57000, Kuala Lumpur, Malaysia.
[3]Radiation Technology, Malaysia Nuclear Agency, Bangi, 43000 Kajang, Malaysia.
[4]Faculty of Engineering, Universitas Malahayati, Kemiling, 35127 Bandar Lampung, Indonesia.

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In this research, thermogravimetric analysis (TGA) was used to measure the amount and rate of change in the weight (weight loss) of abaca fibre reinforced high impact polystyrene (HIPS) composites as a function of temperature. The function determined the composition of abaca fibre reinforced HIPS composites on predicting the thermal stability. The optimum composites designed with composition of abaca fibre 40 wt%; maleic anhydride 1 and 3 wt%, impact modifier 4 and 6 wt% respectively. This paper studied the thermal characteristic of abaca fibre, reinforced HIPS composites as compared to the neat HIPS. The measurements were carried out in temperatures ranging from 25°C to 600°C at heating rate 20°C min^{-1} and nitrogen gas flow of 50 ml min^{-1}. The results from TGA analysis have shown that the combination among the coupling agent maleic anhydride, impact modifier and abaca fibre has improved the thermal stability of composites.

Key words: Thermogravimetric analysis (TGA), abaca fibre, maleic anhydride (MAH), impact modifier, high impact polystyrene.

INTRODUCTION

Thermal analysis is an important and very useful method to characterize any materials such as thermoplastic or thermosetting polymer matrix and also to determine the influence of natural fibres addition into the polymers (George et al., 2004; Luz et al., 2008). One of the accepted methods for studying the thermal properties of polymeric materials is the thermogravimetric analysis (TGA). Thermogravimetric analysis (TGA) provides some information about the thermal stability and decomposition rate of materials and measures weight changes in materials as a function of temperature or time in order to quantify reactions involving gaseous emissions (Villain et al., 2007; Reis et al., 2008). Thermogravimetric analysis (TGA) involves heating a material up to and past its thermal degradation temperature at a controlled rate and monitoring mass loss during the heating process. Thermal analysis is also used as an analytical method in understanding the structural property relationships and thermal stability of composite materials, such as the incompatibility between fibres and polymer matrices.

The incompatibility of components affected deteriorating performance for thermal and mechanical properties. Stress transfer from the matrix to the fibre depends on fibre-fibre and fibre-matrix interactions, cellulose fibres and lignin were important terms on the properties of the composites. The majority of natural fibres as a function of cellulose fibres and lignin, have low degradation temperatures (~200°C), which make them inadequate for processing temperatures above 200°C (Pracella et al.,

*Corresponding author. E-mail: efriyo@yahoo.com, sapuan@eng.upm.edu.my.

2006). 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) in order to enhance the adhesion between matrix and the composite components. The coupling agent more often used for this application is apolyethylene copolymer grafted with maleic anhydride (Keener et al., 2004). Thermal analysis of cellulose fibres and the effects of crosslinking, orientation and crystallinity on the natural fibres as a function of cellulose fibres have been reported by some researchers (Basch and Lewin (1973, 1974), Rodrig et al. (1975). Another previous study reported by Hassan and Nada (2003), investigated performance of the thermal properties of several lignocellulosic fiber reinforced polyester composites where the natural fibers used were rice, straw, bagasse and cotton stalk which showed that all the lignocellulosic fiber reinforced polyester composites were slightly thermally less stable than the neat polyester composite. While, an increasing degradation temperature of the samples with rubber-wood fibre was also studied and reported by Sameni et al. (2003).

High impact polystyrene (HIPS) is produced by the polymerization of styrene in the presence of rubber, commonly known as polybutadiene (BR). BR is added to provide the extensibility, toughness and impact resistance needed in certain applications.

