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## Physical and dynamic mechanical properties of continuous bamboo reinforcement/bio-based epoxy composites

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Tutea Richmond, Louise Lods, Jany Dandurand, Eric Dantras, Colette Lacabanne, et al.. Physical and dynamic mechanical properties of continuous bamboo reinforcement/bio-based epoxy composites. Materials Research Express, 2022, 9 (1), pp.015505. 10.1088/2053-1591/ac4c1a . hal-03589282

**HAL Id: hal-03589282**

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Submitted on 25 Feb 2022

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Richmond, Tutea<sup>ORCID</sup> and Lods, Louise<sup>ORCID</sup> and Dandurand, Jany<sup>ORCID</sup> and Dantras, Eric<sup>ORCID</sup> and Lacabanne, Colette<sup>ORCID</sup> and Malburet, Samuel and Graillot, Alain and Durand, Jean-Michel and Sherwood, Edouard and Ponteins, Philippe  
*Physical and dynamic mechanical properties of continuous bamboo reinforcement/bio-based epoxy composites.* (2022) Materials Research Express, 9 (1). 015505. ISSN 2053-1591

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# Materials Research Express



## PAPER

# Physical and dynamic mechanical properties of continuous bamboo reinforcement/bio-based epoxy composites

### OPEN ACCESS

#### RECEIVED

7 October 2021

#### REVISED

13 January 2022

#### ACCEPTED FOR PUBLICATION

17 January 2022


#### PUBLISHED

28 January 2022

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**Keywords:** bamboo strips, bamboo fibres, bio-based composites, cardanol-based epoxy, Dynamic Mechanical Analysis, Differential Scanning Calorimetry, mechanical properties

## Abstract

Unidirectional bamboo reinforced cardanol-based epoxy composites were prepared by a close mould method. Two morphologies of reinforcements were used in this research: bamboo fibres and bamboo strips. The present article investigates the influence of bamboo reinforcements on the thermal and mechanical properties of the bio based matrix. Differential Scanning Calorimetry analyses showed that the introduction of bamboo does not modify the physical properties of the matrix. DMA analyses in shear mode showed an improvement of the shear conservative modulus that reaches  $1.7 \pm 0.1$  GPa. This value that is independent from the morphology of reinforcements, indicates the existence of physical interactions. The continuity of matter between bamboo strips or bamboo fibres and the matrix observed by SEM confirms this result. Nevertheless, in tensile mode, the improvement of the tensile conservative modulus is specific to the used morphology. Indeed, for bamboo strips composites, it is  $7.7 \pm 0.8$  GPa, while for bamboo fibres composites, it reaches  $9.6 \pm 0.8$  GPa. This result is explained by the optimisation of stress transfer thanks to the specific morphology of bamboo fibres. A significant increase is also observed for the rubbery modulus due to entanglements specific of bamboo reinforcement.

## 1. Introduction

Transportation mainly contributes to global anthropogenic CO<sub>2</sub> emissions [1]. To control the growth of its carbon footprint, measures such as sustainable fuels or technological improvements are needed [2]. A good way to reduce energy consumption, and thus CO<sub>2</sub> emissions, is to minimise structural mass. Nowadays, airplane and automotive interior designs are mainly made with petroleum-based composites. Recent sustainable goals increased the interest of developing composites using resources from biomass as raw materials. Those lightweight materials are required to be as strong as conventional composites.

Natural fibres have a potential use as reinforcement of composite materials. With a lower density than synthetic fibres, their sustainable characteristic coupling with good specific mechanical properties make them an attractive solution for polymer composites [3–7]. Many studies have already discussed the use, as reinforcement in composite materials, of natural fibres as an alternative to synthetic fibres [8, 9]. Several studies have shown the environmental attractiveness of natural fibres compared to glass fibres due to a lower production impact of such fibres [10, 11]. In the automotive industry, natural fibre composites lead to an environmental impact that is 10%–20% lower than glass fibre composites, partly thanks to the reduction in emissions from fuel consumption due to the lighter weight of natural fibres [12]. Moreover, for the aeronautic domain, the potential applications of this type of composites were widely discussed in the literature [13, 14].

