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Research article

Processing and characterization of flame-retardant natural fibre-reinforced epoxy composites and construction of selection charts for engineering applications

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Abstract. Natural fibre-reinforced polymer composite (NFRPC) has been introduced as one of the solutions to overcome the ecological and environmental problems accompanying the widespread usage of polymeric materials in every facet of life. However, the organic nature of both natural fibres (NFs) and polymers increases their flammability behaviour, and this, in turn, limits their application. In this regard, this work concentrates on studying the effect of adding flame retardants (FR) to jute-reinforced epoxy composites (JRECs), either by treating the jute fabric with diammonium phosphate (DAP) or adding DAP powder to the epoxy resin matrix on the flame retardancy performance as well as the mechanical properties. The results showed that the effect of the incorporation method of DAP either to jute fabric or to resin matrix has a significant difference on the flammability test results at low concentrations; however, at higher concentrations, the flame retardancy performance is not affected by the technique of adding FR to the composite system. On the other hand, the mechanical properties are significantly affected by the method of incorporating FR to JRECs at all concentrations. Moreover, the results obtained from JRECs with FR systems were evaluated and compared with the literature by constructing selection charts that relate the flame retardancy level to mechanical properties.

Keywords: *epoxy resin, natural fibre, thermogravimetric analysis, biocomposites, flame retardant, mechanical properties, selection charts*

1. Introduction

Reinforcing the polymeric matrix with natural fibres (NFs) opens the avenue to reduce the depletion of non-sustainable resources and produce more engineering products that have less environmental impact [1]. NFs have several advantages, such as low cost, low energy consumption, availability from renewable resources, easy handling compared to synthetic fibres, ease to process, lightweight and relatively high specific properties [2, 3]. Natural fibre-reinforced polymer composites (NFRPCs) are widely used in various applications ranging from sporting goods, furniture and automotive interior parts to structural applications in civil construction and aircraft [4]. At present, the lower mechanical properties and higher flammability of NFs compared to synthetic reinforcements such as glass and carbon fibres limit their market share [5]. Hybridizing NFs with synthetic fibres comes as a solution to enhance the mechanical properties [6–8]. On the other hand, the high flammability of NFRPC can be reduced by adding flame retardants (FRs) to the polymer matrix or/and treating the NFs with FRs [9]. Jute is one of the most popular cellulosic fibres [10]. Low cost, abundance and high specific strength are the characteristics that have allowed jute fibres to be used in multipurpose applications ranging from carpets, fancy bags and furniture to composite components in

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automotive, rehabilitation and ceiling panels in the construction sector [11–14]. The use of NFs in structural applications can be achieved by reinforcing NFs in textile form with thermosetting polymer.

Amongst all thermosetting resins, epoxy resin is the most used, and it covers a wide range of applications [15–17]. Epoxy resins acquire several characteristics, such as, low shrinkage during curing, high strength, low toxicity, strong adhesion to different substrates and high corrosion resistance [18]. Several studies [19–23] have focused on the effect of reinforcing epoxy resin with NFs as well as the effect of hybridizing NFs/epoxy composites with different synthetic reinforcements (whether in fibre or particulate form) on the mechanical properties. However, little research [24–34] has been conducted to evaluate the effect of adding FRs on both the mechanical and flame retardancy behaviour of epoxy composites. On the other hand, studies [14, 25, 35] have only evaluated the flammability behaviour of NFs/epoxy composites. Table 1 summarizes the most important results obtained from literature related to both flammability tests (in terms of UL-94 and limiting oxygen test (LOI)) and mechanical tests (in terms of flexural strength (FS) and tensile strength (TS)). It is to be noted that two approaches were used to add FR to the composites: either FR added to the matrix or FR added to reinforcement. These two methods are abbreviated flame-retardant matrix (FRM) and flame-retardant reinforcement (FRR). The first column in Table 1 refers to the type of the composite system and processing technique, where CM stands for compression molding, MC stands for mold casting, HLU stands for hand lay-up, HLUCM stands for Hand lay-up followed by compression molding and VB stands for vacuum bagging. Some cells are left empty since these data were not available.

The different types of FRs can be classified according to the functional elements that built up the flame retardant, mechanism of action and mode of action [36]. Halogenated FRs are commonly used since they act in the gaseous phase during combustion and have a significant effect in enhancing the flame retardancy performance of polymeric materials [37]. However, they have been banned in the last decades in many countries since they evolve toxic gases during combustion [38]. Recently, both researchers and industrial sectors have been working on replacing halogenated FRs with more environmentally friendly FRs such as phosphorus-based compounds, silicon-based compounds, borates and metal hydroxides [1, 5].

Referring to our previous review paper [39] and literature done after the publication of this review paper that is summarized in Table 1, it is noted that it is common to incorporate an FR directly into the matrix, and few articles studied the effect of chemical treatment as well as the effect of treating the reinforcement with flame retardants on the flammability and mechanical properties of the NFRPC. Jiang *et al*. [40] studied the effect of adding 9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide (DOPO) to glass fibre-reinforced epoxy (GERE) composites, either to glass fabric or to the bulk epoxy matrix on both flame retardancy and mechanical properties. They found that adding 4.5 wt% DOPO to glass fabric has enhanced the limiting oxygen index (LOI) value to 29%, while incorporating 4.5% DOPO to epoxy matrix has slightly increased to 24% compared to GFRE with LOI 21%. Branda *et al.* [29] investigated the effect of both treating hemp fabric with water glass and the addition of ammonium polyphosphate (APP) to the epoxy matrix on the flame retardancy performance as well as mechanical properties. Cone calorimeter test was used to evaluate flame retardancy performance. It is found that the effect of treating hemp fabric with water glass has slightly reduced peak heat release rate (PHRR) and total heat release rate (THRR). Besides, this treatment has reduced the flexural strength by 15.6%. On the other hand, the addition of 15 wt% APP to epoxy matrix reinforced by hemp fabric has reduced both PHRR and THRR by 66.6 and 43.8%, respectively, without compromising the flexural strength.

Based on the abovementioned works, it is concluded that there is a knowledge gap in studying and comparing the method of incorporating FR into the composite system, whether to add to the matrix or the reinforcement. Besides, it is revealed that several studies have used phosphorus-based FRs in different composite systems. Ammonium polyphosphate (APP), diammonium phosphate (DAP), and other phosphorus- and nitrogen-containing substances are commonly used as a surface fibre-matrix modifier to enhance the flame retardancy performance of the composite system [41]. Indeed, DAP is increasingly being used with natural fibres to improve the thermal resistance of cellulosic fibres, which can be altered by lowering the combustion temperature, reducing weight loss, and increasing residual char [41, 42].

Table 1. Literature on NF/epoxy composites with FRs. The designation in the fourth column refers to the type of matrix followed by fabric type and its wt% content, and finally, FR type and its wt% content; for example, M_F-20_FR1-2 means matrix M reinforced with 20 wt% of Fabric F and incorporated by 2 wt% of flame-retardant type FR1.

