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THERMOGRAVIMETRIC BEHAVIOR OF EPOXY COMPOSITES REINFORCED WITH CONTINUOUS AND ALIGNED JUTE FIBERS[1]

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Abstract
A wide variety of natural lignocellulosic fibers are increasingly being considered as viable alternatives to replace glass fiber in polymer composites. The application of lignocellulosic fiber composites in civil construction and automobile industries requires, however, information not only on the mechanical properties but also the thermal stability. In spite of numerous research works on jute fiber composites, the thermal stability of these composites with epoxy matrix has not yet been investigated. Therefore the present work evaluates the thermal stability of epoxy composites reinforced with up to 30 vol% of continuous and aligned jute fibers by thermo gravimetric, TG/DTG, analysis. The presence of jute fibers induces sensible changes in the thermal stability of the epoxy composites.
Key words: Jute fiber; Epoxy composites; TG/DTG analysis; Thermal stability.

COMPORTAMENTO TÉRMICO DE COMPÓSITOS DE EPÓXI REFORÇADOS COM FIBRAS CONTÍNUAS E ALINHADAS DE JUTA

Resumo
Uma ampla variedade de fibras naturais lignocelulósicas está cada vez mais sendo considerada como alternativa viável pra substituir a fibra de vidro no reforço de compósitos poliméricos. A aplicação de fibras lignocelulósicas na indústria de construção civil e automobilística exige informações não apenas sobre as propriedades mecânicas, mas também a estabilidade térmica. Apesar de numerosos trabalhos de pesquisa sobre compósitos de fibra de juta, a estabilidade térmica desses compósitos com matriz de epóxi não foi ainda investigada. Portanto, o presente trabalho tem por objetivo investigar o comportamento térmico de compósitos de epóxi reforçados com até 30% em volume de fibras contínuas e alinhadas de juta, através da análise termogravimétrica, TGA/DTG. A presença de fibras de juta induz sensíveis alterações na estabilidade térmica dos compósitos de epóxi.
Palavras-chave: Análises térmicas; Compósitos de epóxi; Fibras de rami.

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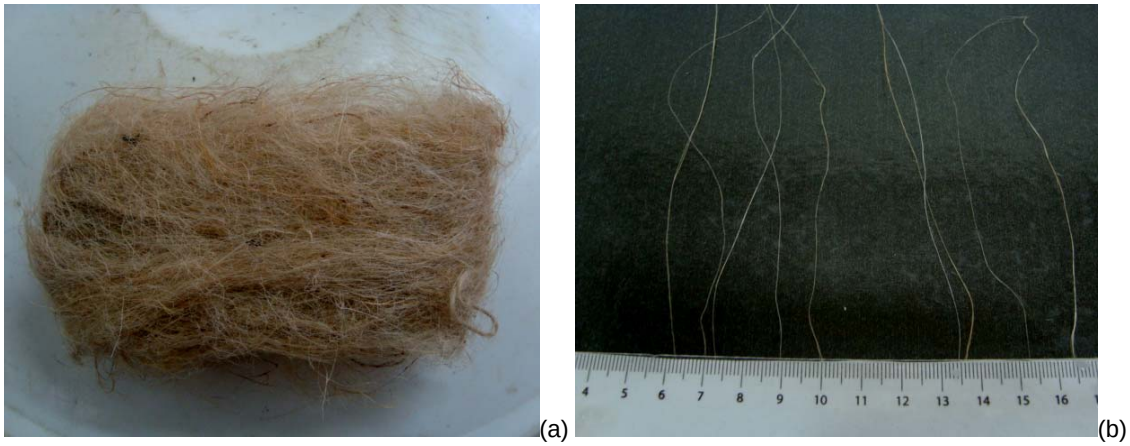
1 INTRODUCTION

