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Influence of fabric properties, hybridization, and thermal aging on properties of flax/jute fibres reinforced epoxy hybrid composites

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Mechanical properties (tensile and three-point flexural), physical properties (density and porosity rate), and thermal properties of flax, jute, and flax/jute fibres reinforced epoxy composites have been investigated. Laminate composites are fabricated by vacuum molding with a total fibre weight fraction maintained at 40%. Results show that the hybridization with jute fibres can significantly improve the mechanical properties of flax composites, while the thermal stability is decreased. The thermal aging at 250 °C during 400 h causes a decrease in tensile and flexural strength of the resulting composites, whereas their rigidity is increased. Morphological studies of fractured surfaces and the evaluation of porosity have been evaluated using scanning electron microscope and optical microscopy respectively.

Keywords: Flax fibres, Jute fibres, Hybridization, Mechanical testing, Natural fibres, Thermal aging

1 Introduction

Natural fibres have interesting specific properties, such as a biodegradability, low density, lower pollutant emissions[1], low cost, specific mechanical and physical properties as compared to those reinforced by glass or carbon fibres[2, 3]. On the other hand, they have various disadvantages, such as poor thermal and dimensional stability, poor adhesion with matrix, weak moisture resistance and poor wettability. Hybridization between fibres can offer the possibility of having a material with high mechanical, physical and thermal properties by compensating the disadvantages of one fibre by adding the other. These mechanical and physical properties of the hybrid composites depend on the properties of fibres, fibres content, layering of fibres, fibre/matrix interface. For this purpose, several researchers have investigated the improvement in mechanical, physical and thermal properties of natural fibres reinforced composite by hybridization.

Ku *et al.*[4] proposed a very detailed study on the tensile properties of composites reinforced by natural fibres. In this review, various aspects such as the effect of volume fraction and surface treatment of fibre on the tensile strength of composites were discussed. Ramnath *et al.*[5] studied the hybridization between jute and flax fabrics with glass fabric to evaluate tensile, flexural, and interlaminar shear. The study shows that the natural hybrid composites have excellent properties under tensile and flexural loading. Chaudhary *et al.*[6] studied the mechanical properties of jute/epoxy, flax/epoxy, and hemp/epoxy composites. Results show that jute /hemp /flax reinforced hybrid composites have maximum Young's modulus of 1.80 GPa and highest tensile strength of 58.60 MPa. Lebrun *et al.*[7] studied hybridization between hemp, flax, and paper fibres. Their results show that the addition of two layers of paper to hemp/epoxy and flax/epoxy composites results in increasing the tensile strength and Young's modulus as well as delamination resistance. Akil *et al.*[8] demonstrated that the addition of pultruded jute fibre to a kenaf/glass fibre composites improves the flexural properties. Dhakal *et al.*[9] made composites based on flax and jute fibres. They found that the flexural properties of 100% jute based composites are higher than that of 100% flax. They further compared the effect of distilled water absorption on the mechanical properties of fibre-reinforced epoxy composites. Results showed that the moisture absorption percentage and diffusion coefficient were higher for reinforced jute composites than that for flax composites. Boopalan *et al.*[10] studied jute/ banana/epoxy hybrid composites. They reported that the addition of banana fibre up to 50 % by weight increases the thermal and flexural properties of jute/epoxy.

On the other hand, to study the thermal stability of natural fibres reinforced composite under high temperature, and its effect on their mechanical and

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physical properties, Ghasemzadeh *et al.*[11] investigated the hybridization between flax and glass fibres. Results showed that the addition of glass fibre to polypropylene/flax composites improves tensile modulus and strength as well as thermal stability. The study also showed that the thermal aging of polypropylene/flax/glass at 85 °C increases the stiffness and tensile strength, but decreases the elongation. Heat treatment of wood fibres at temperatures ranging from 150 °C to 260 °C has been studied by Boonstra *et al.*[12]. They found that the heat treatment increases the modulus of elasticity during the bending test but a reduction in flexural strength (-31%). In another study, Sridhar *et al.*[13] investigated the thermal stability of jute fibre. They found that thermal aging improves the flexural modulus, while the strength of jute fibres decreases.

The present study aims at developing flax/jute reinforced hybrid composites using epoxy with higher mechanical properties (tensile and three-point flexural), optimum physical properties (low porosity rate, low density), and high thermal stability after thermal aging at temperatures ranging from 150 °C to 250 °C as compared to flax/epoxy and jute/epoxy. The influence of hybridization, microstructure, and thermal aging on mechanical, physical, and thermal stability has been studied by morphological observation of fractured surfaces and the evaluation of porosity rate by scanning electron microscope and optical microscope.

2 Materials and Methods

2.1 Materials

The composites consist of an epoxy (EPOCAST 50 -A1 and HARDENER 946, Los Angeles, USA) with a density of 1.18 g/cm^3 reinforced flax and jute fibres with a plain weave architecture. It is composed of eight layers stacked, considering several combinations between the folds of jute and flax fibres. The areal density of fabrics is 298 and 262 g/m² for flax and jute respectively. The composition and characteristics of fibres found by several researchers[14-16] are reported in Table 1.