Composite/ technique	FR method of addition	FR type	Designation	FR [wt%]	UL-94	LOI	*TS* [MPa]	*FS* [MPa]	References
Epoxy_Banana Fibre/(CM)	FRM	Banana fibre (BF)	EP_BF-0	0	HB (29.4)	20.6	50	81.8	[30]
		Banana fibre (BF)	EP_BF-10	10	HB (28)	20.6	58	86	
		Banana fibre (BF)	EP_BF-15	15	HB (27.4)	20.8	64	88.8	
		Banana fibre (BF)	EP_BF-20	20	HB (26.1)	20.8	68	90.1	
		Banana fibre (BF)	EP_BF-25	25	HB (28)	20.2	65	89.2	
Epoxy_coconut shell particle/ (MC)	FRR	Diammonium phosphate (DAP)0	EP_CSP-30	0	HB (21.7)				[25]
		Diammonium phosphate (DAP)	EP_5DAPCSP-30	5	HB (14.8)				
Epoxy_Hemp Fabric/(HLU)	FRR		EP_Hemp-30	0	HB	22	66.5	92.79	[31]
		Hemp treated with 17 mass% phosphoric acid solution and 5% ammonium hydroxide solution	EP_0.51 P Hemp-30	0.51	HB	26	56.18	81.15	
		Hemp treated with sol gel amine type silane and 17 mass% phosphoric acid solution and 5% ammonium hydroxide solution	EP_0.39 P1.45 Si Hemp-30	1.84	HB	28	49.14	71.04	
Vinylester_ Hemp Fabric/ (HLUCM)	FRR		EP_Hemp-46	0		22.8	61.68	93.65	[32]
		Ammonium polyphosphate (APP) coated on Hemp	EP_25APPHemp-46	11.5		29.2	51.51	85.2	
Epoxy_palm empty fruit bunch (EFB)/(CM)	FRM	Microcrystalline cellulose (MCC)/ammonium polyphosphate (APP)/alumina trihydrate (ATH)	EP_EFB-20_APP-10_ATH-5	15	V0				[35]
		Microcrystalline cellulose (MCC)/ammonium polyphosphate (APP)/alumina trihydrate (ATH)	EP_EFB-20_MCC-3_APP-10_ATH-5	18	V0				
		Microcrystalline cellulose (MCC)/ammonium polyphosphate (APP)/alumina trihydrate (ATH)	EP_EFB-20_MCC-3_APP-10_ATH-6	20	V0				
		Microcrystalline cellulose (MCC)/ammonium polyphosphate (APP)/alumina trihydrate (ATH)	EP_EFB-20_MCC-3_APP-10_ATH-7	22	V0				
Epoxy_Jute Fabric/ (HLUCM)	FRR		EP_Jute			19.1			[14]
		Polydopamine (PDA) coated jute	EP_PDAJute			26.1			

DAP is a non-durable water-based FR, thus, it has a relatively low cost compared to other FRs [43]. This research focuses on two main points. First, to study the different methods of adding DAP with

Table 1. Continuously 1.

Composite/ technique	FR method of addition	FR type	Designation	FR [wt%]	UL-94	LOI	*TS* [MPa]	*FS* [MPa]	References
Epoxy_Flax Fabric/(HLU)	FRM		EP_2LFlax	0	NR	21.3	47		[24]
		Ammonium polyphosphate (APP)	EP_2LFlax_APP-10	10	NR	22.4	40		
		Ammonium polyphosphate (APP)	EP_2LFlax_APP-20	20	V1	25.5	39		
		Aluminium hydroxide (ALH)	EP_2LFlax_ALH-20	20	NR	22.5	42		
		Aluminium hydroxide (ALH)	EP_2LFlax_ALH-30	30	NR	23.5	46		
		Aluminium hydroxide (ALH)	EP_2LFlax_ALH-40	40	NR	24.5	41		
Epoxy_oil palm empty fruit bunch (OPEFB)_PET yarn/(HLU)	FRM	Magnesium hydroxide (MgOH)/PET yarn/OPEFB	EP_MgOH-5_PET-5	5	HB (22.2)		10.79		[33]
		Magnesium hydroxide (MgOH)/PET yarn/OPEFB	EP_MgOH-5_PET-5OPEFB-20	25	HB (11.5)		4.29		
		Magnesium hydroxide (MgOH)/PET yarn/OPEFB	EP_MgOH-5_PET-5_OPEFB-35	40	HB (14.4)		3.88		
		Magnesium hydroxide (MgOH)/PET yarn/OPEFB	EP_MgOH-5_OPEFB-50	55	HB (17.3)		3.63		
Epoxy_Flax Fabric/ (HLUCM)	FRM	Ammonium polyphosphate (APP)	Ep_Flax-40	0	NR			124	[26]
		Ammonium polyphosphate (APP)	Ep_Flax-40_APP-12	12	V0			116	
Epoxy_Kenaf fibre_PET yarn/(HLU)	FRM	MgOH/PET/Kenaf	EP_MgOH-5_PET-5	5	HB (22.2)		10.87		[34]
		MgOH/PET/Kenaf	EP_MgOH-5_PET-5 -Kenaf-20	25	HB (19.6)		19.96		
		MgOH/PET/Kenaf	EP_MgOH-5_PET-5 -Kenaf-35	40	HB (14.6)		25.24		
		MgOH/PET/Kenaf	EP_MgOH-5_PET-5 -Kenaf-50	55	HB (20.5)		32.02		
Epoxy/Unsaturated Polyester _Sisal Fabric/(CM)	FRM	Nanoclay, bis(2-hydroxy-ethyl) methyl hallow ammonium (Cloisite 30B)	EP-95_UP-5_ASTF-30	0	V1	25	119	180	[28]
		Nanoclay, bis(2-hydroxy-ethyl) methyl hallow ammonium (Cloisite 30B)	EP-95_UP-5_ASTF-30_NC-1	1	V1	27	128	191	
Epoxy_Hemp Fabric/(VB)	FRM & FRR		Epoxy_Hemp-25	0			109		[29]
		Waterglass treated Hemp fabrics/epoxy composite	Epoxy_WGT-Hemp-25	0			92		
		Hemp fabrics treated with waterglass/ epoxy/ammonium polyphosphate (APP)	Epoxy_Hemp-25_APP-15	15			110		
		Hemp fabrics treated with waterglass/ epoxy/ammonium polyphosphate (APP)	Epoxy_WGT-Hemp-26_APP-15	15			94		

various concentrations to JREC on both flammability and mechanical properties. Secondly, to construct flame-retardant selection charts for NFs reinforced epoxy composites based on the data collected from Table 1 and the present work. These charts will guide the designer to select the appropriate NFRPC with FRs that can replace more conventional materials in different engineering sectors (construction, electronics, automotive, *etc*.). These charts correlate two main criteria, flammability, and mechanical properties for NFRPCs with FRs. Besides, researchers can easily use these selection charts and find knowledge gaps in the field of NFRPC with FRs.

2. Experimental work

2.1. Materials

Epoxy (EP) resin was supplied by AXSON Technologies, France, in the form of Biresin CR82-6 epoxy resin to be mixed with Biresin CH80-6 as the hardener. This resin system is characterized by very low viscosity and outstanding wettability of reinforcements, making it suitable for composite preparation. Table 2 shows the main physical properties of this system, as given by the material's datasheet. Jute Fabric (JF) with an areal density of 235 g/m^2 is based on a plain weave with 5 threads/cm in warp direction and weft direction, supplied from the local market, Egypt. Dia-ammonium phosphate (DAP) of mean diameter 300 nm was supplied by Morgan Chemicals, Egypt.

2.2 Jute fabric treatment

Firstly, the jute fabrics were washed and rinsed in tap water and then dried in ambient air for 2 days. Then the fabrics were further oven dried at 70 °C for 48 h. Secondly, the dried JFs were immersed in DAP solutions with different concentrations (6, 18 and 32%). The ratio of fibre to DAP solution was 1 g fibre to 20 ml DAP solution. After 24 h, the fabrics were removed from the DAP solution and dried in ambient air for 2 days, followed by an oven drying at 70 °C for 24 h. The cooled samples were kept in plastic bags for further processing. The chemical uptake or pick-up for the treated jute fabrics was calculated according to Equation (1):

$$\text{Chemical add on [\%]} = \frac{W_2 - W_1}{W_1} \cdot 100 \quad (1)$$

where W_2 and W_1 are the weight of dried fabric after and before treatment, respectively. In this work jute treated with DAP weight gained are 9, 27 and 54 wt%. The treated jute fabrics were designated as 9DJ, 27DJ and 54DJ.

Table 2. Physical properties of epoxy system resin.