In past decades the world is ever more demanding materials that are not only less intensive in terms of processing energy but also environmentally friendly. Actually, global issues like generalized pollution and the planet warming[1] are renewing the interest of natural materials in substitution for synthetic ones.[2] In particular, natural lignocellulosic fibers obtained from cellulose-rich plants are gaining attention as an alternative to more expensive synthetic fibers that are also associated with CO_2 emission, which is the main responsible for global warming. In fact, lignocellulosic fibers such as jute, sisal, and hemp among others, present comparative environmental, economical, social and technical advantages.[3-7] In particular, the jute fiber has in recent years been investigated as an effective reinforcement of polymer composites,[8,9] some of which are already applied in civil construction parts[10] and automobile components.[10,11] Despite the superior mechanical strength and stiffness contribution of the jute fiber, its effect on the thermal stability of composites is still a matter of discussion.[12-18] Since the application of these jute fiber composites may involve relatively higher temperatures during processing as well as in use condition, their limits of thermal stability need to be specified. The literature failed to report investigations on the thermal behavior of epoxy composites reinforced with jute fibers. Thus, the objective of this work was to perform a thermo gravimetric analysis of epoxy matrix composites incorporated with different amounts of continuous jute fibers.

2 EXPERIMENTAL PROCEDURE

The jute fibers used in this work were supplied as a 5 kg lot by the Brazilian firm Sisalsul. Figure 1 illustrated a bundle from the as-received lot of jute fibers as well as isolated fibers extracted from the bundle. The dimensional characteristics of the fibers were statistically analyzed from 100 randomly picked from the lot. An average diameter of 0,07 mm and mean length of 300 mm were found and reported elsewhere.[19] The polymer employed as the composite matrix was a type diglycidyl ether of the bisphenol – A (Degeba) with DER 374 g/mol and equivalent weight of 187.3 g/equiv. mixed with an stoichiometric amount, phr = 13, of triethylene tetramine (TETA) as hardener.



(a) (b)

Figure 1. (a) A bundle of as-received; and (b) individually separated jute fibers.

The as-received jute fibers were cleaned with water and then dried in a stove at 60°C for 24 hours. For composite fabrication, the fibers were initially accommodated inside

ISSN 1516-392X

a cylindrical mold with 5.5 mm in diameter and 20 mm in length. Different amounts of continuous and aligned jute fiber were used for composites with 0 vol%, 10 vol%, 20 vol% and 30 vol% of fibers. The still fluid DGEBA/TETA epoxy resin was then poured into the mold and the composites allowed to cure for at room temperature, about 25°C, for one day. Each distinct composite was cut in thin discs with 2 mg of weight, corresponding to about 1 mm in thickness. These composite discs, associated with the different volume fraction of jute fiber embedded in epoxy, were thermo gravimetric analyzed to obtain TG/DTG curves varying with temperature. The analysis was conducted in a Perkin-Elmer equipment at a heating rate of 10°C/min under nitrogen, from 20°C to 800°C.

3 RESULTS AND DISCUSSION

Thermogravimetric results of the neat epoxy and the jute fibers reinforced composites are presented and discussed. Figure 2 shows the TG/DTG curves for the neat DGEBA/TETA epoxy obtained after room temperature curing of one day. In this figure, one should notice that the TG curve displays a slightly decreasing, less than 1% of weight loss, first stage up to 300°C. This is followed by a second stage up to about 550°C in association with a greater weight loss, above 90% of the total sample's weight.