The composites manufacturing was conducted at the Algerian airline maintenance center (Algiers, Algeria). Five composite laminates were elaborated using vacuum molding technique as per the following procedures:

- The work table is cleaned with acetone to obtain a smooth plate.
- Deposit the sealant around the surface and form a rectangle (dimensions of our plate) to prevent the exit of the air during the evacuation.
- Deposit the impregnated fabrics in the rectangle sealant, followed by the different separator films (perforated film and tearing film) for the purpose of absorbing excess resin, and a drainage felt (it must not touch or exceed the sealant). Finaly, a vacuum bagging film is then positioned on the drainage felt. Each setup must be examined for leaks on the entire surface of the plate.
- Pump the air and recover the excess resin by inserting the vacuum pump hose between the vacuum film and the drainage felt.
- The polymerization of the resin is carried out for a period of 6 - 8 h; after this period, the plate is unmolded.

The sample code and composition of the laminate composites are given in Table 2.

2.2 Methods

2.2.1 Density Measurement

The density of the composites was measured according to ASTM D1895[17]. The composite mass and volume were determined using an analytical balance (RADWAG) with an accuracy of 10^{-4} g and a VERNIER caliper respectively. Before testing, all samples were dried at 75 °C for 2 h.

Table 1 — Composition and characteristics of flax and jute fibres

Parameters	Jute fibre	Flax fibre	Reference
Cellulose, %	61–75	64–85	14
Hemicellulose, %	11–15	5–20	14
Lignin, %	8–15	0–5	14
Density, g/cm^3	1.3–1.5	1.5	15
Moisture, %	12–14	8–12	15
Angle microfibrillar, deg	8	11	15
Elongation, %	2.0–3.0	2.0–4.0	16
Tensile strength, MPa	200–770	350–1040	16
Yong's modulus, GPa	20–55	28–70	16

Table 2 — Designation and composition of the flax/jute reinforced epoxy composites

Sample code	Composition, %	
	Jute	Flax
J0F100	0	100
J25F75	25	75
J50F50	50	50
J75F25	75	25
J100F0	100	0

2.2.2 Porosity Measurement

The porosity of the polished samples was calculated using the free software "ImageJ" (National Institute of Health, USA). A sufficient number of micrographs were taken by optical microscope at various positions from each sample in different locations (top, middle, and bottom). The pore size was calculated as the diameter of the equivalent area of the pore, and the porosity was calculated as the percentage of the area fraction of all the pores.

2.2.3 Thermogravimetric Analysis (TGA)

The thermal stability of the composites was studied using a thermogravimetric analyzer (TA Instruments Q500 NETZSCH, Germany). The composites samples were heated from 25 °C to 600 °C, with a heating rate of 10 °C/min in a nitrogen atmosphere (N_2).

2.2.4 Thermal Aging

2.2.4.1 Effect of Thermal Aging on Density and Weight Loss of Composites

The effect of temperature on the weight and density of the different composites (thermal aging) was studied in a forced air circulation oven operating at 150 °C and 250 °C during 7 h. The density and weight loss were determined for each sample.

2.2.4.2 Effect of Thermal Aging on Mechanical Properties

To investigate the influence of thermal aging on mechanical properties (tensile and flexural properties), a heat treatment was made in the oven on each composite under a forced air circulation at 250 °C during 400 h.

2.2.5 Tensile Test

The tensile test was carried out using an Instron model 5980 universal machine at 23 °C according to AFNOR NF ISO 57-105 standard (equivalent to ASTM D790-07 standard)[18] with dimension of 250 × 25 × 4.5 mm^3. Five specimens were tested and the average values were reported.

2.2.6 Flexural Test

The flexural test was carried out on an Instron 5980 machine at 23 °C, using the three-point bending fixture according to AFNOR NF ISO 57-105 standard (equivalent to ASTM D790-07 standard)[18] with dimension of 100 × 15 × 4.5 mm^3. The distance between supports is adjusted to comply with the condition imposed by the standards, Lo = 16h (h = 4.5 mm). Five specimens were tested and the average values were reported.

2.2.7 Morphological Analysis

The morphology of the different composites was observed with a JEOL JSM 6360 scanning electron microscopy (SEM) at different magnifications. SEM micrographs were taken from fractured surfaces of samples after the tensile and flexural tests. Before SEM observation, the fractured surfaces were coated with a thin carbon layer.

3 Results and Discussion

3.1 Density

The density values of the composites having different volume fractions of fibres are illustrated in Fig. 1. It can be observed that the increase in jute fibre content decreases the density of epoxy reinforced composites, which can be attributed firstly to the low density of jute fabric (with areal density of 2.618 g/cm^2) compared to that of flax (2.98 g/cm^2). Moreover, the higher density of the flax fibres is attributed to the fact that these fibres have much smaller lumens compared to the jute fibres, as reported by Oksman *et al.*[19]. Secondly, due to their different chemical compositions[14], the densities of cellulose and hemicellulose are higher than that of the other components of the fibres (lignin)[20], thus the fibre density increases. Finally, the pore size between threads of fabrics is larger in jute fabric as compared to that in flax fabric.