Among the existing natural fibres, reinforcements extracted from bamboo are good candidates to replace man-made fibres like glass fibres [15]. Their worldwide availability makes them environmentally interesting since the localisation of the fibre production is an important factor to consider. It is well accepted that the outstanding mechanical performances of bamboo mainly comes from its fibres which are the structural part of the plant [16]. Accordingly, in the past, they were designated as ‘natural glass fibres’ [17]. In the literature, many authors have studied bamboo fibres for their mechanical performances [16, 18]. Like any natural composite material, natural fibres exhibit considerable variability in their mechanical properties depending on the degree of organisation (bulk, technical fibres, elementary fibres...), extraction method, species, biochemical composition, and the microfibrillar angle [7, 15, 19, 20]. Since the mechanical performances of composites are governed by the reinforcements, it is important to consider the reproducibility of fibre properties.

Numerous studies on short bamboo fibre reinforced composites with thermoplastic or thermoset matrices are reported in the literature [21–25]. To our knowledge, only a few works were carried out with continuous unidirectional bamboo fibres [26–28]. The extraction of bamboo fibres has been extensively discussed in the literature: the conclusion is that it is difficult to obtain long and defect-free fibres [7, 15, 29, 30]. In this work, bamboo fibres were extracted based on a hybrid mechanical-chemical method using sodium hydroxide (NaOH). The alkali solution allows the extraction of technical fibres due to the solubilization of hemicelluloses and lignin [31, 32]. Moreover, this treatment increases the accessibility of the free hydroxyl groups on the fibre surface, allowing physical interactions with the polymeric matrix. It also produces a rougher surface topography which favours interactions with the matrix [31, 33]. In fact, this situation optimizes stress transfer at the fibres-matrix interfaces.

Natural fibres are mostly hydrophilic; accordingly, the choice of the matrix is a key parameter to limit the water absorption of the composites. Thermoplastics have specific advantages such as recyclability. The restriction comes from the processing temperature, which is limited by the thermal stability of natural fibres. It has been shown that the temperature should be below 200 °C to avoid any thermal degradation of the natural reinforcements [34]. Such limitations favour thermosets. With a target of sustainable composites, it is thus relevant to use a thermoset that is derived from renewable resources.

Among thermosets, epoxies are the most widely used. Thanks to their versatility, they are used in many industrial sectors such as aerospace, automotive and marine. Indeed, the properties of the final materials can be tailored for a specific application by optimising the chemical structures of epoxides or hardeners, their functionalities or the epoxide/hardener ratio [35]. Natural epoxidized oils currently constitute the largest, renewable, low-cost and non-toxic alternative family to traditional epoxy [36]. Based on long and flexible carbon chains, such materials have low thermo-mechanical properties. Cardanol is a natural alkylphenolic commercially available. This compound is synthesized from cashew nutshell liquid (CSNL) which is an annually renewable bio sourced material [37, 38]. Although, Cardanol bears an aliphatic chain, the aromatic ring provides a strong chemical and thermal resistant structure which makes Cardanol an interesting starting material for technical applications [38].

This manuscript presents the thermo-mechanical properties of bamboo reinforced epoxy composites. In order to have a sustainable material, a bio-based epoxy matrix was used. This work concerns continuous unidirectional bamboo reinforcement [19]. Two kinds of reinforcement were studied: bamboo fibres (BF) and bamboo strips (BS). Bamboo strips were characterized in a previous study [34]. Accordingly, the best properties are obtained from strips extracted at the periphery of bamboo culm while the location of strips along the culm length has no influence. The thermal stability of the composites was checked by ThermoGravimetric Analysis (TGA) to define the processing conditions. A complementary study of thermal transitions was performed by Differential Scanning Calorimetry (DSC). The investigation of mechanical performances was carried out by Dynamic Mechanical Analysis (DMA) in both shear and elongation modes.