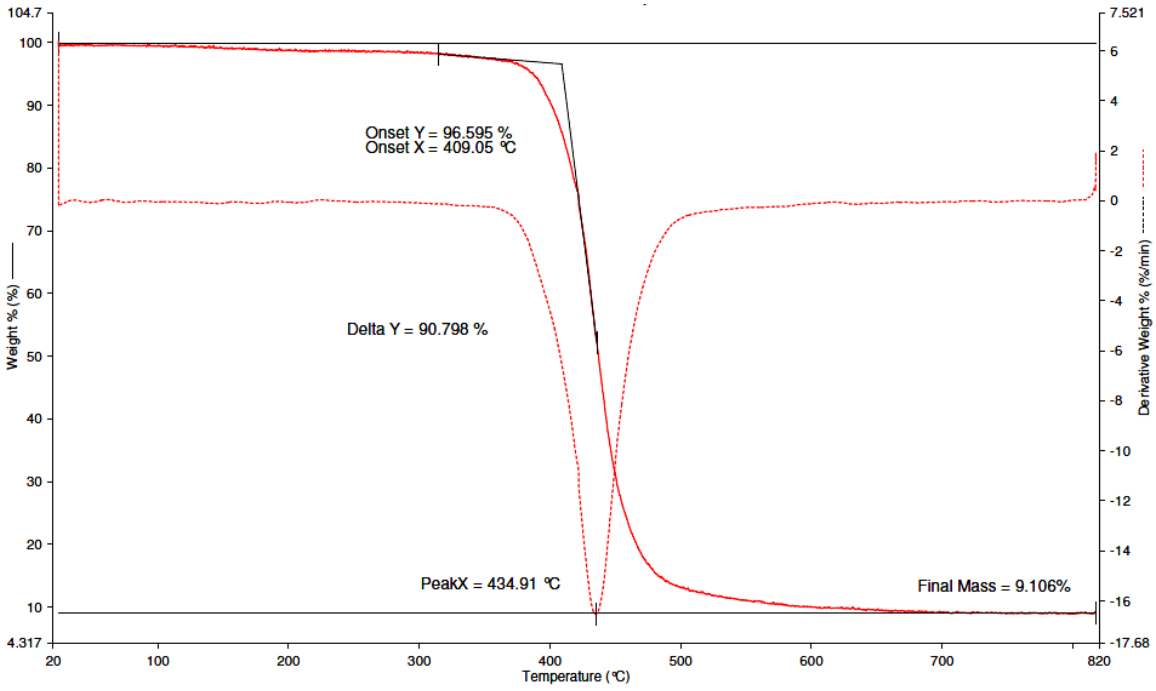


Figure 2. TG/DTG curves of the neat DGEBA/TETA epoxy.

At even higher temperatures, from 550°C to 800°C, a third stage, very slowly declining with weight loss corresponding to 9.1% of the sample (Figure 2). The DTG curve displays only one symmetrical and uniform peak related to a maximum rate of weight loss at 435°C. Such well defined peak is typical of pure polymeric resins that are also associated with a relatively small amount of high temperature residues.[20] In neat polymers, this unique DTG peak is related to the main thermal decomposition mechanism of macromolecules chain degradation or depolymerization.[21] The thermogravimetric parameters obtained from the TG/DTG curves in Figure 2 are presented in Table 1.

ISSN 1516-392X

Table 1. Thermogravimetric parameters of the neat DGEBA/TETA epoxy and related composites with up to 30 vol% of jute fibers

Sample	Initial moisture peak (°C)	Onset of the second stage (°C)	Shoulder peak in the second stage (°C)	Main peak in the second stage (°C)	Final residue (%)
Neat DGEBA/TETA epoxy	-	409	-	435	9.1
Epoxy – 10% fiber	86	378	434	450	12.2
Epoxy – 20% fiber	83	375	420	458	13.6
Epoxy - 30% fiber	80	381	408	459	14.3

Figures 3 to 5 show TG/DTG curves for the DGEBA/TETA epoxy matrix composites reinforced with 10 vol%, 20 vol% and 30 vol% of jute fibers, respectively. The main thermogravimetric parameters obtained from these curves are also listed in Table 1. The results presented in Figures 3 to 5, jointly with the values in Table 1, reveal sensible differences with respect to corresponding results in Figure 2 for the DGEBA/TETA epoxy, which serves as composite matrix. Different than the pure epoxy, the first stage in the composites, up to 300°C, exhibits small peaks between 83°C and 86°C, in association with an appreciable weight loss increasing from 2.1% to 2.9% with the amount of jute fiber. These initial peaks at lower temperatures are characteristics of natural fiber reinforced polymer composites and attributed to the release of water adsorbed on the surface of all lignocellulosic fibers.[22-25] Although small, these initial peaks could be considered as a first limit for the thermal stability of lignocellulosic fiber reinforced polymer composites, particularly those of jute fiber in epoxy matrix.