3.2 Porosity Measurement

The porosity of the composites with different volume fractions of fibres is plotted in Fig. 1. It is noted by the "ImageJ" that the porosity type is a closed porosity, which has a weak influence on the mechanical properties of the composites[21, 22]; unlike

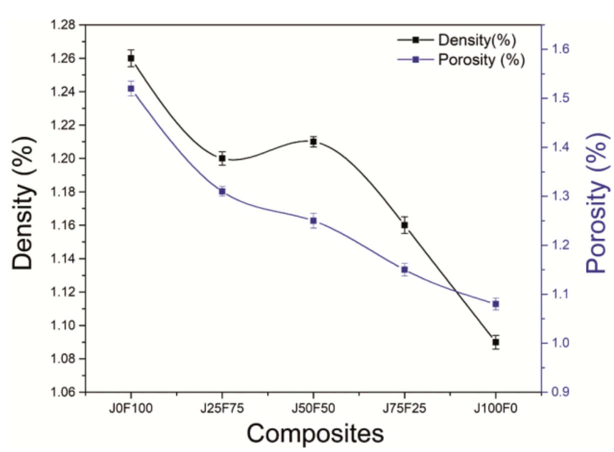

Fig. 1 — Change in composites density and porosity according to fibre volume fractions

open porosity, which has the distinction of connecting at least two faces of the porous material.

According to the estimates given by the “ImageJ" analysis, it is noted that the porosity scales in the composites are micro and mesoscopic. The first type of porosity is caused by the high viscosity of the resin at the time of impregnation (capillarity) and the existence of luminal cavities in plant fibres, which are identified by air-filled cavities in the fibres (fibre porosity). This porosity scale (microscopic porosity) has a weak influence on the mechanical properties of the composites if it is well distributed [22]. The second type (mesoscopic porosity) is broken down to three states. Firstly, the gaps between the fibres and matrix are caused by the heterogeneous form and dimensions of plant fibres (due to the air-filled cavities at the fibre/matrix interface regions)[23]; secondly, there are compatibility between jute/flax fibres and matrix (interface porosity); and finally, due to the wettability of the matrix in the bundles (impregnation porosity). The macroporosity is absent which is due to good permeability of the resin in the plain weave architecture.

Figure 1 shows a decrease in the porosity with the addition of jute fibres to flax reinforced epoxy composites because of the difference in the chemical composition, morphology, and the adhesion between the jute/flax fibres and the matrix. There is good wettability of the matrix in the jute bundles as compared to that of flax, which decreases the voids between jute fibres of the same bundle (impregnation porosity)[24]. This result may be interpreted by a good adhesion between jute fibres and matrix. Moreover, the bundles twisting can also influence the composite porosity, because of the fact that the maximum level of twisting reduces the permeability of the matrix in the bundles (jute fibre contains a minimum level of twisting compared to flax)[25]. Finally, the difference in the pore size of flax and jute fabrics (jute fabric has larger pores size than flax fabric) causes increase in the permeability of the matrix in the fabric, and decrease in the interface porosity between the bundles and the matrix, as illustrated in Fig. 2 (interface porosity). Rahman *et al.*[26] observed that the increase in pore size in fabric increases the air permeability and moisture.

Moreover, it should also be pointed out that the density and porosity of the laminate composites follow a similar trend, as shown in Fig. 1. As we increase the jute content, the density and porosity decrease simultaneously. This behavior can be mainly attributed to the low porosity with a small difference between all the composites (from 1.08% for J100F0 to 1.55% for J0F100). Besides, the flax density (298 g/m²) is higher than that of jute (261 g/m²). Thus, it has been found that the composites with a high flax content have the highest density despite having a relatively high porosity rate than that based on jute.

3.3 Effect of Hybridization on Tensile Properties

Figure 3 shows the tensile test results where the addition of fibres to the epoxy resin remarkably improves the tensile properties. It can also be seen that the addition of jute fibre to a flax fibre-based composite increases Young’s modulus with the percentages of 1.30%, 10.40%, 11.60%, and 19.60% for the composites J25F75, J50F50, J75F25 and J100F0 respectively as compared to J0F100.

Concerning the tensile strength, we notice an improvement after the addition of jute fibres especially for the J50F50 and J70F25 with a percentage improvement of 10.70% and 1.03% respectively. Those remarkable improvements in stiffness and tensile strength properties of flax fibres reinforced epoxy composites after the addition of jute fibres are because of the excellent adhesion between the jute fibres[6, 9]. At the same time, the good wettability of the epoxy matrix in the jute bundles as compared to that in the flax can also improve these properties, which increases the contact area and adhesion between the jute fibres and the matrix [Fig. 2 (b)]. Chaudhary *et al.*[6] studied the tensile

