The Effect of the Jute Fabric Reinforced Vinyl Ester Composite Treated with Ammonium Polyphosphate on the Physical, Flammability and Thermal Properties

Khairul Naseem Mohd Rodzi, Mohd Iqbal Misnon, Ahmad Faiza Mohd, Muhammad Ismail Abdul Kadir , Mohamad Faizul Yahya

Abstract*: *Natural fibre composite is potentially used as low load bearing material in building infrastructure field (light weight application). Nevertheless, the ability of materials against thermal and fire must be good. In this work, the incorporation of the phosphorus based intumescent flame retardant (IFR), ammonium polyphosphate (APP) into the jute fabric composite (JC) with different formulations were fabricated to analyze its fire-retardant properties. The physical, flammability and thermal properties of the untreated and treated composites were investigated with the support by FESEM and EDX analysis. Based on the results, the density of treated JC was higher than untreated sample due to the impregnation of APP. In terms of fire-retardant properties, the JC samples treated by APP were able to improve the thermal and flammability of the composites. The enhancement of fire-retardant properties of JC can improve its potential to be used as low load bearing material in building infrastructure applications.

Keywords: Ammonium polyphosphate (APP), fire retardant properties, jute fabric, physical properties, vinyl ester.

I. INTRODUCTION

An engineered bio based composite materials that are made from rapidly renewable resources and biodegradable can potentially replaces non-structural building and infrastructure materials. This can lessen the environmental issues arise from traditional composite materials [1]. This can be achieved by taking advantage of natural fibres that are getting more importance lately, such as jute, hemp, sisal, kapok, rice husk, and bamboo which most widely used in polymer composite industry as reinforcing materials [2]. Jute as fibre reinforcement for composites has many advantages as compared to traditional composite such as relatively lightweight composite with high specific strength and stiffness properties, low density material, plant based fibre, environmental friendlies and renewable [3], [4]. Jute fibre also possesses good mechanical and physical properties among all other natural fibres.

The most recent researches in natural fibre composites fields are enhancement of the adhesion in between fibre and fibre matrix to improve the mechanical properties of the fibres. However, due to the strict and safety regulations on the composite manufacturing applications retarded the utilization natural fibre composites (NFC) in industrial field because of the poor tolerant against thermal and fire of fibre reinforcement materials [5], [6]. On top of that, the existing fire-retardant additives such as halogen base flame retardant are highly effective for polymer composites yet non-environmentally and high toxicity of the material [5], [7], enforcing researches to finding a new fire-retardant material. Ammonium polyphosphate (APP) as phosphorus based intumescent flame retardants (IFR) system is one of those phosphorus-based additives which fulfilling all criteria mentioned before. It is a very well-known, APP fire-retardant able to improve the thermal stability and fire-retardant properties of composites, and unlike the other phosphorus-based additives, is not harmful to health [8]. Many works found on jute chemical treatments that enhance the adhesion between reinforcement and matrix [9]– [12]. Nevertheless, to the best of authors' knowledge, fewer efforts were found on the enhancement of jute composite fire-retardant and its degradation properties [13]– [15]. Hence, this study is mainly focused on the investigation of the jute natural fibre composite treated with ammonium polyphosphate.

II. MATERIALS AND METHOD

Vinyl ester resin (SH 7000) was purchased from BT Science Sdn Bhd. Jute Fibre were supplied by Yeo Hup Kee Sdn Bhd. Sodium hydroxide and acetic acid were used as the alkalization treatment for surface modification on jute fabric.

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Ammonium polyphosphate (APP)/Exolit AP 435 was purchased from DKSH Malaysia Sdn Bhd and used to enhanced the fire-retardant properties of fabric composites.

A. *Materials*

1) Preparation of jute/vinyl ester and jute/vinyl ester/ APP composites

The jute fabric was first be treated with 2% alkali (NaOH) treatment for 2 hours soaking time. In composite fabrication, the vinyl ester resin was prepared by mixing five different percentages of APP on the ratio 100:5, 100:10, 100:15, 100:20, and 100:25 before MEKP was added into the mixture. with the ratio of 1:44 by weight. The composite was produced by using hand lay-up technique in this study. The prepared mixture was applied on four fabric layers with size (350 × 350 mm) and the steel roller was used to evenly spread and squeezed out the excessive resin.