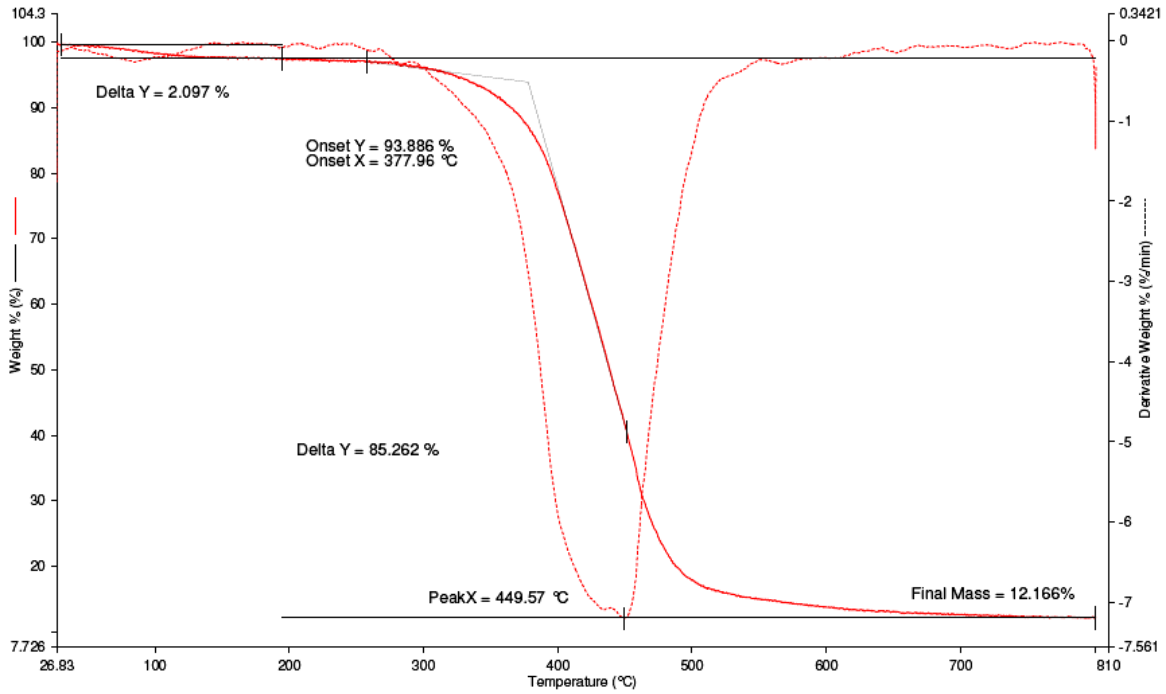


Figure 3. TG/DTG curves for DGEBA/TETA epoxy composites reinforced with 10 vol% of jute fibers.