		Resin	Hardener
Mixing ration by weight		100	27
Viscosity at 25 °C	[mPa·s]	900	10
Density at 25 °C	[g/cm^3]	1.13	0.95
Mixing viscosity at 25 °C	[mPa·s]	230	
Density of the cured product at 23 °C	[g/cm^3]	1.08	

2.3. Composite preparation

JRECs panels were fabricated using the vacuum bagging technique. Six plies of jute fabric each of 270×200 mm were used. The stack of JF coated with epoxy resin (applied by hand lay-up technique) was centred on a steel plate coated with a releasing agent and further sealed under a bagging film. A Vacmobile SVM 2S series vacuum pump was used, and a vacuum of 100 kPa (absolute) was applied. The laminate stack was left under vacuum to cure overnight. The laminate was then post-cured for further 6 h at 60 °C. Figure 1 illustrates a schematic sketch for the composite preparation of adding FR to jute fabric or epoxy matrix. Jute fabric (treated and untreated) was kept within the range of 23–26 wt% in all composite samples. The density of the prepared composite samples was measured based on the Archimedes principle using a density kit in combination with the ADAMEQUIPMENT PW254 sensitive balance. Moreover, void content has been calculated for each composite composition based on the measured density and the calculated theoretical density. Table 3 presents the compositions, nomenclature and void content of the prepared composites.

2.4. Characterization

The surface morphology of untreated and treated jute was analyzed by a scanning electron microscope SEM Quanta FEG250-FEI-made in the USA using high vacuum mode, 20 kV and spot size 3.5 and optical microscope OLYMPUS BX51.

The Fourier transform infrared (FTIR) spectra for untreated and treated jute fabric were investigated using JASCO FT/IR-4700 spectrophotometer. The transmission spectra were measured in the range of 500–4000 cm^{-1} with a resolution of 2.0 cm^{-1} using ATR mode.

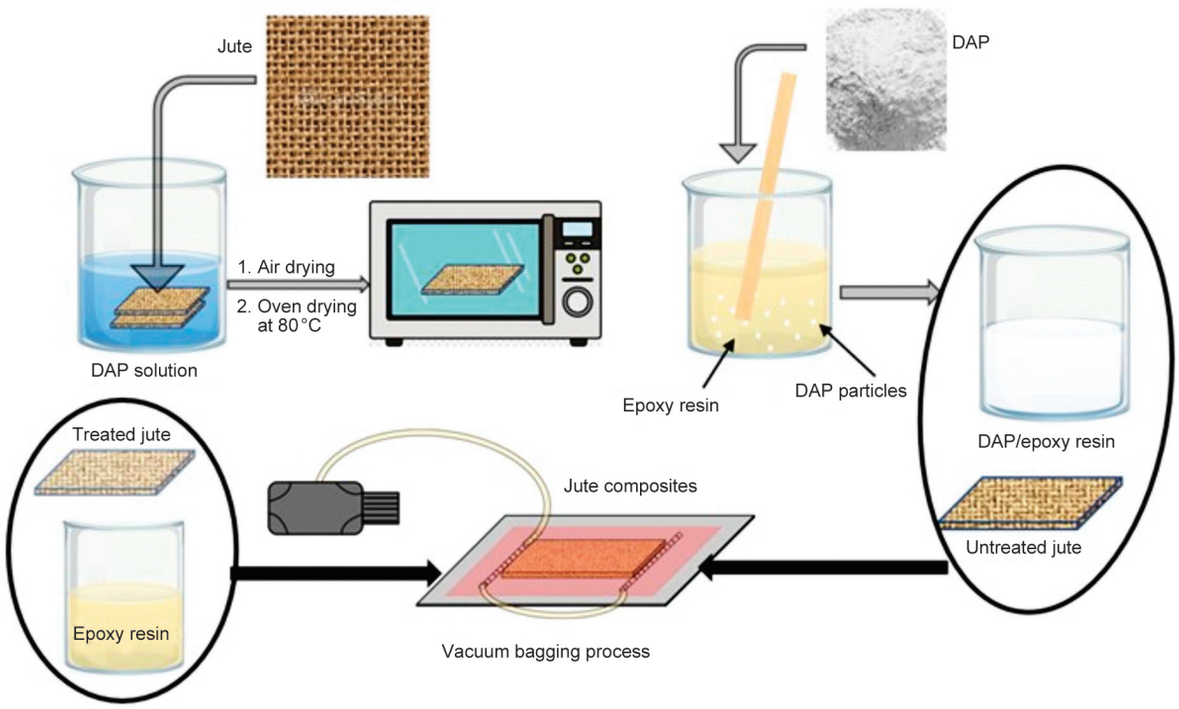

Figure 1. Schematic sketch for composite preparation.

Table 3. Composite designation.

Sample	DAP% in jute	DAP% in epoxy	DAP% in composite	Void [%]
UJE	0	0	0	4.3
UJ3DE	0	3	2.25	5.9
UJ9DE	0	9	6.25	6.0
UJ18DE	0	18	12.50	4.7
9DJE	9	0	2.25	1.8
27DJE	27	0	6.25	3.3
54DJE	54	0	12.50	10.6

The thermal decomposition of pure epoxy resin, jute fabric, and treated jute fabric with FR and JFREC with FR was investigated using a TA Q500 thermogravimetric analyzer. TGA tests were conducted on specimens with a 5–10 mg mass and heated from 40 to 700 °C at a heating rate of 10 °C/min under a nitrogen atmosphere. The mass loss curves were recorded as a function of temperature for different samples. These curves were analyzed to obtain the onset decomposition temperature at 5% weight loss, degradation temperature at maximum decomposition rate and char residue percentage at 700 °C.

To study the flammability of composites, UL-94 vertical burning test was conducted according to ASTM D3801, the samples that did not pass UL-94 vertical test underwent UL-94 horizontal burning test, and the test was performed according to ASTM D635. Besides, limiting oxygen test (LOI) was also performed according to ASTM D2863.

The mechanical characteristics were evaluated by conducting 3 point bending test according to ASTM standard D-790 with a sample size of 130×13×3.5 mm using an LRXPLUS universal testing machine equipped with a 5 kN load cell at a crosshead speed of 5 mm/min and measuring. 8 samples were considered for analysis. Charpy impact strength for un-notched eight samples (80×10×2 mm) in accordance with DIN EN ISO 179-1 using a 2 Joule hammer to strike the sample at its edge side.

3. Results and discussion

3.1. Surface morphology for jute fabric

The surface morphology of untreated and DAP-treated jute fabrics was observed by optical microscope and SEM. Figure 2a illustrates the longitudinal appearance for jute fibre. Jute fibre has a cylindrical shape with no curl. Figure 2b shows jute fabric with 9 wt% DAP, it is clearly observed the precipitates of DAP on Jute fibre. Figure 2c and Figure 2d represent jute fabric with add on 27 and 54 wt% DAP, respectively. It can be noticed that clear, flaky crystals have appeared, and their content increased as DAP concentration increased (Figure 2c, Figure 2d). This can be attributed to the mechanism of precipitation of DAP on jute fabric. To prepare jute fabric treated