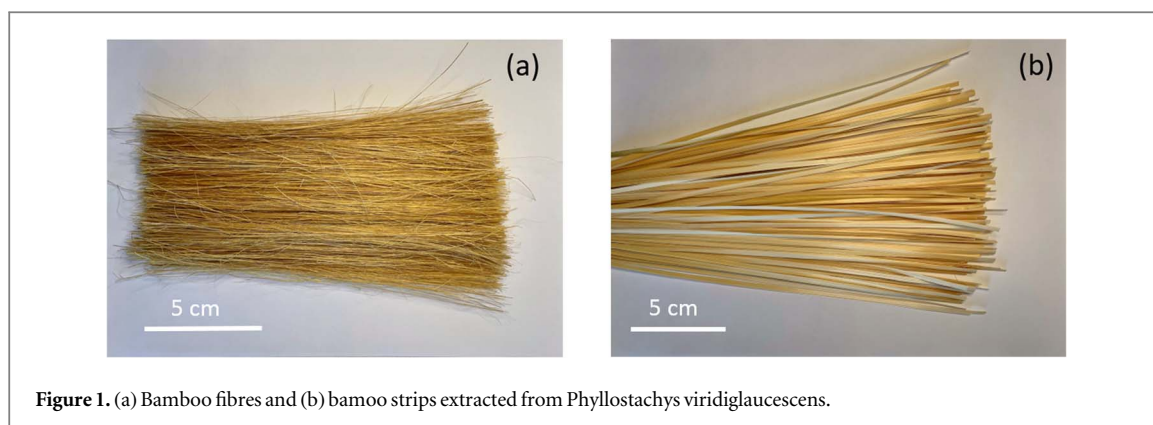
## 2. Materials and methods

### 2.1. Materials

#### 2.1.1. Bamboo reinforcements

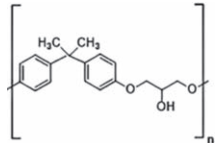

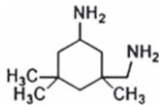
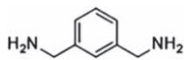
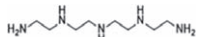
For this study, *Phyllostachys viridiglaucens*, a locally growing bamboo, was provided by Cobratex/France. The two types of bamboo reinforcements used - bamboo fibres and bamboo strips - are shown in figures 1 (a) and (b). **Bamboo strips** (BS) were extracted by a purely mechanical process by Cobratex. With dimensions of 0.3 mm thick and 5 mm width, those reinforcements show a Young’s modulus of 10 GPa and a tensile strength of 210 MPa [34].

Technical fibres, also called fibres bundles, referred as **bamboo fibres** (BF) herein, were extracted by a hybrid mechanical-chemical process with a chemical treatment by an alkaline solution at 1% NaOH described in our previous work [39]. It is important to recall that such treatment does not modify the cellulose component as



**Figure 1.** (a) Bamboo fibres and (b) bamboo strips extracted from *Phyllostachys viridiglaucensens*.

**Table 1.** Chemical composition and structures of FormuLITE 2501A and FormuLITE 2501B.

	CAS	CAS Name	Chemical structure	Mass content
Epoxide resin	250-68-38-6	Liquid epoxy resin		80%–90%
	68413-24-1	Cashew, nutshell liq., polymer with epichlorohydrin		10%–12%
Hardener	2855-13-2	3-aminomethyl-3,5,5-trimethylcyclohexylamine		25%–55%
	1477-55-0	m-Phenylenebis(methylamine)		5%–12%
	112-57-2	3,6,9-Triazaundecamethylenediamine		1–2,5%

shown by XRD analyses reported in the literature [33]. The mean diameter of the extracted fibres is 397  $\mu\text{m}$  with a Young's modulus of 26 GPa and an average tensile strength of 323 MPa [39].