Current research issues (Lee et al., 2003; Antich et al., 2006) are focused on the development of performance HIPS or natural fibre reinforced HIPS composites including chemical resistant, flame retardant grade and mechanical properties. Other research evaluated functions of catalyst addition in HIPS (Marcilla et al., 2007). This research described characteristics of natural fibre reinforced HIPS composites in optimum condition composition (wt% of abaca fibre, maleic anhydride and impact modifier) which were in transition temperatures compared to neat HIPS. Abaca (Musa textile Nee) is a family of banana with familiar name of Manila hemp and a member of the genus Musa plant, which comes from pseudo-stems; yields a strong and resilient fibre. Abaca fibre was widely used in the production of handicrafts, manufacture of marine ropes and in the fishing industry due to its resistance to humidity and salty water (Sharrock, 1998). Nowadays, innovation of abaca fibre reinforced composites implicated in under floor protection for passengers and was patented by Daimler Chrysler's researchers (Knothe et al., 2000).

MATERIALS AND METHODS

Abaca (*Musa textilis*) fibres were obtained from Ridaka Hand Craft, Pekalongan, Central Java, Indonesia. High impact polystyrene (HIPS), Idemitsu PS HT 50, density 1.04 g/cm3, and melt index 4.0 g/10 min was 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 styrene butadiene styrene (SBS) copolymer rubber (Cyclo resin) was supplied by PT. Wahana Makmur Kencana, Jakarta - Indonesia.

Formulation of the samples

All composites in this work were formulated with 40 wt% of abaca fibre. When the chemical additive, Maleic anhydride (MAH) and impact modifier were used, the proportion of the MAH as a coupling agent was 1 and 3 wt%, while the proportion of the impact modifier were 4 and 6 wt% respectively (Table 1).

The formulation of abaca fibre reinforced HIPS composites (A, B, and C) were prepared by optimum condition technique (Box Behnken Design) as reported in the previous paper (Agung et al., 2011).

Composite processing

The abaca fibres were dried under the sun light between 27 to 30°C for four days. The dry abaca fibres were cut into 2 to 3 mm by means of an electronic cutting machine. Based on the proportion of abaca fibre, Maleic anhydride (MAH), and impact modifier which were incorporated into the neat HIPS. The processing of abaca fibre reinforced HIPS composites were accomplished using a rolling machine as shown in Figure 1. The working temperature of the rolling machine was kept at approximately 200°C with speed kept in a slow rate. The speed of the first cylinder roll was on 6.0 m/min and the second cylinder roll was on 10.8 m/min. The process was continued until all the materials were well-mixed and produced sheets of abaca fibre reinforced HIPS composites with an average thickness of 1 mm.

Thermogravimetric analysis (TGA)

The samples used for the thermogravimetric analysis were cut from the sheet of composite in order to have a weight of 7 to 20 mg. After weighing, the samples were dried in an oven for 1 h at 100°C. The TGA equipment (Mettler Toledo SDTA 851 analyser) was programmed for heating from 25 to 600°C with a heating rate of 20°C min-1, under a nitrogen flow of 50 ml/ min-1.

RESULTS AND DISCUSSION

The characterization of a material requires the use of thermal analysis. Thermogravimetric analysis (TGA/DTG) analyses the moisture content (1st step), polymer content (2nd step) and the residue (ash) at the final temperature (3rd step). TGA analysis is a description of a change in thermal stability which is examined in terms of percentage weight loss as a function of temperature. Derivative thermogravimetric (DTG) is a type of thermal analysis in which the rate of material weight changes upon heating versus temperature is plotted. DTG is used to simplify the recorded weight loss versus temperature.

This research used the thermogravimetric (TG) curves to release the thermal degradation and thermal stability of optimum composition of abaca fibre reinforced HIPS composites then compared with neat HIPS. The TG and DTG analysis are presented in Figures 2 and 3. TGA was performed with temperature rang offing from 25 to 600°C.

Table 1. Formulation of the composites in weight percentage (wt%).