Then, the wet fabrics was laid in between thick glass plates (400 × 400 × 100 mm in dimension). Then this assembly was compressed with a weight placed on top of this mixture to remove the excess resin and left to cure under room temperature for 24 hours before post curing process take placed in an oven for four hours at 80°C. The fabricated sample were abbreviated according to the APP percentages treatments on the jute composites as follows; JC0, JC5, JC10, JC15, JC20 and JC25.

B. *Characterizations*

1) Physical Measurement

Relative density, thickness, fibre volume fraction, and matrix content volume percentage of composite have been carried out using ASTM D792, ASTM D3039 and ASTM D3171-11 respectively.

2) Morphological analysis and chemical characterization (FESEM/EDX)

The cross section and JC fibre surface were examined by using a field emission scanning electron microscopy (ZEISS SUPRA 40BP) at 5kV voltage. Thin layer of gold was coated on the sample for electrical conductivity and avoid charging effect purpose to provide a better resolution of the images taken.

For the chemical characterization, energy dispersive X-ray spectroscopy (EDX) which supported by FESEM images was used to investigated the chemical compositions of treated and untreated JC sample.

3) Flammability Testing

Flammability properties were investigated using burning test method according to ASTM D635. This fire-test response test method is used to compare the relative linear rate of burning of composites in the form rectangular specimen in the horizontal position. The measured data will be the rate of burning for the material.

4) Thermogravimetric analysis

Thermal analysis was performed using thermogravimetric analyzer (Shimadzu TGA 50), under inert atmosphere of a nitrogen gas. The range of temperature used was between 30°C and 1000°C and at constant heating rate of 10 °C/ min.

III. RESULTS AND DISCUSSION

A. *Physical Properties*

As shown in Table- I, control sample (JC0) showed the highest fibre volume fraction (31.46%) among all of other samples. Whilst other samples possessed lower and the trend was decreased as the amount of APP treatment was increased. The vinyl ester volume fraction however, showed inverse trend to fibre volume with the highest point was showed by sample JC25 (80.89%). This is due to the increment of APP deposited on the composite material which increases the thickness and weight of fabricated composites. Similar reason is applied for the increment trend of sample densities.

Justification from the cross-section view in the Fig. 1(b-d) at 100μm magnification, clearly visible the fibre, matrix and APP particle at the JC5, JC20 and JC25 in comparison with Fig. 1(a) where at the resin part the surface was clear and smooth. In this study, EDX as chemical analysis along with FESEM were used to observed features on the surface and investigate the elemental analysis of the untreated and treated jute composite (JC). The result showed that JC consist of four main element which are carbon, oxygen and phosphorus while aurum (gold) was being exempted because of an external element that was used to coating the sample for the conductivity and avoid charging effect purpose as shown in Fig. 2(a-b).

Table- I: Physical properties of untreated and treated JC.

Sample	Relative Density (g/cm3)	Thickness of Composites	Fibre Volume Fraction (%)	Matrix Volume Fraction (%)
JC0	1.213	3.23	31.457	67.595
JC5	1.24	3.80	26.157	74.444
JC10	1.24	3.85	25.817	74.786
JC15	1.24	3.9	25.486	75.120
JC20	1.29	3.97	24.066	79.638
JC25	1.297	4.08	23.291	80.888

The C and O element were found to be the major element present in the X-ray emission spectrum as they are theoretically followed the main element in the reinforcement and matrix. Results of EDX in Table- II shows the weight percentages (wt %) of each element in all JC samples. The main element of JC0 contains of carbon, oxygen with weight percentages of 39.57% and 12.90%. While for treated sample JC10 and JC20 there was an increment observed from 1.02% to 1.66% of the phosphorus element as the amount of APP increased. As the present of the phosphorus element higher in the JC, it should promote a better thermal stability and flammability properties for JC samples. Meanwhile for the JC5 sample, the traces of phosphorus element were not found in EDX analysis, this cased could be quantification error as sample volume for EDX analysis is small compared to large sample produced. This cased might be an aggregation of APP particles in the JC which obstructed the dispersion of the mixture and thus reduce homogeneity of the JC sample.

Table-Energy dispersive X-ray analysis of untreated and treated JC.