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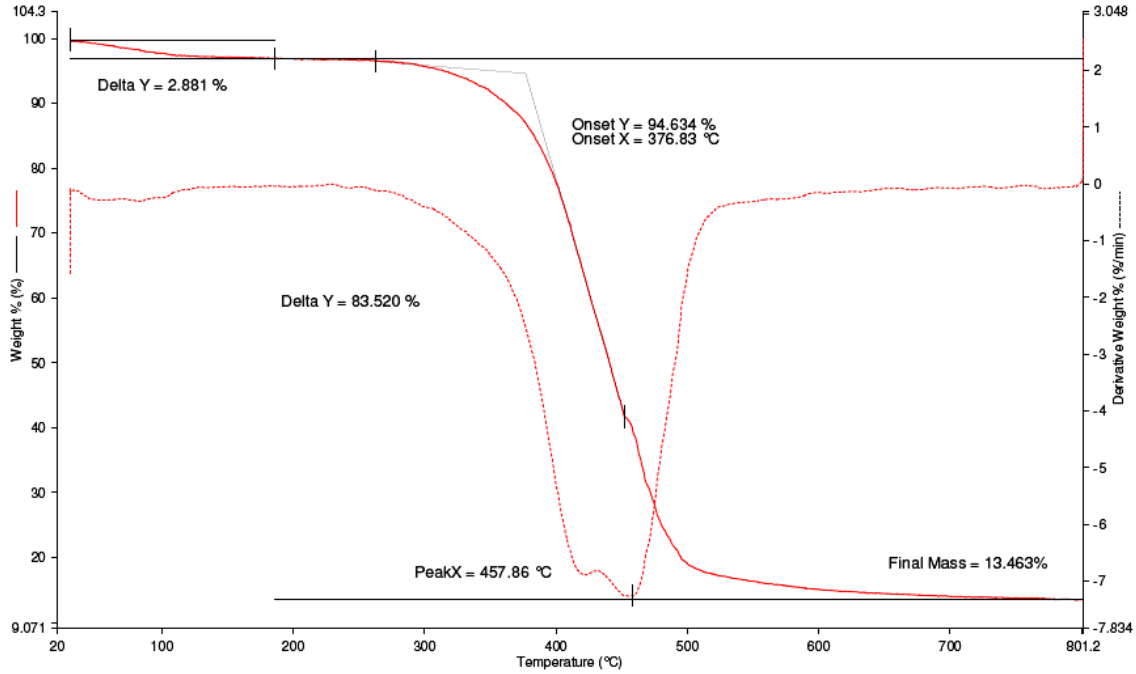

Figure 4. TG/DTG curves for DGEBA/TETA epoxy composites reinforced with 20 vol% of jute fibers.

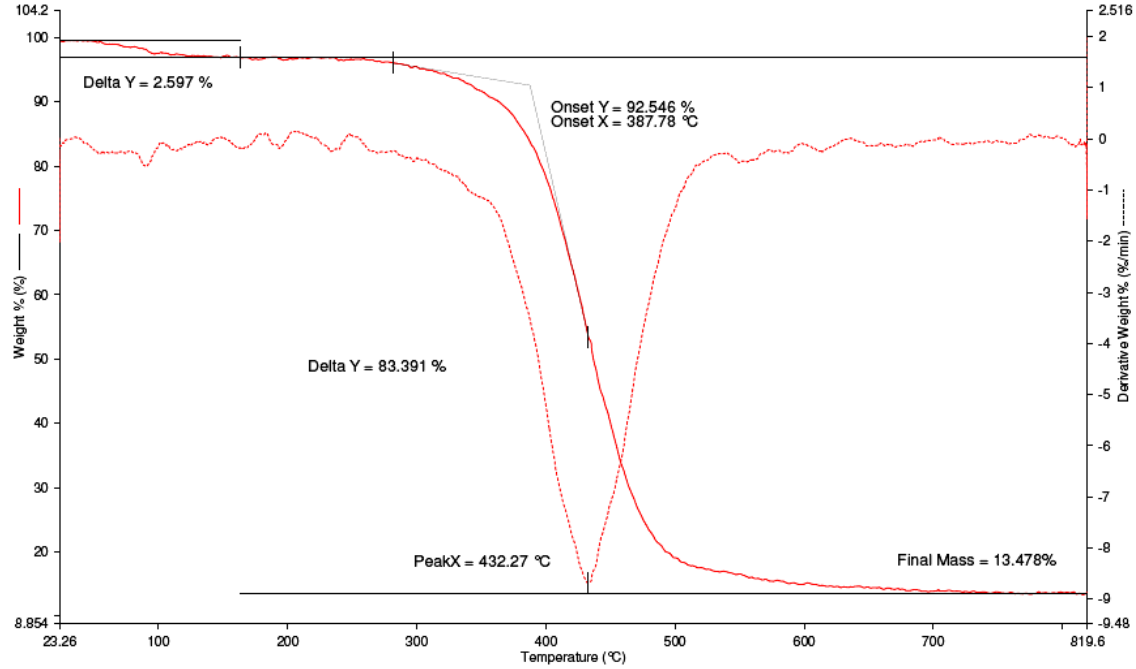

Figure 5. TG/DTG curves for DGEBA/TETA epoxy composites reinforced with 30 vol% of jute fibers.