### 2.1.2. Bio-based epoxy matrix

The epoxy system used as matrix for this study was supplied by SPECIFIC POLYMER. It's a bio-based matrix with an epoxide resin (Cardolite FormuLITE 2501A) and an amine-based hardener (Cardolite FormuLITE 2501B). All components were extracted from renewable resources that include Cardanol so that the system has 34% of bio-content. Based on the information on the ingredients provided in the safety data sheet, the chemical composition of the epoxy system is listed in table 1 [40, 41].

### 2.1.3. Composites processing

Before composites processing, bamboo reinforcements were dried in an oven at 100 °C. The bamboo percentage for all composites is 40 weight %. The bio-based epoxy system was prepared according to a mixing ratio in weight of 100:31, specified in the technical datasheet. The mixing was then degassed in an oven under vacuum to eliminate bubbles. Unidirectional composites were then prepared by first aligning bamboo reinforcements in the cavity of a mould and adding the epoxy system to cover all reinforcement. The mould was then closed and transferred in an oven. The curing process undergoes two steps: a first isotherm at 100 °C for 2h then an isotherm at 130 °C for 1h.

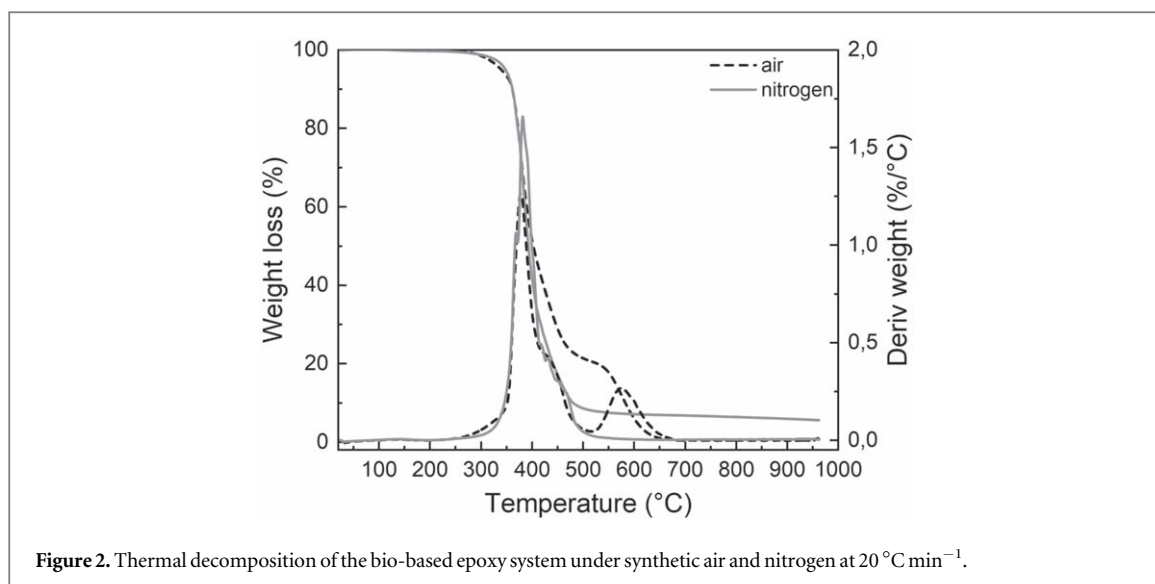


Figure 2. Thermal decomposition of the bio-based epoxy system under synthetic air and nitrogen at  $20\text{ }^{\circ}\text{C min}^{-1}$ .

## 2.2. Methods

### 2.2.1. Scanning electron microscopy

To check the interface between bamboo reinforcements and epoxy matrix, Scanning Electron Microscopy observations were carried out on a JEOL JSM 6700F with an accelerating voltage of 5 kV.