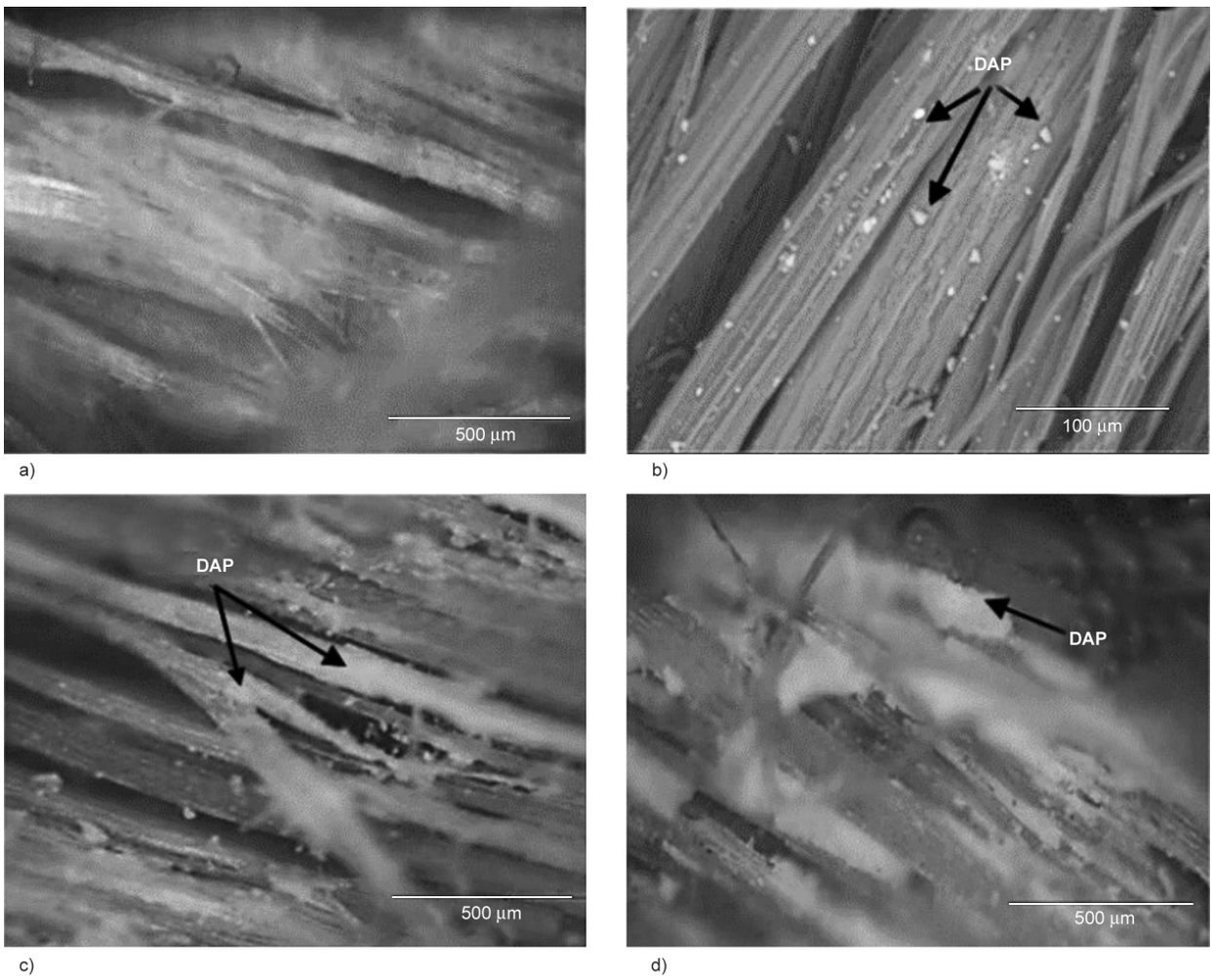

Figure 2. Surface morphology for a) untreated jute, b) 9 wt% DAP jute (9DJ), c) 27 wt% DAP jute (27DJ), d) 54 wt% DAP jute (54DJ).

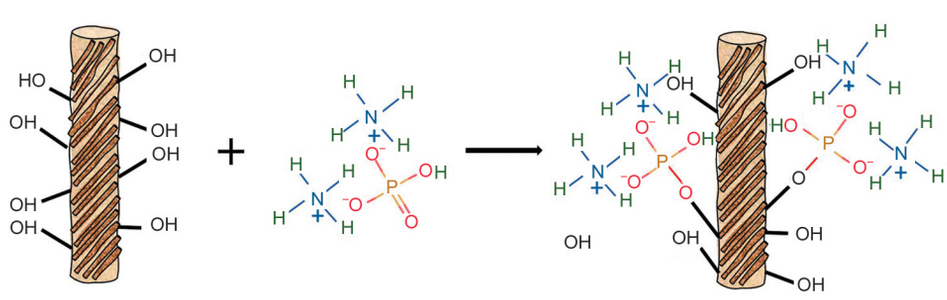

Figure 3. Schematic of jute fibre reaction with DAP solution.

with DAP, firstly, DAP solution was prepared, DAP crystals were dissolved in water to form phosphate anion and diammonium cation. Secondly, the jute fabric was immersed in the solution, followed by drying the jute fabric in air. Figure 3 represents a schematic for the reaction that occurs between DAP and jute fibre. It can be noted that part of the hydroxyl groups on jute fibres were bonded by the double oxygen bond in the phosphate ion. Additionally, during the drying process, the phosphate anion and diammonium cation bond together again to form DAP salt. Thus, as the concentration of DAP in the water solution is low, small precipitates of DAP were formed on the jute fabric (Figure 2b). However, as DAP concentration increases, large flaky crystals appear on the jute surface (Figure 2c, Figure 2d).

3.2. FTIR

The FTIR spectra for untreated and DAP treated jute are displayed in Figure 4. The peaks at 3308, and

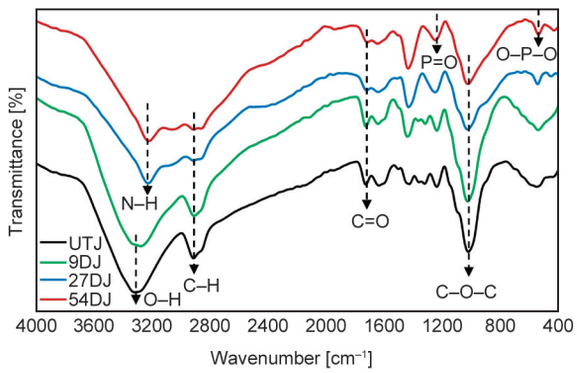

Figure 4. Fourier transform infrared spectra of untreated and DAP-treated jute.

2901 cm^{-1} untreated fibre correspond to the stretching vibration of O–H group of lignin and C–H bond of cellulose, respectively [44]. The peaks appearing at 1726 and 1620 cm^{-1} are attributed to C=O group that is present in hemicelluloses and lignin [45]. The peak of C–O–C corresponds to 1019 cm^{-1} [46]. It is to be noted that the intensity of C–O–C for treated jute is reduced, and this indicates that the hydrogen atom in cellulose, hemicellulose and lignin is replaced by another functional group [47]. Compared with untreated jute, new peaks at 3042, 1243, 1027 and 422 cm^{-1} are attributed to the stretching vibration of N–H, P=O, P–O–C and O–P–O, respectively [46, 48, 49] for treated jute samples. These results indicate that DAP is well coated on jute fabric.

3.3. Flammability results

The limiting oxygen index (LOI) and UL-94 burning tests were performed to evaluate the flame retardance of Jute/epoxy composites, and the test results are shown in Table 4. Figure 5 displays and compares representative video screenshots obtained during UL-94 vertical test for the composite materials. Untreated JREC (UJE) is easily ignited and burns up to the clamp within 56 s in the vertical burning test, with a LOI of 21%. Compared with the UJE composite, the incorporation of DAP into JREC has increased the LOI values to different degrees. UJ3DE (Composite system with 2.25 wt% Dap by adding DAP powder to epoxy matrix) has slightly increased the LOI to 22.8%. In contrast, 9DJE (composite system with 2.25 wt% DAP by adding DAP-treated jute fabric) has increased LOI to 26.9%. Meanwhile, the LOI values for UJ18DE and 54DJE, which are both

Table 4. Ul-94 and LOI burning test results.