Another relevant difference between the neat epoxy and the composites refers to the onset of the second TG stage associated with the greater weight loss, up to around 500°C. As presented in Table 1, this onset for the neat epoxy occurs at 409°C, which is a temperature sensibly above those, 375°C-381°C, for the composites. Similar behavior was reported by Mohanty, Verna e Nayak[17] for polyethylene matrix composites reinforced with jute fibers. The reason for this reduction in the second stage onset temperature, which is technically accepted as the composite limit of stability, is ascribed to the process of jute fiber thermal decomposition. In fact, as any lignocellulosic fiber, the jute fiber begins its thermal decomposition by the lignin at temperatures as low as 220°C.[26] At approximately 300°C-400°C, this decomposition

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is maximized. It is then suggested that the jute fiber lignin degradation be the responsible for the composite thermal stability limit. In other words, the stability limit of jute fiber reinforced DGEBA/TETA epoxy composites should be given by the onset temperature of the second TG stage as listed in Table 1.

In particular, it is worth mentioning that a work of Doan, Brodowsky e Mäder[18] on thermal analysis of isolated jute fibers revealed initial peaks of water release around 30°C. Furthermore, in the isolated jute fiber the onset of thermal decomposition occurs at about 290°C owing to the degradation of lignin and hemicelluloses. In addition the authors[18] reported main peaks around 350°C that were assigned to cellulose degradation. These previous results showed that the isolated jute fiber is less thermally resistant than both the DGEBA/TETA epoxy and related composites given in Table 1.

Another important thermogravimetric aspect of the composites shown in Figures 3 to 5 worth discussing is the existence of shoulder peaks in the DTG curves. These shoulder peaks are also observed in other lignocellulosic fiber composites[24,27] and attributed to the degradation of the fiber constituents specially the hemicelluloses and cellulose.[26] A main DTG peak is observed not only for the neat epoxy, at 435°C in Figure 2, but also at higher temperatures 450°C-459°C for the composites in Figure 3 to 5. This main peak associated with the epoxy decomposition, is apparently affected by the presence of jute fibers. It is proposed that the fiber cellulose macromolecules interact with those of the epoxy and increase the thermal stability of the composite. This behavior has not yet been reported in the literature and deserves to be further investigated.

As a last point to be discussed, the introduction of jute fibers, as presented in Table 1, increased the amount of high temperature residue from 9.1%, in the neat epoxy (Figure 2), to 12.2%-14.3% in the composites (Figures 3 to 5). This can be ascribed to the participation of carbonaceous matter and tar/char, predominantly formed in the pyrolysis process of the jute fiber.[26] This also results in lesser weight loss in the second thermo gravimetric stage of the composites (Figures 3 to 5).

4 CONCLUSIONS

- Composites with stoichiometric DGEBA/TETA epoxy matrix reinforced with up to 30 vol% of jute fibers display sensible changes in the thermal behavior evaluated by thermo gravimetric, TG/DTG, analysis;
- the incorporation of jute fibers causes a decrease in the composite onset temperature, which characterizes the beginning of a second stage of greater weight loss, as compared to the neat epoxy, owing to the degradation of the jute fiber;
- in the composites, but not in the neat epoxy, initial DTG peaks were observed around 80°C-86°C due to water release form the jute fibers;
- the main DTG peak around 435°C for the neat epoxy, which can be assigned to the degradation of the macromolecular chain, is slightly displaced to higher temperatures, 450°C-459°C as a consequence of jute fiber/epoxy matrix interaction;
- shoulder peaks, observed only in the composites, were attributed to decomposition of hemicellulose and cellulose contained in the jute fibers;
- the final thermal degradation residue increases in proportion with the amount of jute fiber due to the greater formation of carbonaceous matter and tar/char.

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