### 2.2.2. Thermogravimetric analysis

In order to evaluate the thermal stability of epoxy, samples were investigated by ThermoGravimetric Analysis (TGA) on a Q50 analyser from TA Instruments. Samples were placed in a closed pan with an initial sample amount of 10 mg. Analyses were done under an oxidizing (synthetic air) and inert gas atmosphere (N<sub>2</sub>) at a heating rate of  $20\text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$  from 30 to  $1000\text{ }^{\circ}\text{C}$ . Three replicates of each TGA thermogram were recorded in order to ensure repeatability. The thermograms obtained on neat epoxy are presented in figure 2.

The thermal decomposition of the biobased epoxy started at  $250\text{ }^{\circ}\text{C}$  based on the derivative weight. For both atmospheres, an intense degradation phenomenon is observed with a maximum intensity at around  $380\text{ }^{\circ}\text{C}$ . Müller *et al* [42] studied the decomposition of an epoxy DGEBA/IPDA system by coupling TGA with Fourier transform infrared Spectroscopy. They found that two competing decomposition pathways occurred depending on the heating rate applied. The first one corresponds to a preliminary dehydration and direct radical scission. With a faster heating rate, a one-step decomposition via direct radical scission is predominant. Based on this observation; we suggest that a balanced ratio of both decomposition pathways is occurring. The total residue under nitrogen atmosphere is 5.6%. This residual mass at this temperature is due to the formation of char as the aromatic cycles are known to promote charring. Under oxidising conditions, a second degradation phenomenon is present leading to a complete decomposition of the sample, which results from oxidation of the first degradation residue.

### 2.2.3. Differential scanning calorimetry

The glass transition ( $T_g$ ) of neat epoxy and composites was determined on a DSC7 Differential Scanning Calorimeter (DSC) manufactured by Perkin Elmer. Samples were sealed in closed aluminium pans for a total of analysed mass between 10 to 15 mg. Experiments consist of two heating runs and two cooling runs under nitrogen flow. For each sample, the  $T_g$  value was measured by the tangent method on the second ramp temperature. The first heating run aimed to erase the hygrothermal history of the sample. Measurements were performed between  $50$  to  $150\text{ }^{\circ}\text{C}$  at a constant heating rate of  $20\text{ }^{\circ}\text{C min}^{-1}$  under a nitrogen flow.

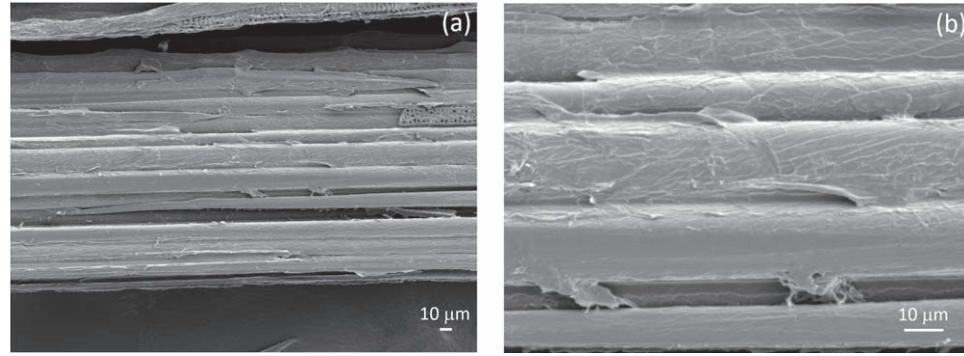
### 2.2.4. Dynamic mechanical analysis

DMA was used to study and characterise the thermomechanical properties of materials. Moreover, as the interfacial bonding in composites is a key parameter for performances, DMA can serve as a technique to identify the quality of the reinforcement/matrix interface. This technique allows us to measure the complex mechanical modulus [43, 44]. In the isofrequency mode ( $\omega_0$ ), the temperature dependence of the modulus is given by:

$$M_{\omega_0}^*(T) = M_{\omega_0}'(T) + iM_{\omega_0}''(T)$$

with  $M_{\omega_0}'(T)$  and  $M_{\omega_0}''(T)$  are respectively the storage modulus and the loss modulus. The analytical representation of the dynamic mechanical behaviour is based on the Maxwell model and is defined by the