Type of composite sample	Weight percentage (%)				
	C	*O*	*P*	*AU*	*TOTAL*
JC0	39.57	12.90	-	47.53	100
JC5	36.43	11.52	-	52.05	100
JC10	37.73	14.20	1.02	47.02	100
JC20	39.03	11.57	1.66	47.74	100

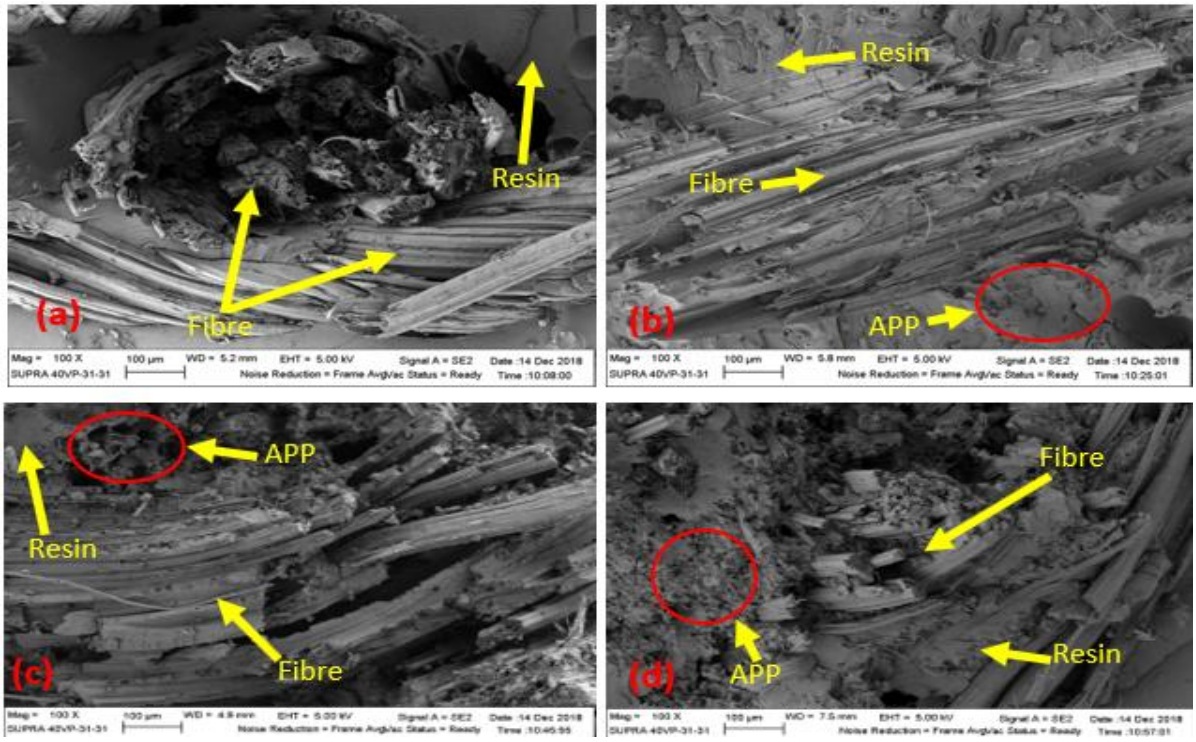

Fig. 1. Cross section view; FESEM micrographs of tensile fracture (a) JC, (b) JC5, (c) JC20 and (d) JC25.