Sample	Proportion in weight percentage (wt%)			
	HIPS	ABACA	MAH	IM
HIPS	100			
HIPS/ABACA/MAH/IM (A)	55	40	1	4
HIPS/ABACA/MAH/IM (B)	53	40	3	4
HIPS/ABACA/MAH/IM (C)	51	40	3	6

Figure 1. The production of abaca fibre reinforced HIPS composites by rolling machine.

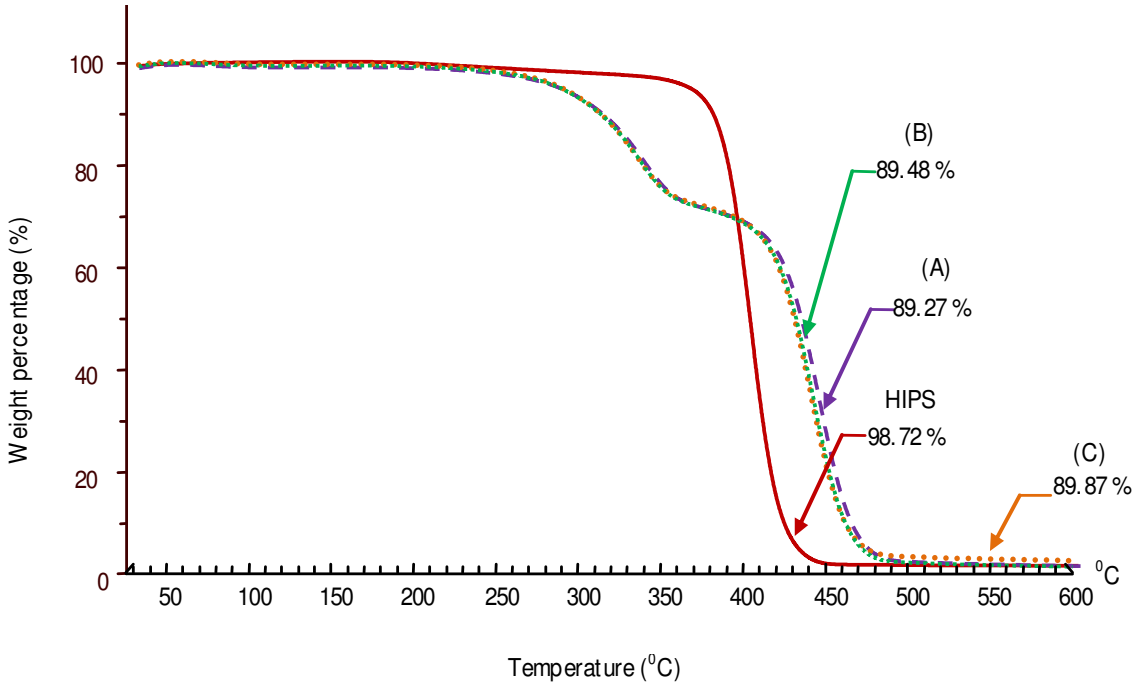

Figure 2. The TG analysis of HIPS, abaca fibre reinforced HIPS composites with formulation (A), (B), and (C).