Sample	FR [wt%]	UL-94 rate	LOI [%]
UJE	0	HB (13.2 mm/min)	21.8
UJ3DE	2.25	HB (12.2 mm/min)	22.8
9DJE		HB (0 mm/min)	26.9
UJ9DE	6.25	V0	26.0
27DJE		V0	26.8
UJ18DE	12.5	V0	26.8
54DJE		V0	35.8

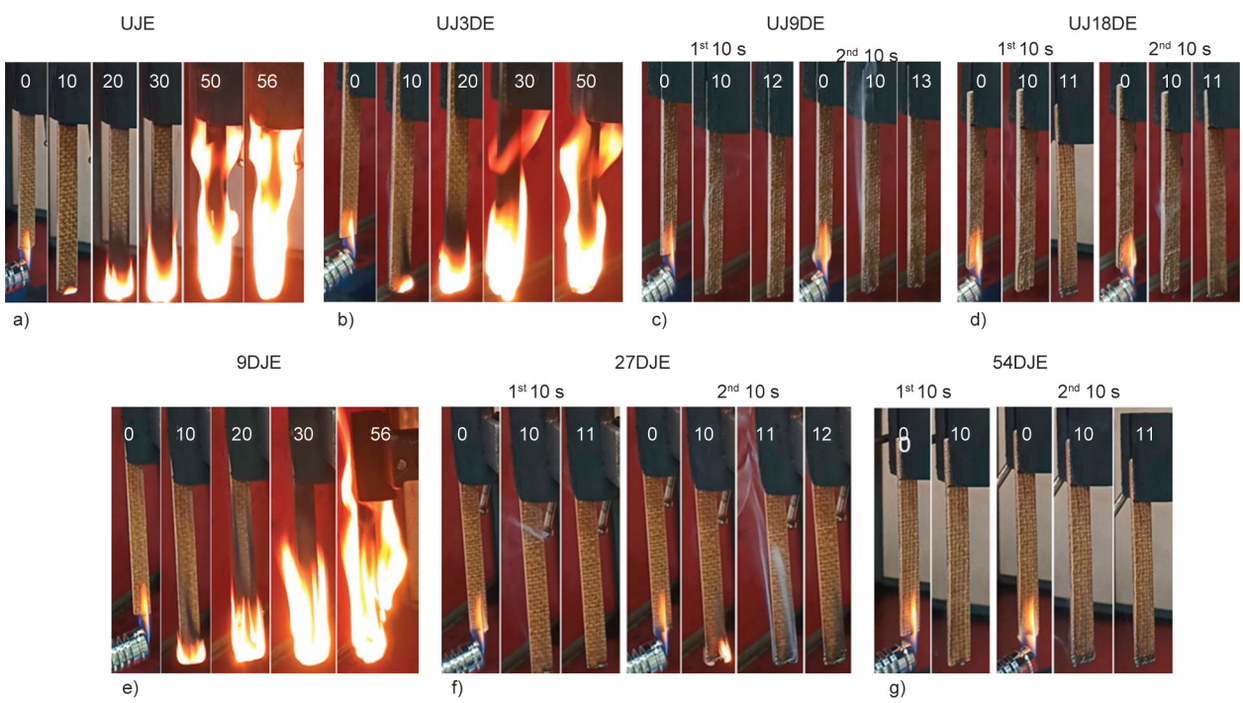

Figure 5. Video screenshots obtained during UL-94 vertical test for the untreated jute/epoxy and JREC filled with different concentrations of DAP a) UJE, b) UJ3DE, c) UJ9DE, d) UJ18DE, e) 9DJE, f) 27DJE, g) 54DJE.

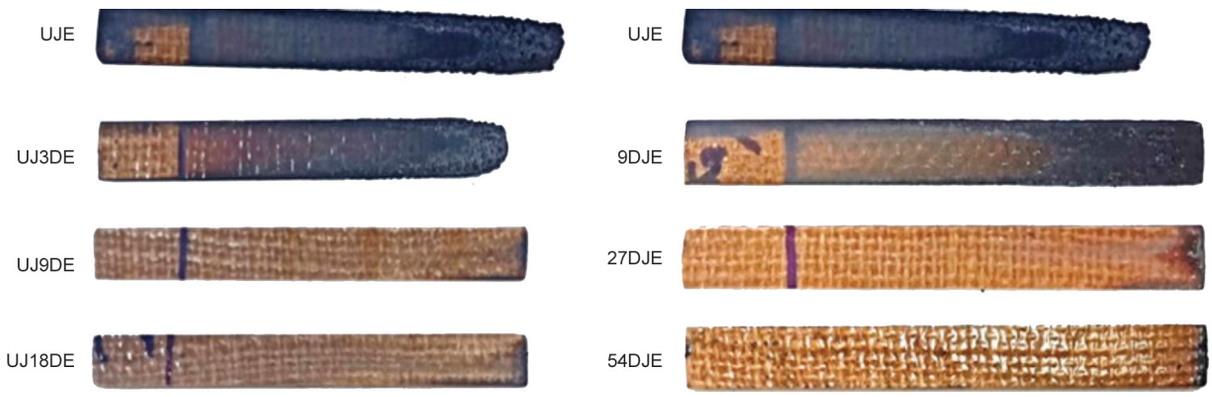

Figure 6. Composite samples after UL-94 vertical burning test.

composite systems with 12.5 wt% DAP, but different ethods of preparation, are 35.5% and 35.8%, respectively. Figure 5 demonstrates that neither UJ3DE nor 9DJE can pass the UL-94 vertical test. However, both successfully passed the UL-94 horizontal burning test, while 9DJE burns at a slower rate. The other composites with 6.25 and 12.5% DAP, regardless of preparation method, are all rated in the UL-94 test by V0 with no dripping. Figure 6 shows the composite samples after UL-94 vertical burning test. It is observed that UJE has burned completely. Figure 7 depicts a schematic illustration of the effect of adding DAP either to the matrix or reinforcement on the flame retardancy performance of the composites. It should be noted that both DAP concentration and DAP addition technique to composites influence DAP particle size and, as a result, particle volume fraction across the entire composite system. When DAP is introduced to jute fabric at a low concentration, the DAP particles are very small in size, causing an increase in particle volume fraction. When the same amount of DAP (2.25 wt%) is added to the epoxy matrix, the particle size increases, and the particle volume fraction decreases. Therefore, the larger the volume fraction of DAP in the composite system, the more char is produced during burning (Figure 7a and Figure 7b). In contrast, regardless of the DAP addition process, the incorporation of large amounts of DAP yields particle agglomeration and increases the amount of char produced during combustion (Figure 7c and Figure 7d).

3.4. Thermogravimetric analysis

TGA measurements were carried out under nitrogen atmosphere in order to determine the thermal stability and understand the flame retardation mechanism of treated jute fabric and JFREC treated with DAP. The recorded TGA and derivative thermogravimetric (DTG) curves of the different composites are plotted in Figure 8 and Figure 9, respectively, while additional data are summarized Table 5. The onset degradation temperature is taken at 5% mass loss. The thermal degradation of natural fibres occurs over several stages, including the dehydration of water, cross-linking of cellulose to form dehydrocellulose, decomposition of dehydrocellulose to produce char and volatiles, formation of levoglucosan, and decomposition of levoglucosan to produce flammable and non-flammable volatiles [49]. Cellulose, hemicellulose and lignin decompose at temperatures of around 260–400, 200–260and 160–800 °C respectively [49, 50]. From Table 4 and Figure 8a, it is observed that untreated jute decomposes in two steps, the first step for lignin and hemicellulose decomposition and the second step for cellulose decomposition. In the case of DAP-treated jute, decomposition occurs in three steps. The first step may be due to DAP decomposition and early decomposition of hemicellulose, second and third step refer to lignin and cellulose decomposition. Treating jute fabric with DAP results in two obvious facts: (1) as the concentration of DAP increases, more char residue remains, and (2) as the concentration of DAP increases, the temperature of maximum weight loss (degradation temperature) decreases. The reason behind these facts is that DAP degrades at a low temperature of around 175 °C to phosphoric acid and ammonia gas. This acid catalyzes dehydration and chain scission of cellulose fibres [27]. As a result, pyrolysis products decompose and vaporize at low temperatures below the boiling temperature of levoglucosan [51].