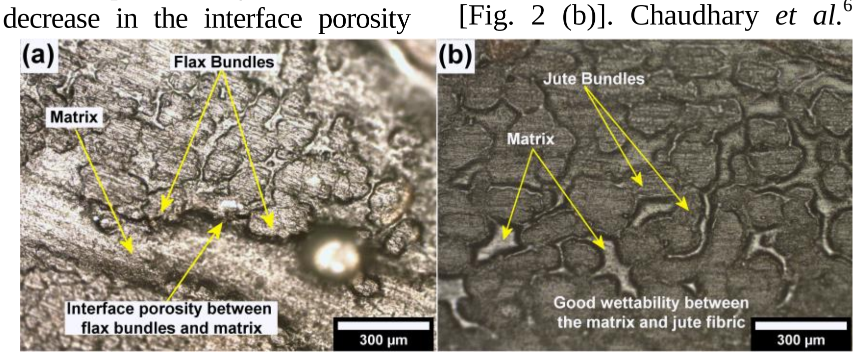

Fig. 2 — Optical micrographs of the composites (a) J0F100 and (b) J100F0

properties of jute/epoxy, flax/epoxy, and hemp/epoxy composites. They found that for flax/epoxy and jute/epoxy composites the respective values of Young's modulus and tensile strength are 1.60 GPa & 46.20 MPa, and 1.60 GPa & 43.30 MPa. Ramnath *et al.*[27] performed a comparative study between composites based on jute, abaca and glass fibres. They found that jute fibres have a good wettability as compared to abaca, and, therefore, good load transfers. This excellent adhesion between jute fibres and matrix is related mainly to the excellent treatment of jute fibres before their use, including chemical[28], physical[29], and biological[30] treatment, eliminating the amorphous parts of the fibres. These parts are responsible for the poor adhesion between fibres and matrix. Secondly, there is difference between the microfibril angle of the two fibres[15]. In fact, the smaller the microfibril angle, the higher is the rigidity and resistance of the fibre [31]. Finally, the difference in the chemical composition of the two fibres is found, considering percentage of cellulose, hemicelluloses and lignin[14], because the rigidity of these components is not the same[32].

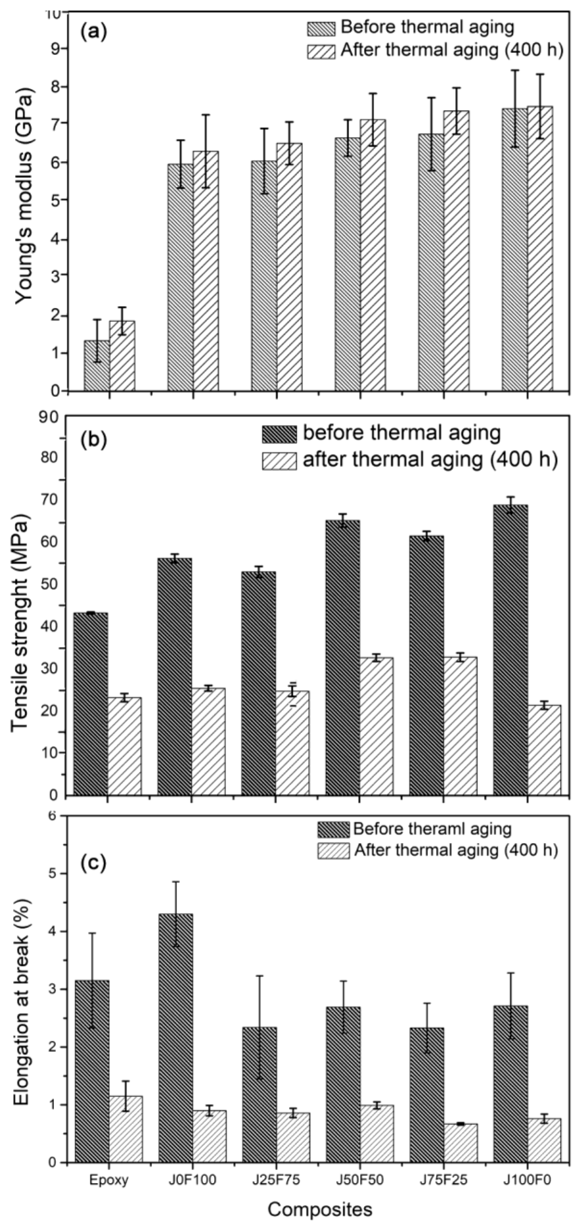

Fig. 3 — Tensile properties of flax/jute reinforced epoxy composites before and after thermal aging (400 h)

3.4 Effect of Hybridization on Flexural Properties

Figure 4 presents the flexural properties of flax/jute reinforced epoxy composites. Flexural strength of epoxy increases after the addition of jute to the flax fibres reinforced epoxy composites. This improvement in epoxy flexural strength is due to the ability of natural fibres to resist at the bending force.

Hybridization between jute and flax fibres improves the mechanical properties such as flexural modulus and flexural strength of composites. This improvement in flexural properties after the hybridization of the two fibres leads to the high capacity of composites in front of the shear and compressive strength, because bending test represents a tensile, compression, and shear stress of specimen.

Compression resistance properties of kapok/sisal-unsaturated polyester hybrid composites are investigated according to fibre content by Reddy *et al.*[33]. They found that the addition of sisal fibres to kapok reinforced composite increases the compressive properties of the hybrid composites. Some researchers have done the similar type of study using two or more fibres as reinforcing the composites. Oksman *et al.*[19] investigated the flexural properties of flax/PP and jute/PP; results show that jute/PP has flexural modulus and strength higher than that of flax/PP. Chaudhary *et al.*[6] studied the flexural properties of jute/epoxy, flax/epoxy, and hemp/epoxy composites; results reveal that the hybrid composite flax/jute/hemp/epoxy has the highest flexural modulus and flexural strength as compared to flax/epoxy, jute/epoxy, and hemp/epoxy. Alavudeen *et al.*[34] reported that the bending properties are high in hybrid banana/kenaf/epoxy composites as compared to that in banana/epoxy and kenaf/epoxy. Boopalan *et al.*[10] studied jute/ banana/epoxy hybrid composite. They found that the addition of jute fibre with a percentage of 50% to the composite banana/epoxy increases the thermal and flexural properties of banana/epoxy. Das *et al*.[35] hybridized jute with paper fibres. The results show that the addition of jute fibres increases the flexural properties of composite paper/epoxy.