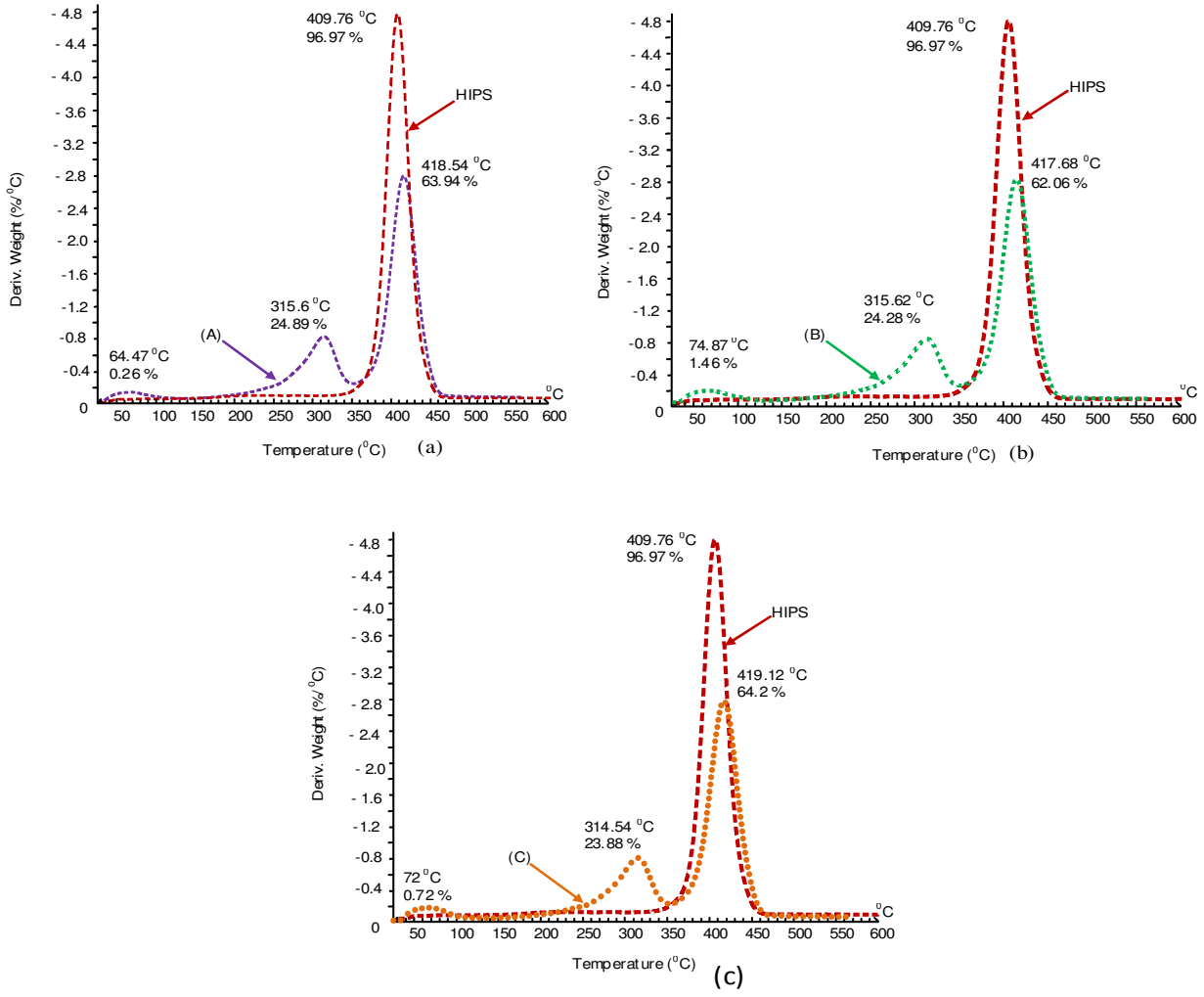

Figure 3(a, b, c). DTG curve of HIPS, abaca fibre reinforced HIPS composites with formulation (A), (B), and (C).

The neat HIPS consisted of one stage of weight loss process and had a transition temperature from 348.88℃ and the final transition at 468.83℃. This stage released the peak transition temperature of HIPS at 409.76℃ with weight loss of 96.97% and residue was 1.28% (Table 2). This result is in agreement with previous studies which observed that the thermal decomposition of HIPS occurred in one single stage, between 369 to 490℃ with its maximum temperature transition at 433℃ and had a residual weight of 1.2% (Vilaplana et al., 2007). The other relevant issue of TGA investigation of neat polystyrene (PS) reported that its decomposition started at 288℃ and had four stages of weight loss process (Nair et al., 2001).