In terms of composite decomposition, it should be noted that composites produced by adding DAP to the matrix degrade in two steps, whereas composites prepared by adding DAP to jute fabric degrade in three steps (Figure 9b). This is due to the fact that,

like many natural phenomena, the first step of the combustion process begins on the surface with ignition. As a result, if DAP is added to the matrix, the ignition will begin on epoxy containing DAP,

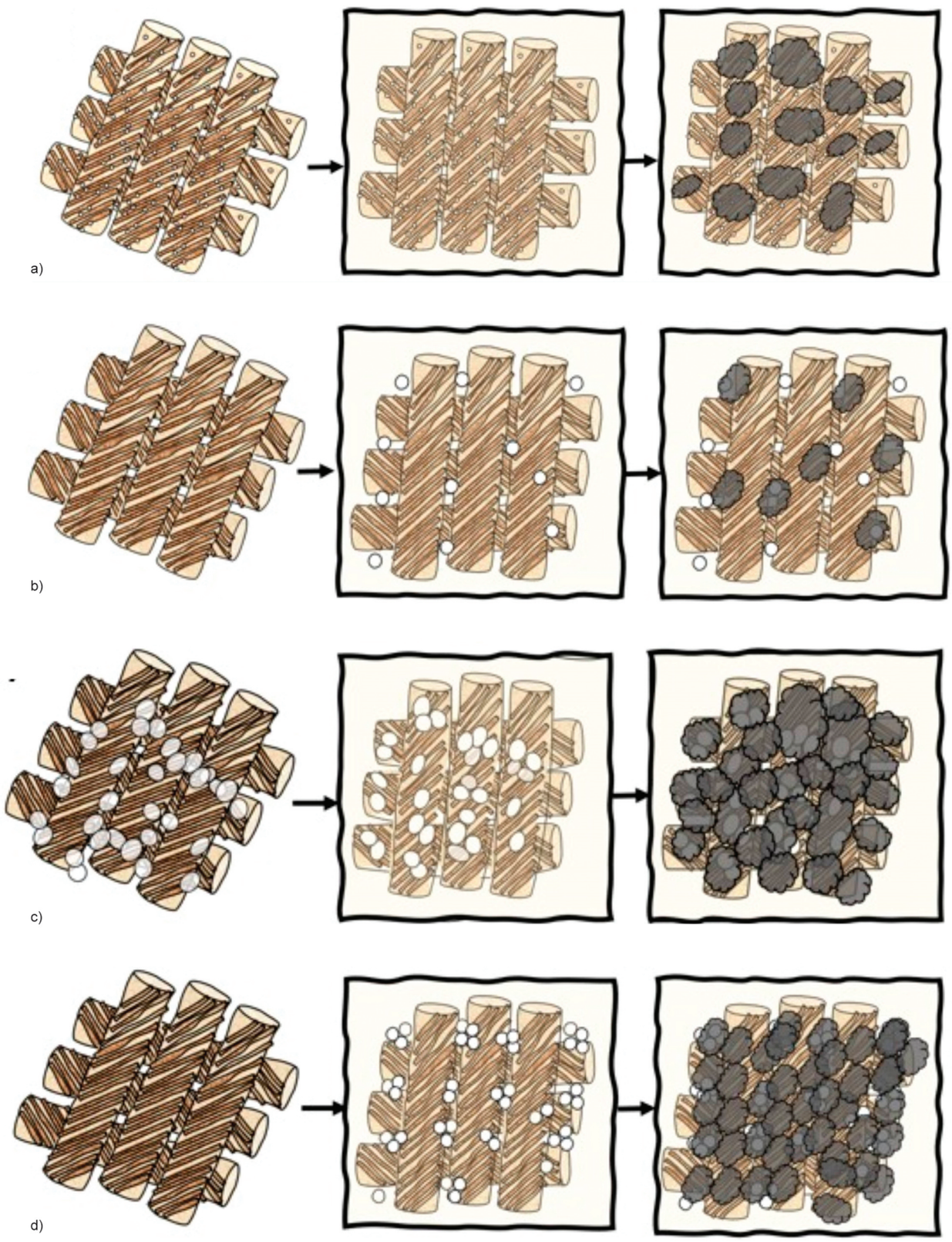

Figure 7. Schematic for the effect of DAP addition technique and DAP concentration on the flame retardancy performance of composites a) 9DJE (low concentration of DAP treated jute/epoxy), b) UJ3DE (untreated jute with low concentration of DAP/epoxy composite), c) 54DJE (high concentration of DAP treated jute/epoxy) and d) UJ18DE (untreated jute with high concentrated of DAP/epoxy composite).

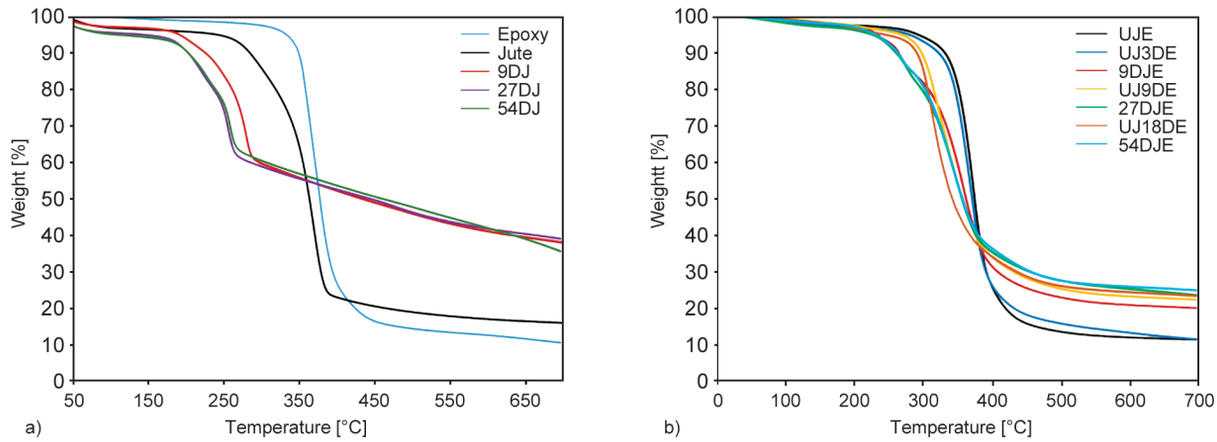

Figure 8. TGA curves for a) pure epoxy, jute and DAP-treated jute, b) epoxy/jute composites with DAP.

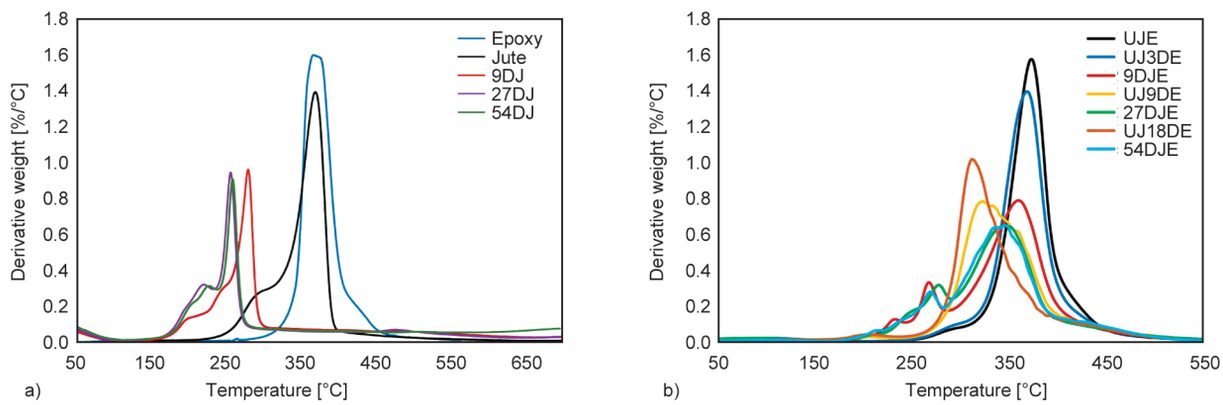

Figure 9. DTA curves for a) pure epoxy, jute and DAP-treated jute, b) JREC and epoxy/jute composites with DAP.

lowering the degradation temperature of the entire composite system. Furthermore, DAP decomposes in the epoxy resin matrix, forming a charring layer that shields the entire system from further combustion propagation. However, when DAP is applied to jute fabric, the ignition begins on pure epoxy resin, and the flame propagation continues until it reaches the DAP-treated jute fabric, therefore, the composite system goes through the same stages of degradation as treated jute.

3.5. Mechanical test results

Figure 10 shows the effect of DAP addition method (FR added to matrix abbreviated FRM, FR added to

Table 5. Extracted data from TGA and DTA curves.