It is observed that the increase of flax percentage in hybrid composites tends to make it ductile. Indeed, there is an increase of the flexural strain (Y) from 7.01 mm of the composite J0F100 to 9.14 mm of the composite J100F0.

3.5 Thermal Aging

3.5.1 Effect of Thermal Aging on Weight Loss and Thermal Stability of Composites

Figure 5 presents the weight losses of the composites at different temperatures (150 and 250 °C).

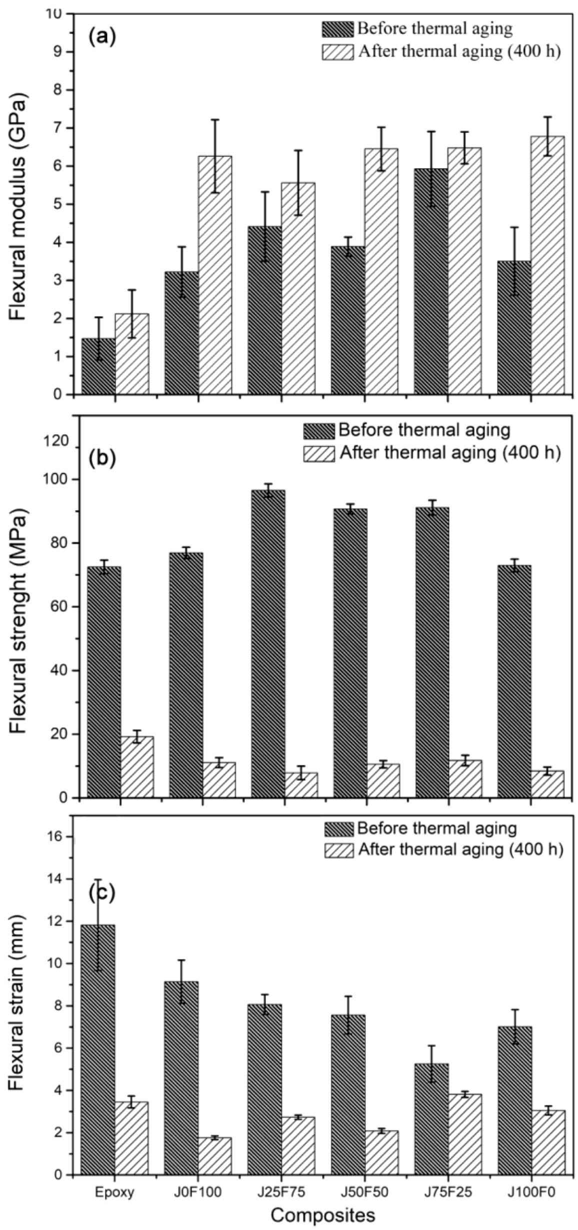

Fig. 4 — Flexural properties of flax/jute reinforced epoxy composites before and after thermal aging (400 h)

To study the effect of increase in temperature on the weight losses of the composites, the temperatures (150°C and 250 °C) are maintained for 7 h. It can be observed that all curves follow a similar trend, a first quasi-linear part followed by a second asymptotic part. The linear part corresponds to the first step (weight loss by thermolysis), which represents the release of volatiles & acids, after the chain rupture, and the evaporation of moisture from the samples. The weight losses are between 0.7% and 2.8% for 150 °C and between 2.7% and 4.70% for 250 °C with a maximum time of about 1 h.

The asymptotic part represents the thermo-oxidation (diffusion of oxygen in the composite materials). A remarkable decrease in the weight losses is observed up to the stabilization. The oxidation (burning) is limited to only the amorphous zones or the interphases crystalline-amorphous zones. This difference in oxidability between the two phases is commonly attributed to the permeability of the amorphous zones to molecules such as dioxygen[36].

It is observed from the Fig. 5 that the incorporation of fibres decreases the overall thermal stability of the epoxy. It is also observed that the increase of jute fibres increases the weight losses at both the temperatures 150 °C and 250 °C. The reason for this increase is expected to be the degradation process in