The thermal stability of abaca fibre reinforced HIPS composites with three formulations (A), (B), and (C) noticed that after a small initial drop in weight loss with temperatures ranging from 34.73 to 36.75 to 103.55 to 143.99℃ respectively (Table 2), characterizing a first constant stage. This initial drop in weight for natural material of abaca fibre reinforced HIPS composites (formulation (A), (B), and (C)) are normally attributed to the release of absorbed moisture or the vaporization of water related to the humidity on the surface from the fibres. The interaction between hydrogen bonding allowed the thermal energy to be distributed over many bonds and is a major source of stability in cellulose (Andrzej et al., 2010). This was correlated with dehydration of the abaca fibre. Figure 2 shows that after the first constant stage, a sloping decrease in weight then occured until the approximate temperature for each composition (A), (B), and (C) of abaca fibre reinforced HIPS composites is 353.24, 351.92 and 349.88℃ in temperatures respectively (Table 2). This sloping decrease can be associated with the decomposition of the abaca fibres constituents including lignin, cellulose and hemicelluloses. With increasing temperature, a second constant stage in all TGA curves of formulation (A), (B), and (C) from abaca fibre reinforced HIPS composites exist up to the limit of 600℃ established for the analysis (Figure 2). The total weight loss for each formulation (A), (B), and (C)

Table 2. Results of TG Analysis for abaca fibre reinforced HIPS composites with formulation (A, B and C).

Composition of sample	Number of transition	Initial decomposition temp. (℃) (Ti)	Maximum decomposition temp. (℃) (Tm)	Final decomposition temp. (℃) (Tf)	Weight lost at transition (%)	Residual weight (%) at 600℃
Abaca reinforced HIPS composites, (A) with composition:	1	34.73	64.47	103.55	0.26	
Abaca (40 wt%) Maelic Anhydride (1 wt%)	2	218.63	315.60	353.24	24.89	10.73
Impact Modifier (4 wt%)	3	353.24	418.54	472.35	63.94	
Abaca reinforced HIPS composites, (B) with composition:	1	36.75	74.87	143.99	1.46	
Abaca (40 wt%) Maelic Anhydride (3 wt%)	2	211.38	315.62	351.92	24.28	10.52
Impact Modifier (4 wt%)	3	356.96	417.68	468.83	62.06	
Abaca reinforced HIPS composites, (C) with composition:	1	35.65	72.00	118.19	0.72	
Abaca (40 wt%) Maelic Anhydride (3 wt%)	2	209.22	314.54	349.88	23.88	10.13
Impact Modifier (6 wt%)	3	355.67	419.12	469.19	64.20	
High Impact Polystyrene	1	348.88	409.76	468.83	96.97	1.28

of abaca fibre reinforced HIPS composites were 89.27, 89.48 and 89.87% respectively. The minor differences of total weight-loss are interpreted as constituents of abaca fibre, maleic anhydride and impact modifier that contributed to thermal decomposition of abaca reinforced HIPS composites.

Figure 3 (a, b, c) shows the DTG curves for abaca fibre reinforced HIPS composites. From Table 2, the first 3 peaks were at the 64.47, 74.87, and 72℃ for formulation (A), (B), and (C) of abaca fibre reinforced HIPS composites respectively. The peaks confirm that the release of humidity of each formulation (A), (B) and (C) of abaca fibre reinforced HIPS composites were affected by a distinct atmosphere. The weight percentage addition of maleic anhydride of the composites that were studied in formulations (A) and (B), increased degradation temperature by 10.4℃ at the first peak. On the other hand, the weight percentage addition of impact modifier studied between formulations (B) and (C) decreased the degradation temperature by 2.87℃. The weight loss of the first peaks were found at 0.26% (A), 1.46% (B), and 0.72% (C) from abaca fibre reinforced HIPS composites respectively. Meanwhile, a previous study showed that the starting decomposition temperatures of abaca fibre were defined at 2% weight loss (Andrzej et al., 2010).