**Figure 3.** SEM images of bamboo (a) fibre bundles after a hybrid-mechanical extraction process (b) fibre bundles composed of several elementary fibres. The roughness of the surface is evident.

relationships:

$$M'_{\omega_0}(T) = M_r + (M_g - M_r) \frac{\omega_0^2 \tau (T)^2}{1 + \omega_0^2 \tau (T)^2}$$

$$M''_{\omega_0}(T) = (M_g - M_r) \frac{\omega_0 \tau (T)}{1 + \omega_0^2 \tau (T)^2}$$

where  $M_r$  is the rubbery modulus,  $M_g$  is the glassy modulus and  $\tau$  is the relaxation time. The tensile storage and loss moduli are designated as:  $E'_{\omega_0}$  and  $E''_{\omega_0}$ . The shear storage and loss moduli are called  $G'_{\omega_0}$  and  $G''_{\omega_0}$ . The damping factor  $\tan \delta$  is defined as the ratio of storage modulus to loss modulus,  $\delta$  being the angle between the in-phase and out-of-phase components of the modulus. DMA were performed on the ARES G2 strain-controlled rheometer manufactured by TA Instruments. Rectangular samples dimensions are 50 mm × 10 mm × 0.3 mm for the shear geometry mode and 50 mm × 10 mm × 0.3 mm for the tensile geometry mode. Trials were carried out over the temperature range –130 to 150 °C at a heating rate of 3 °C min<sup>-1</sup>. For each sample, two consecutive runs were performed in the isofrequency mode at  $\omega_0 = 1$  rad s<sup>-1</sup>. This frequency has been selected since it corresponds to the lowest noise/signal ratio. The relative strains are 0.1% for the shear mode and 0.03% for the tensile mode for remaining in the linear viscoelastic region in both cases. All experiments were replicated three times to check repeatability.

For the isotropic epoxy matrix, only the shear mode was used while for the anisotropic bamboo/epoxy composites, both shear and tensile modes were investigated.

### 3. Results and discussion

#### 3.1. Morphology analysis

SEM images of alkali extracted bamboo fibres surfaces are shown in figures 3(a) and (b). They show that fibres are not damaged by the extraction process. Two effects of the treatment can be observed: the fibres surface was cleaned making it easier to identify single bamboo fibres. Moreover, a rough surface may be beneficial to improve the mechanical interaction with the matrix.

Unidirectional bamboo composites are presented on figures 4(a) and (b). It is interesting to note there are no voids or cavities are on the surface area.

Composites samples were then fractured under liquid nitrogen condition and observed by SEM in order to check reinforcement/matrix interactions. Fractured surface are presented in figure 5(a) and 5(b). The interface matrix/filler is illustrated by the black dashed line. In these images a continuity of matter indicates a good interface between matrix and both reinforcements.

As no coupling agent was used this work, it means that the hydrogen bonding inherent to bio-based epoxy promotes good interfacial interactions with bamboo reinforcements.

#### 3.2. Thermal transitions of bamboo/epoxy composites

The thermal transitions of the epoxy matrix (E) bamboo strips/epoxy (BS/E) and bamboo fibres/epoxy (BF/E) composite were investigated by DSC. The DSC thermograms are presented in figure 6.