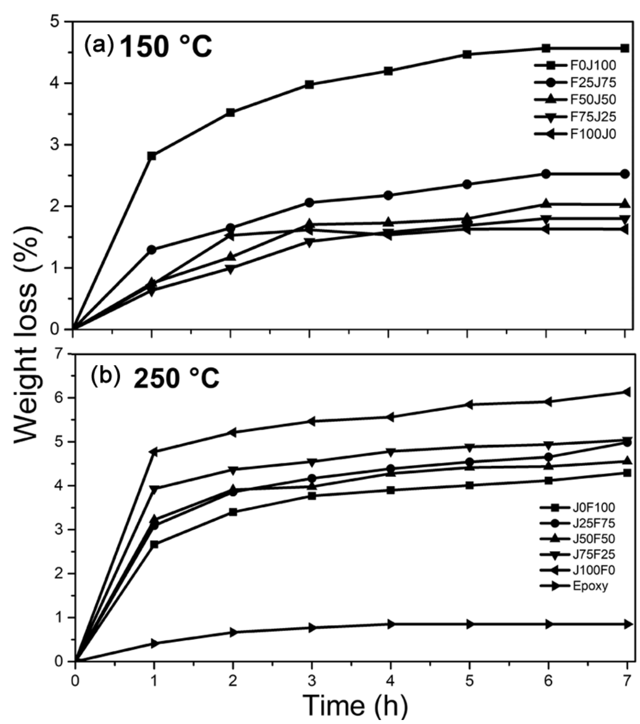

Fig. 5 — Weight losses of different composites as a function of time at (a) 150 °C and (b) 250 °C

the first part owing to the high moisture of jute fibres in comparison with the flax fibres[9], and the high level of lignin in jute fibre which begins to degrade partially between 110 °C and 200 °C; condensation reactions are present and there is real degradation that would intervene only beyond 400 °C. An increase in weight losses is observed with temperature increasing from 150 °C to 250 °C. For example, after 4 h, the weight losses at temperature 150 °C are 1.5% for F100J0 and 4.25% for F0J100, while at 250 °C; the corresponding values are 3.75% for F100J0 and 5.70% for F0J100. This variation in weight losses leads to the elimination of hemicelluloses when the temperature increases to 250 °C, which starts degrading at temperature range 225°-325 °C. This confirms many results reported for other lignocellulosic materials, and the degradation of the carbohydrates in the lignin samples between 180 °C and 250 °C which are converted into volatile gases, such as CO, CO_2, and CH_4[37].

The thermal stability of pure epoxy and flax/jute fibres reinforced hybrid composites is also evaluated using a thermogravimetric analysis (Fig. 6). From TGA curves, it is evident that the incorporation of fibres decreases the overall thermal stability of the epoxy. The thermal degradation of pure epoxy occurs in one step at much higher temperature, but it exhibits significantly higher weight loss with temperature, which is mainly attributed to the degradation of their network. However, it can be seen that all composites display two main parts of weight loss, viz (i) the initial weight loss of composites occurs at low temperature range 90-250 °C, due to vaporization of adsorbed water or moisture, the elimination of hemicelluloses, and the degradation of carbohydrates; and (ii) the second part is started in the temperature range 250-500 °C, and it presents the decomposition of fibre components (cellulose at 350 °C and lignin beyond 380 °C), and the epoxy matrix. Moreover, with the hybridization of flax fibres with jute fibres, the thermal stability of epoxy composites is affected within a similar temperature range 250-400 °C. Therefore, the highest the jute content, the lower is the thermal stability. Thus, the thermal stability of flax fibres is better as compared to jute fibres. However, further heating to 600 °C increases the final residue with the jute fibres addition, but with relatively less residual content for the F0J100. These results might be due to higher lignin content present in jute fibres, where the lignin degradation occurs slowly when exposed to high temperature[38].

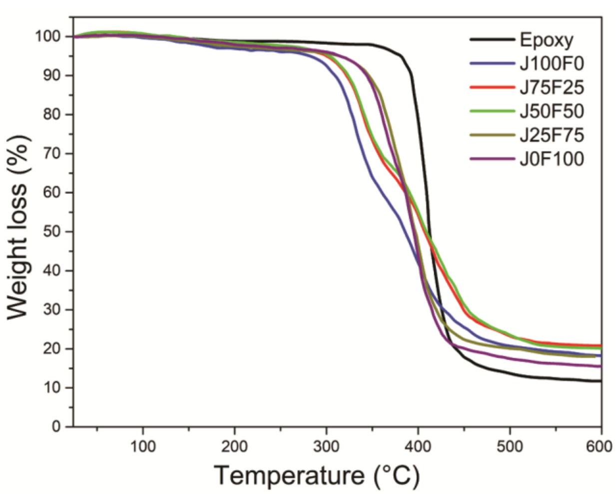

Fig. 6 — TGA curves of pure epoxy and flax/jute reinforced epoxy hybrid composites

3.5.2 Effect of Thermal Aging on Tensile Properties

Figure 3 shows that the thermal aging increases Young's modulus of the epoxy and all composites, decreases the tensile strength of epoxy and all composites, and decreases the elongation of epoxy and all composites.

The increase in Young's modulus after thermal aging can be attributed to the reactions into the main polymeric components of composites (cellulose, hemicellulose and lignin, moisture). The thermal aging at 250 °C causes a degradation of hemicellulose (hemicelluloses are the most thermal-chemically sensitive to temperature which degrades from 225 °C with a decrease of galactose, xylose and arabinose content) [12], and recrystallization of the amorphous regions of hemicellulose (xylan and mannan), which increases the relative amount of crystalline cellulose, therefore increasing Young's modulus of composites. Zeronian *et al.*[39] stated that the thermal aging increases the crystallinity of the fibre. For example, the amorphous part of the cellulose (*Fam)* decreases from 0.38 to 0.30 when heated at 240 °C for 6 h. On the other hand, the cross-linking of lignin network, in fact, acts as a matrix, holding together the cellulose microfibrils (polycondensation reactions are present and not a real degradation that would intervene only beyond 400 °C)[37], and has an influence on Young's modulus, because it makes the structure around the microfibril/fibril of cellulose rigid.