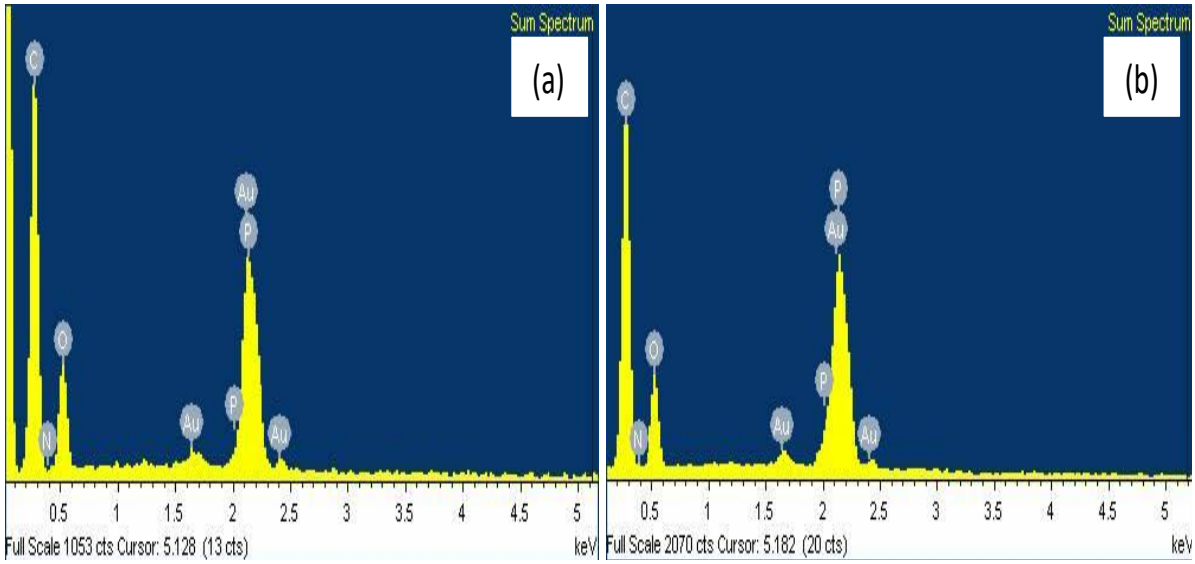

Fig. 2. EDX analysis on the untreated (a) and treated JC sample (b)

A. *3.2. Flammability Testing*

1) Burning testing

Burning characteristics of JC were investigated in accordance to ASTM D635 and the result was tabulated in Table- III. In this test, 30s-time exposure of small flame was set and exposed within 25mm mark on the composite specimens. Among all the sample, only the JC0 sample was burned completely Fig. 3(a). The time taken was 235.06 s for the flame to spread and reached 100mm marks. While the burning process progressing, it was also observed that the sample was burned in yellowish orange flame, produced thick black smoke and released pungent smell like stronger burnt plastic mix with paper which might be correspond to the mixture of the jute fabric and vinyl ester resin. The linear burning rate of this sample was measured as 19.17 mm/min.

After the test, a huge amount of ash was produced and only a little residue was left behind of this sample. This indicates that the JC0 sample is poor in flame retardancy performance. For the JC5 sample, it was partially burned with yellowish orange black smoke where the flame spread reach 25 mm marks but doesn't reached 100 mm marks. Besides that, the sample shape was maintained when the test finished due to char formation present that prevent the flame to spread and disintegrate the composition into an ash. The burnt part of sample became brittle at the char area as shown in Fig. 3(b).

As the content of APP increases from 10 phr to 25 phr, it produced less and thin smoke while the abilities of the composite samples to retard the flame from spreading keep increasing Fig. 3(c-d). This is proven by the failure of the flame to reach even the first mark as shown in Table- III. Hence, it can be concluded that the increasing of APP content improves the flame retardancy of the composites sample

Table- III. Results of the burning test of all fabricated composites

Sample	1st mark (25 mm)	2nd mark (<100 mm)	3rd mark (100mm)	Elapsed time (s)	Linear burning rate (mm/min)
JC0	/	/	/	235.06	19.17
JC5	/	/	X	113.345	X
JC10	X	X	X	X	X
JC15	X	X	X	X	X
JC20	X	X	X	X	X
JC25	X	X	X	X	X