For both epoxy matrix and BF/E composite, no residual exothermicity was detected. This result indicates the complete curing with a fully cross-linked resin. On the first run, a small endothermic event is present due to water/sorption onto the sample. On the second run the event detected is an endothermic second-order

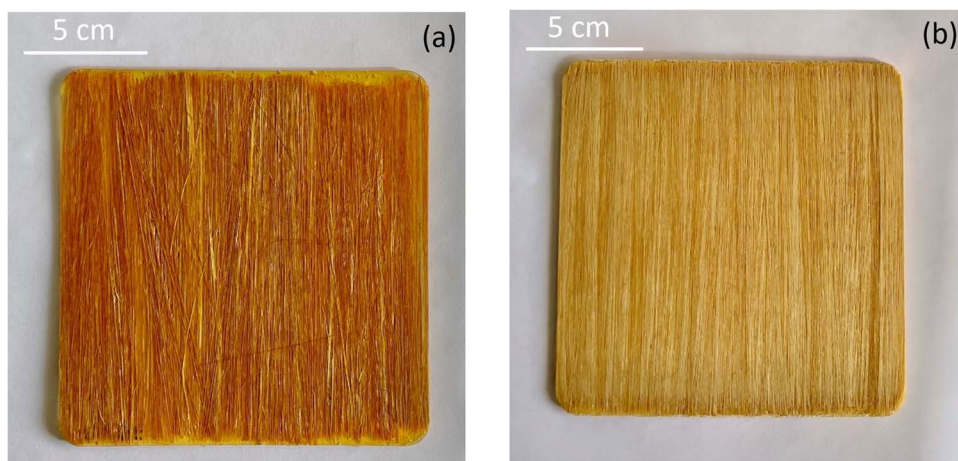


Figure 4. Images of bio-based epoxy composites reinforced with (a) bamboo fibres and (b) bamboo strips.

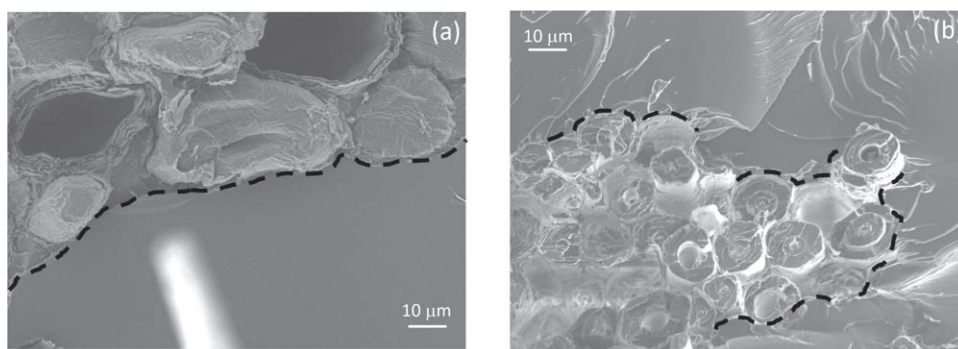


Figure 5. SEM observations of the interface between epoxy and (a) bamboo fibers and (b) bamboo strips. Dashed lines are guidelines for interface.