Moreover, the elimination of some amount of moisture after the thermal aging makes the composites more rigid. Mathew *et al.*[40] investigated the effect of heat treatment on the crystallinity and dynamic modulus of the PLA-based composites in a hot air oven at 80 °C for 3 days. They found an improvement in the dynamic modulus and thermal stability of the composite by the increase in the crystallinity.

The reduction in tensile strength after thermal aging is attributed to the cleavage of secondary bonds between hemicellulose and cellulose on the one hand, and the covalent bonds between hemicellulose and lignin on the other hand. Therefore, the load-sharing capacity of the lignin-hemicellulose matrix, which can be considered as a coating for the unidirectional cellulose microfibril/fibril, is reduced. This behavior does not match those reported by Ghasemzadeh *et al.*[11], who found an increase in tensile strength as the treatment temperature (85 °C) is not found enough to achieve this transformation. On the other hand, the effect of natural defects presented in the composites after thermal aging, such as resin pockets, voids, and pores represented by the decrease in the density of composites and weight losses [Figs 5 (a) and (b)], decreases the tensile strength which is sensitive to these defects.

Meanwhile, fibre-epoxy interface interaction can take place between the hydroxyl groups (-OH) of the main backbone chain of the matrix and the fibres surface, resulting in hydrogen bonds. However, the major problem of the natural fibres is its hydrophilic behavior [41]. Water molecules tend to attract and react with the hydroxyl group (-OH) of the fibre's cellulose, and thus, it inhibits their effective reaction with the matrix. However, thermal exposure to high temperature leads not only to complete water removal but also to degrade the fibre's components (pectins and the cellulose above 180 °C and 230 °C, respectively)[42]. Therefore, thermal aging affects both fibres and fibres-matrix interface properties by developing defects, such as porosities. Thus, it leads to a drop in mechanical properties of the composites. The work of Baley *et al.*[43] showed that drying flax fibres causes a reduction in their mechanical properties, which is transferred to the composite. They also reported that the moisture removal will affect the properties of the fibre surfaces, thus, results in poor fibre/matrix interfacial bonding. However, in contrast, Doan *et al.*[44] showed an increase in the tensile strength of jute/ polypropylene composites in humidity aging conditions, which can be attributed to the improved both polymer and interfacial adhesion strength.

3.6 Effect of Thermal Aging on the Flexural Properties

It is observed from the results obtained after the flexural test, that the effect of thermal aging on the flexural properties is the same to those of the tensile tests, with a significant improvement in flexural modulus, and significant reduction in flexural strength & flexural strain.

Bezerra *et al.*[45] studied in the thermal aging of a composite based on carao fibres placed in forced air circulation oven operating at 110±5 °C. They reported an increase in flexural modulus and a decrease in flexural strength of the matrix and composites significantly after 40 days of the test, which may be related to polymer degradation upon exposure to heat.

3.7 Morphological Analysis

Figures 7 (a) and (b) show that the rupture of jute and flax fibres is slightly ductile. The external surface of the fibres is rough with hemicelluloses, lignin, and other sugars remaining even after chemical treatments. Moreover, it is not recommended to completely eliminate them since they act as binders between the fibrils of cellulose[19], compared to that of glass fibres (smooth external surface). However, between the flax and jute fibres, the fibres are separated in the flax bundles unlike the jute fibres, which remain glued in the bundles; this can be explained by the fact that the lignin in jute fibres is relatively higher as compared to that in flax fibres, which acts as binders between the fibres. The presence of lignin contributes to enhance the cohesion and causes an improvement in the mechanical properties of composites.

Figures 7 (a) and (b) also show that flax and jute fibres break at different heights, often at the nodes of the fibres. It can be seen that the fibres belonging to the same bundles do not break in the same plane (ductile behavior), unlike the glass or carbon fibres that break in the same plane (brittle behavior). Figures 7 (c) and (d) show that the diameter of jute fibres is higher than that of flax, as confirmed by Ahmad *et al.*[46].

Moreover, it is observed that the fracture surfaces do not present delamination phenomenon. This is maybe attributed to the fabrics weaving, which is the same. It tends to increase the delamination resistance of the material, and it is difficult for cracks to propagate. Indeed, this delamination phenomenon slightly occurs when the laminate composite is composed of folds with different orientations or unidirectional fibres, and the plain weave architecture of the fabrics. Alif *et al.*[47] studied the effect of weave structure on the interlaminar fracture behavior of orthogonal woven fabric glass/epoxy and carbon/epoxy composite laminates. They found that the delamination resistance of plain weave architecture is higher than that of the twill and satin weave armature. Another reason for the absence of delamination is the low porosity in the composites,