X indicated the expected event had not happened

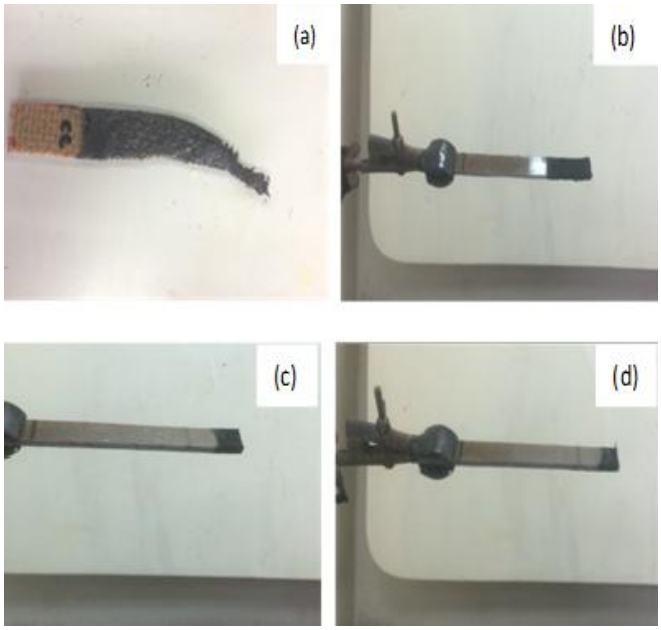

Fig. 3. Images of all types of fabricated composite sample after burning test; (a) JC0, (b) JC5, (c) JC10, (d) JC20

A. *Thermal Properties*

Fig. 4 showed TGA curves of jute fire retardant composites against flame retardant. Temperature at 15% weight loss of the specimens are characterized and were taken as the initial thermal stabilities as shown in Table- IV. From Fig. 4, it can be observed that the JC5, JC10 and JC20 showed lower thermal stabilities compared to the JC0 sample.

The temperature at 15% weight loss of the JC0 and JC5 sample was decreased from 319 $^{\circ}$C to 314 $^{\circ}$C respectively. Further decreasing of the temperature to the 309 $^{\circ}$C at 15% weight loss by addition of APP up to JC20 led to the deterioration of the thermal stability. This study follows the similar trend of the other studies in thermal stability of the natural fibre reinforced polymer composites (NFRPC) which have been reported by previous researchers [5], [16]– [18].

When exposed to high temperature, APP is reported to released phosphoric acid and non-flammable gases which then produced intermolecular dehydration and charring. Many oligomers and small molecular species are produced by an acid from the pyrolysis of APP catalysts –C–O– bond breaking [16]. Based on these facts, it was presumed that the first stage of weight loss caused by an acid released from the APP decomposition due to APP pyrolysis state and advance decomposition of the jute fibres.

It can be implied that the addition of the APP content improves the thermal stability of the jute composites and increases char residues at high temperature as shown in Table IV. The char residue of all the treated JC sample were still remained whereas the untreated JC was degraded completely. This is because at the elevated temperature, the intumescent char is forming thus preventing the inner matter of the composites from further degradation.

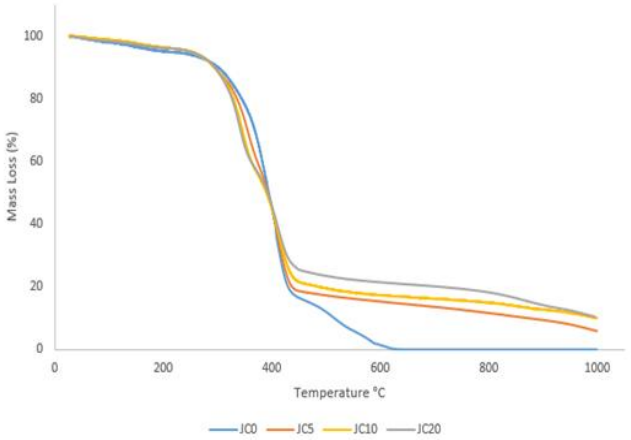

Fig. 4. TGA curves for JC0, JC5, JC10 and JC20.

Table- IV. Effect of APP content on jute fabric vinyl ester composites.

Designation	APP PHR	T15% (at weight loss)	Char residue at 600 °C
JC0	0	319	0
JC5	5	314	15.24
JC10	10	315	17.32
JC20	20	309	21.4

IV. CONCLUSION

Incorporation of APP as flame retardant in JC exhibited the enhancement in its fire-retardant properties. In terms of physical properties, thickness and matrix volume fraction were increased whilst fibre volume fraction was decreased as the amount of APP was increased. This is most probably due to the impregnation of APP that makes the composite become denser. The treated JC sample showed the good performance against thermal and fire. For the burning test, the ability of the treated JC to inhibit fire was observed to be higher as the amount of APP increased. In the thermogravimetric results, it was clearly showed that, the char residue of all the treated JC composites is still remained while the untreated JC was completely degraded. Thus, the treatment using APP was highly effective to enhance and improve the thermal and flammability properties for natural fibre reinforced polymer composites.

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