In the same review (Table 2), the second peaks were at 315.60, 315.62, and 314.54℃ respectively for formulations (A), (B), and (C) of abaca fibre reinforced HIPS composites. The DTG curve at the second peaks of abaca fibre reinforced HIPS composites released a minor difference of peaks temperature, these peaks indicated that maleic anhydride and impact modifier did not induce a reaction on abaca fibre reinforced HIPS composites (Figure 2 (a, b, c). This condition also clearly showed that the second peaks had a more effective degradation effect to the constituents of the fibres. A reference explained the second peaks of weight loss are associated to decomposition of cellulose and thermal depolymerization of hemicelluloses (Chu, 1970).

A relevant aspect of the third peaks was where the peaks correspond to a maximum rate of decomposition of abaca fibre reinforced HIPS composites. From DTG curve Figure 2 (a, b, c), it was observed that the weight loss of abaca fibre reinforced HIPS composites (formulation A, B, and C) were 63.94, 62.06, and 64.2% which were found at 418.54, 417.68, and 419.12℃ respectively. These peaks described minor differences in weight loss and that peak temperatures among formulations (A, B, and C) had occurred. Compared to the neat HIPS, weight loss of neat HIPS (96.97%) had a more contrasting difference with all abaca fibre reinforced HIPS composites (A, B, and C) formulations. The peak temperatures of abaca fibre reinforced HIPS composite for the formulations (A, B, and C) were lower than neat HIPS. Lower temperature peaks indicated a complex thermal degradation process related to the rupture of molecular chains with different energy levels. The research also described that weight percentage composition of maleic

anhydride and impact modifier affected the process of thermal degradation significantly. The weight percentage addition of maleic anhydride of the composites that were studied in formulations (A) and (B), decreased degradation minor temperature by 0.86°C at the third peak. Also studied in the peaks from formulations (B) and (C) of abaca fibre reinforced HIPS composites, increased the temperature by 1.44°C when weight percentage addition of impact modifier was done. This phenomenon indicated characteristics of weight loss in abaca fibre reinforced HIPS composites. Therefore, weight percentage composition of maleic anhydride and impact modifier had the potential to affect the process of turning thermal decomposition of the composites to ash.

Another result described in Table 2 was the values of the residues. Residual weight of HIPS and abaca fibre reinforced HIPS composites (formulations A, B, and C) were 1.28, 10.73, 10.52, and 10.13% respectively. There are larger significant differences between neat HIPS and abaca fibre reinforced HIPS composites (A, B, and C). This is apparently an indication of another effective loss of degradation rate, the faster degradation rate attributed to lower residual weight. The formulations of abaca fibre reinforced HIPS composite inhibited the rate of decrease of thermal stability. Therefore, it can be concluded that the thermal stability of the composites had potential to be more flexible in its application. In this research, thermal stability of abaca fibre reinforced HIPS composites had correlated with weight loss transition, interpretation of maximum decomposition temperature, and residual weight loss summarized in formulation (B); abaca 40 wt%, maleic anhydride 3 wt%, and impact modifier 4 wt% was better than the formulation A and C.

Conclusions

The thermal stability of formulation for the optimum design of abaca fibre reinforced HIPS composites (A, B, and C) has been successfully studied and experimentally compared to neat HIPS. The char yield of abaca fibre reinforced HIPS composites was found to be higher than neat HIPS. The residual weight of abaca fibre reinforced HIPS composites and neat HIPS were 10.73, 10.52, 10.13, and 1.28% respectively. Three stages of structural decomposition of abaca fibre reinforced HIPS composites (formulation A, B, and C) displayed maximum weight loss associated with clear peaks at (first stage) 64.47, 74.87 and 72°C, (second stage) 315.60, 315.62, and 314.54°C, (third stage) 418.35, 417.68 and 419.12°C, and one stage for neat HIPS 409.76°C. Composition of maleic anhydride and impact modifier has the potential to influence thermal degradation to correlate with the total weight loss of all compositions (A, B, C) of abaca fibre reinforced HIPS composites. Lower temperature peaks of abaca fibre reinforced HIPS composite indicate a complex thermal degradation process related to the rupture of molecular chains with different energy level and had potency to more flexible in its application.

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