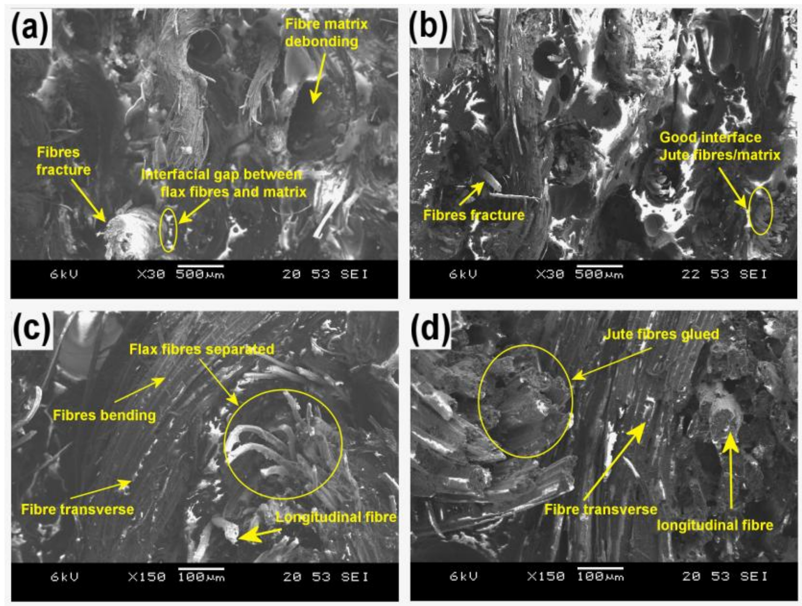

Fig. 7 — SEM micrographs of fractured tensile and flexural surfaces of composites before thermal aging (a) tensile test J0F100, (b) tensile test J100F0, (c) flexural test J0F100, and (d) flexural test J100F0

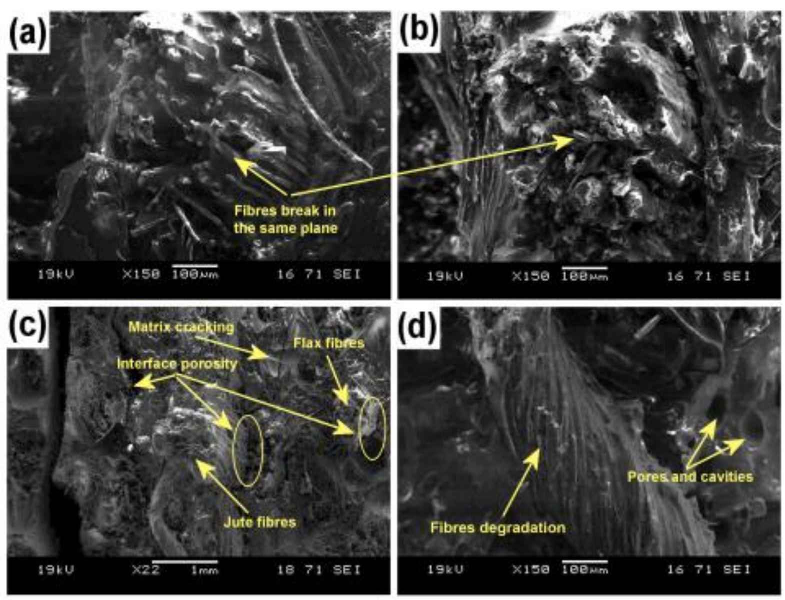

Fig. 8 — SEM micrographs of fractured tensile and flexural surfaces of composites after thermal aging (400 h) (a) flexural test J0F100, (b) flexural test J100F0, (c) flexural test J50F50, and (d) tensile test J50F50

which increases the resistance to delamination[48]. The transverse rupture is almost non-existent for the same reason mentioned for delamination.

Figure 8 (c) exhibits the presence of microcracking in the matrix after thermal aging, which can be attributed to the degradation of the matrix. It is clear from Figs 8 (c) and (d) that the thermal aging causes damage to the composites like pores and cavities, which confirm the decrease in the weight losses of all composites after thermal aging.

Figures 8 (a) and (b) show that jute and flax fibres belonging to the same bundles break in the same plane after thermal aging (brittle behavior), which confirms the increase in Young's modulus values and decrease in elongation, unlike before thermal aging when flax and jute fibres break at different heights, as illustrated in Figs 7 (a) and (b).

4 Conclusion

The present study deals with the mechanical and thermal properties of flax, jute and flax/jute fibres reinforced epoxy composites. Based on the obtained results, the following facts can be drown:

4.1 Variation in composites density and porosity according to fibre volume fractions is noticed due to the difference in chemical composition, morphology between the two fibres, and the fibre-matrix interface.

4.2 An improvement in the mechanical properties, such as tensile and flexural, after hybridization between jute and flax fibre is observed. Hybridization causes the following effects:

- An increase in mechanical properties of epoxy after the addition of jute and flax fibres.
- An increase in stiffness of the composites after the addition of jute to flax fibres reinforced epoxy composites.
- Improvement in flexural properties (flexural modulus, flexural strength, and flexural strain) of all hybrid composites after the hybridization between jute and flax fibres.

4.3 The decrease in thermal stability of the epoxy at 150 °C and 250 °C after the addition of jute to flax fibres is reported, along with the reaction of the degradation products of the fibres on the matrix. TGA analysis can further confirm the above results.

4.4 The thermal aging at 250 °C causes a decrease in tensile and flexural strength of the composites, but an increase in the elasticity modulus.

4.5 SEM observations show that the rupture of jute and flax fibres is slightly ductile.

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