Sample				$T_{5\%}$ [°C]	First step		Second step		Third step		Char residue at 700 °C
					Temperature [°C]	T_{peak} [°C]	Temperature [°C]	T_{peak} [°C]	Temperature [°C]	T_{peak} [°C]	
Epoxy				333.24	296–431	371.00	–	–	–	–	10.71
Jute fabric	Jute			241.14	182–303	285.60	303–395	369.01	–	–	16.02
	9DJ			188.57	124–227	201.86	227–268	244.3	268–314	278.79	38.04
	27DJ			148.43	135–189	194.28	189–242	218.29	242–275	255.75	39.15
	54DJ			115.00	134–195	200.64	195–239	226.06	239–278	255.72	39.29
Composites	0% DAP	UJE		296.90	247–321	292.00	321–425	372.77	–	–	11.42
	2.25% DAP	UJ3DE		279.10	234–314	289.41	314–424	367.62	–	–	11.55
		9DJE		232.31	172–241	230.97	241–289	267.93	289–404	359.02	20.00
	6.25% DAP	UJ9DE		270.26	134 –270	204.33	270–411	326.37	–	–	22.35
		27DJE		223.43	179–242	204.33	242–288	276.52	288–404	344.41	23.59
	12.5% DAP	UJ18DE		240.39	134–250	203.46	250–401	312.62	–	–	23.44
		54DJE		226.20	150–220	212.00	220–276	268.00	276–402	340.98	24.98

reinforcement abbreviated FRR) as well as DAP concentration on flexural strength (FS). It demonstrates that in the case of FRM, flexural strength for UJ3DE (2.25 wt%) has increased slightly by 3.3% compared to UJE, which has an FS of 91.2 MPa. UJ9DE (6.25 wt% DAP) has reduced FS by 10.3% that of UJE. Beyond 6.25 wt% DAP (UJ18DE), there is an insignificant change in FS compared to UJ9DE. To explain the loss in FS at 6.25% and beyond, where there is no subsequent drop in FS, it has to be considered that the mechanical properties of particle-reinforced composites are affected by both particle-particle interactions as well as the interfacial interaction between the particles and the polymer matrix [52, 53]. The van der Waals attractive force between particles rises as particle size decreases, resulting in particle agglomeration. This can explain the drop occurred in flexural strength by adding 6.25 wt% DAP. On the other hand, increasing DAP content beyond 6.25%, where the agglomerates of DAP are larger in size, will result in increasing the repulsion force between particle agglomerates and that in turn enhance the adhesion between the matrix and the particles. Thus, there is no subsequent decrease in FS beyond 6.25 wt% DAP. However, in the case of FRR, the flexural strength decreases continuously as DAP concentration increases. This can be attributed to the fact that jute fabric is treated with DAP solution, an alkaline medium with a pH value of 8, which may induce a chemical attack on cellulose fibres. Consequently, the mechanical strength of the jute fabric is reduced, lowering the FS for jute/epoxy composites. Therefore, the higher the concentration of DAP used to treat jute fabric, the more chemical damage occurs to the cellulosic fibre and the lower the mechanical properties of the jute fabric.

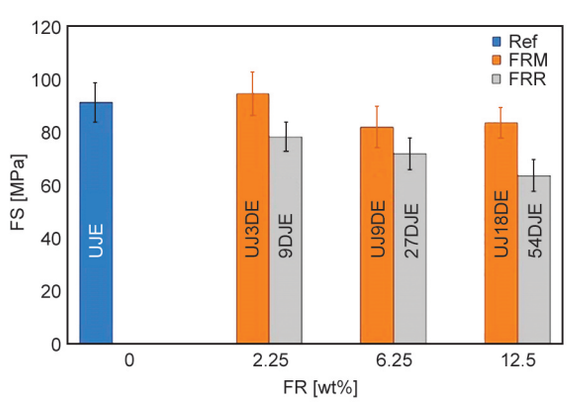

Figure 10. Flexural strength for Jute/epoxy composites with FRs.

Figure 11 shows the impact strength (IS) of the hybrid composites with respect to increasing DAP concentration as well as method of DAP addition to JREC. In contrast to flexural strength, it can be depicted that there is an increase in impact strength in the case of FRM with increasing particle content to 2.25% DAP, beyond which impact strength decreases but still has a comparable strength to UTJ composite. Also. In the case of FRR, there is an insignificant decrease in impact strength. To explain this behaviour, we should emphasize that the particle size, particle volume percentage, and particle-matrix interface all have an impact on the various toughening mechanisms in particulate-reinforced composites. Toughening mechanisms in particulate polymer composites include crack front pinning and crack tip blunting. The crack front pinning mechanism is more likely to occur when there is good bonding between particle and matrix bonding, whereas weak interfacial bonding between particle and matrix leads to fracture by crack tip blunting [54]. As previously stated, increasing DAP level above 2.25 wt% induces agglomeration and weak adhering to the matrix. Thus, we may conclude that up to 2.25 wt%, the composite has good bonding properties that supports the occurrence of the fracture by the front pinning mechanism, however, beyond 2.25% DAP, the weak bond enables the crack tip blunting mechanism to occur.

The fracture surface of impact specimens of JREC composites was studied by SEM. SEM micrographs of the 9DJE, UT3DE, 54DJE and UJ18DE were illustrated in Figure 12 respectively. Generally, the fracture surface of the specimens verifies the results obtained from the mechanical testing. The micrographs reveal that the particle size and shape are significantly influenced by both the DAP weight content and the preparation technique used. Specifically,

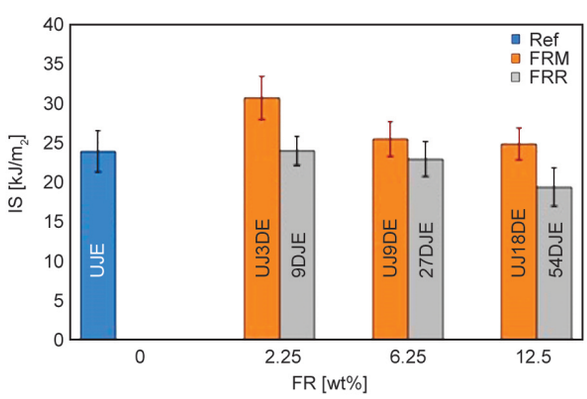

Figure 11. Impact strength for Jute/epoxy composites with FRs

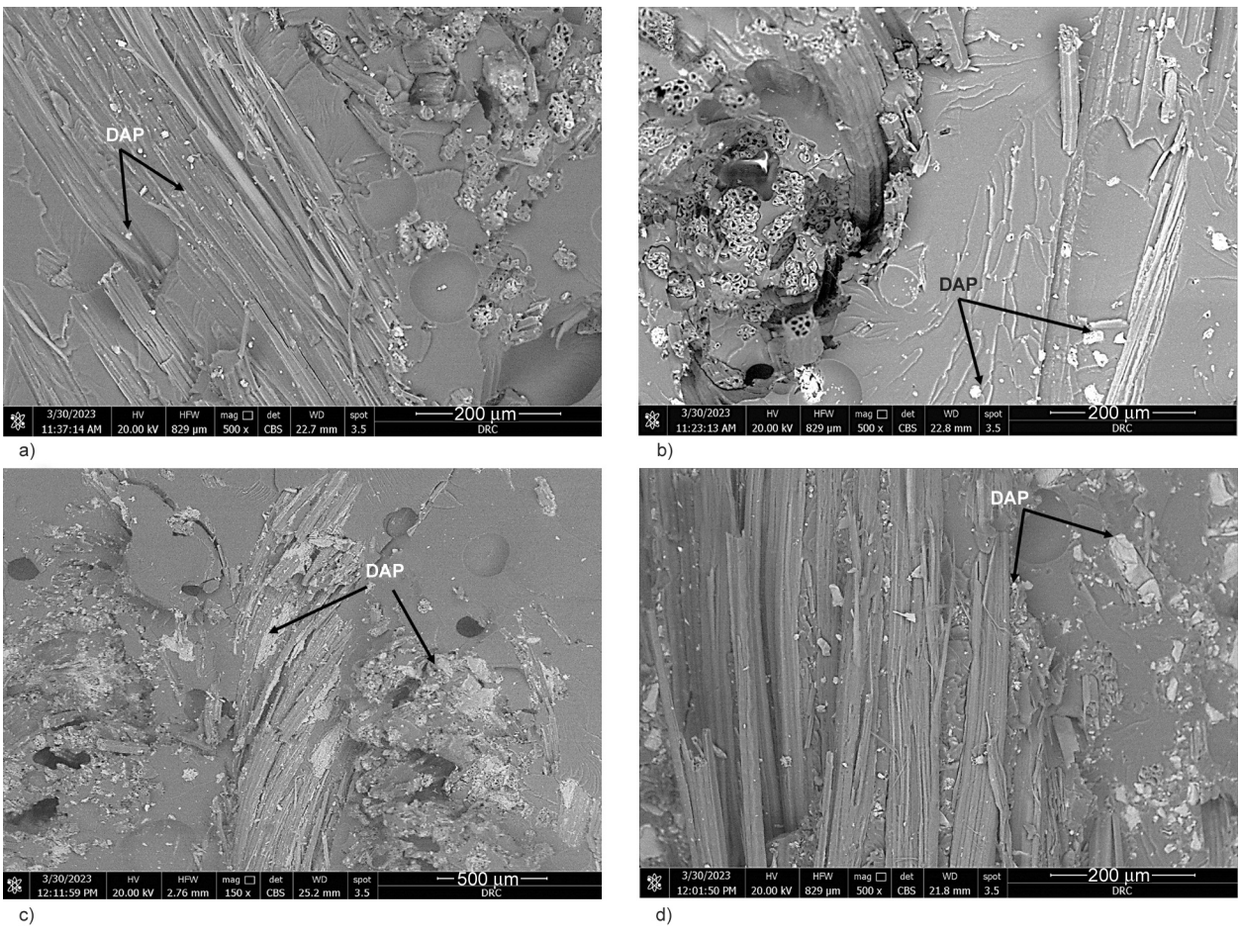

Figure 12. SEM micrographs of the impact fracture surface of a) 9DJE, b) UJ3DE, c) 54DJE and d) UJ18DE.

when DAP is added to jute fabric at a low concentration (2.25 wt%), the resulting particle size is smaller compared to adding DAP to epoxy resin (UJ3DE). Conversely, when high DAP content is incorporated either into jute fabric (54DJE) or epoxy resin (UJ18DE), the particle size becomes coarser and exhibits agglomeration.

4. Construction of flame retarded natural fibre epoxy composites selection charts

To summarise the effect of FR on both the flame retardance and mechanical properties results, Figure 13a and Figure 13b show selection charts that combine UL-94 and LOI test results with the mechanical strength (whether flexural strength (FS) or tensile strength (TS)) for this study and other results obtained from literature, respectively. It should be noted that although including FRs in various NF/epoxy composites improved flame retardancy performance, the mechanical strength was reduced. This sort of chart can assist the designer through entering it by the required mechanical strength and flame retardancy level to determine the appropriate NFRPC with FR for the desired application. The mechanical properties for each FR/composite system were normalized and plotted versus UL-94 and LOI, as shown in Figure 13c and Figure 13d, respectively, to provide a simpler chart that can easily evaluate the efficiency of FRs regardless of the type of composite system to assist researchers in tracking the latest research and identifying gaps of knowledge that need to be investigated. Therefore, relative tensile and flexural strength were calculated based on dividing the mechanical strength of the FR/composite system by the mechanical strength of the composite. The calculated value below one means that the mechanical strength has decreased. The points are named according to certain designation, which is matrix type_FR wt% FR type Fabric type_FR wt% FR type in the matrix for example EP_10 APPHemp_5ATH, this means epoxy matrix reinforced by hemp treated by 10 wt% APP and epoxy matrix filled by 5 wt% ATH. The points are evaluated according to their relative position to the origin. Highly ranked points are located in the upper right of the chart. Flax Fabric reinforced epoxy system filled with 12 wt% ammonium

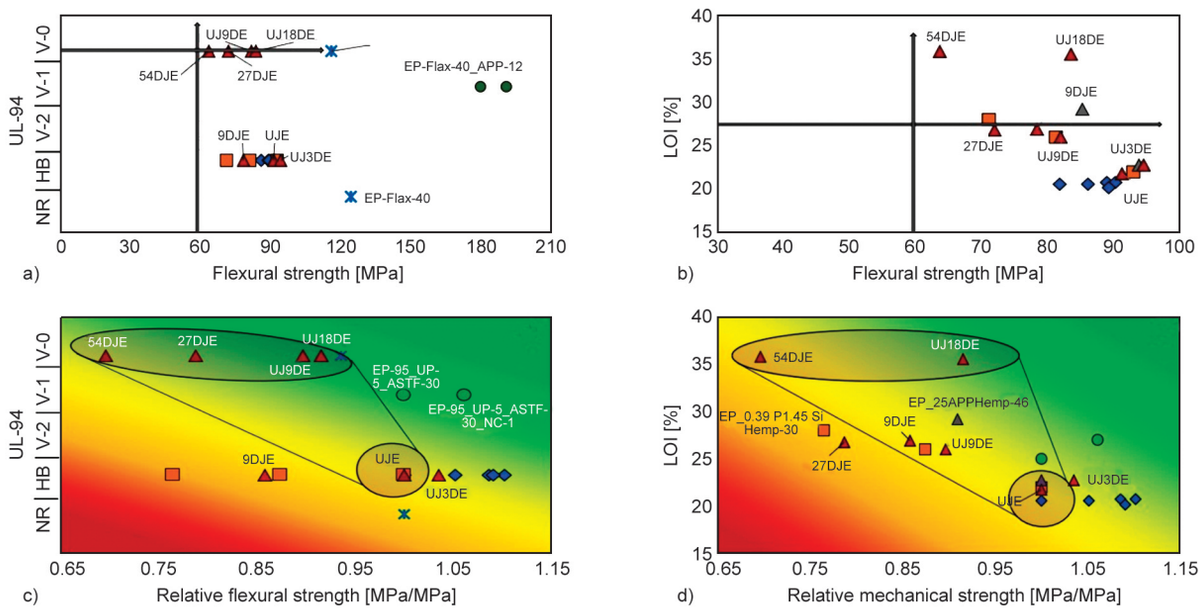

Figure 13. Selection chart for NFRPC with FRs a) UL-94 *versus* mechanical strength; b) LOI% *versus* mechanical strength; c) UL-94 *versus* relative mechanical strength, d) LOI% *versus* relative mechanical strength.
Symbols: ◆ – EP_BF (RFS) [30], ■ – EP_Hemp,P,Si (FS) [31], ✱ – EP_Flax,APP (FS [26]), ● – EP/UP_Sisal, NC (FS) [28], ▲ – EP_Jute, DAP (FS) [PW], ▲ – Vinisester_Hemp, APP (FS) [32].

polyphosphate shows the highest rank. Regarding this present work (PW), UJ18DE is concerned the best as it is located in the green zone.

5. Conclusions

In this study, flame-retardant jute fabric-reinforced epoxy resin composites were produced using two different approaches. On the one hand, DAP was added to the epoxy matrix, and on the other hand jute fabric was treated by DAP. At low DAP concentrations in the composite, the jute fabric modification reduced the flammability of the reference jute/epoxy composite more than the approach of adding DAP to the matrix. However, at higher DAP concentrations, the flame retardancy performance of jute/epoxy composites increased regardless of the method of incorporating FR into the composite system. Meanwhile, the technique of applying FR into JRECs has a considerable influence on the mechanical characteristics at all DAP concentrations. DAP was added with different concentrations (2.25, 6.25 and 12.5 wt%) to JREC. UJ18DE composite can be considered an optimal system that has enhanced flame retardancy performance without deterioration in mechanical strength. It passed V0 in the UL-94 test and achieved an LOI of value 35.5%. Additionally, various selection charts for flame-retardant natural fibre-reinforced epoxy composites were constructed. These charts correlate different flame retardancy levels with mechanical strength. Designers can quickly utilize these charts to determine the best flame retardant NFRPC for the desired application. Furthermore, researchers can make use of these charts to find knowledge gaps to study. Furthermore, new data can be used to update these selection charts.

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