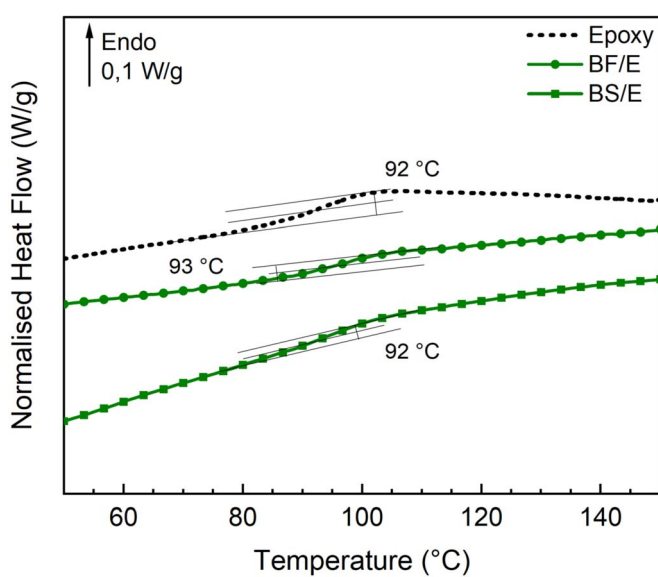
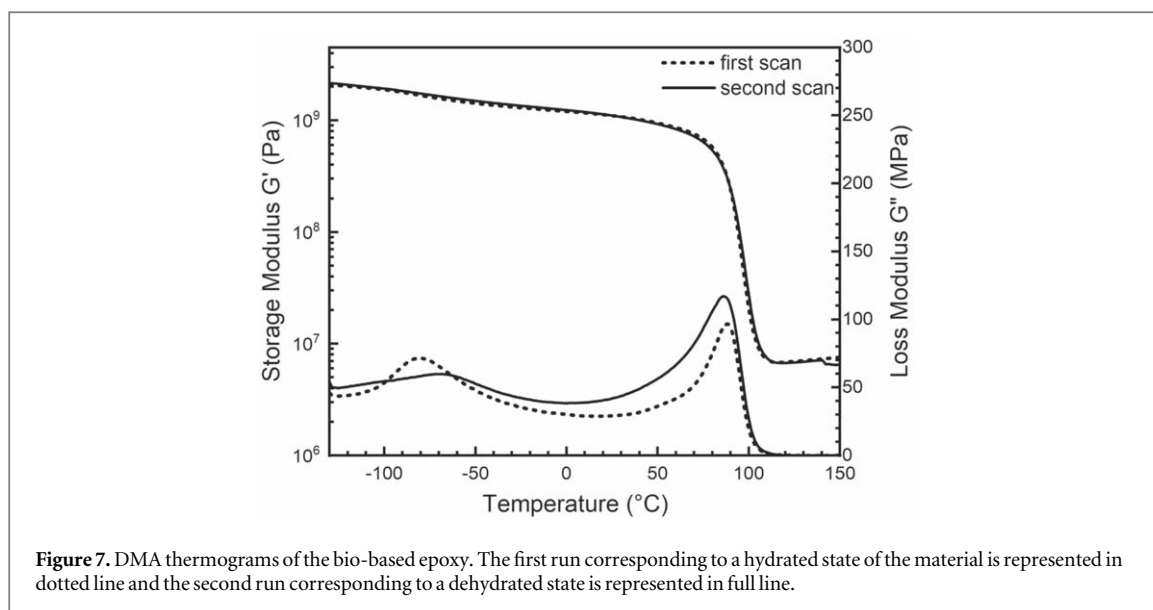


Figure 6. DSC curves of the bio-based epoxy and bamboo/epoxy composites.





**Figure 7.** DMA thermograms of the bio-based epoxy. The first run corresponding to a hydrated state of the material is represented in dotted line and the second run corresponding to a dehydrated state is represented in full line.

transition characteristic of the glass transition of epoxy. The  $T_g$  of pristine epoxy  $92^\circ\text{C}$ , agrees with the value reported by Mu *et al* [45]. This value is of  $93^\circ\text{C}$  and  $92^\circ\text{C}$  for bamboo fibres/epoxy composites and bamboo strips/epoxy respectively. The slight variation between matrix and composites shows that the incorporation of bamboo reinforcements doesn't modify the formation of epoxy network. This result is explained by the fact that the chemical treatment is made with an alkaline solution at 1%NaOH. In previously published data on epoxy/bamboo fibers, it was reported that the increase of the glass transition temperature only happens for treatments with higher NaOH concentrations [46]. In the present work, there is only physical interactions between matrix and fibers so that such composites are reusable.

### 3.3. Thermo mechanical analysis of the epoxy matrix

The thermomechanical properties of the bio-based epoxy were assessed by DMA in the shear mode. Experiments were done in the thermal stability range of the matrix previously defined by TGA, e.g.  $-130$  to  $150^\circ\text{C}$ . Thermograms are shown in figure 7.

Two consecutive runs were recorded: the first one corresponds to a hydrated state of the matrix and the second one to its dehydrated state.

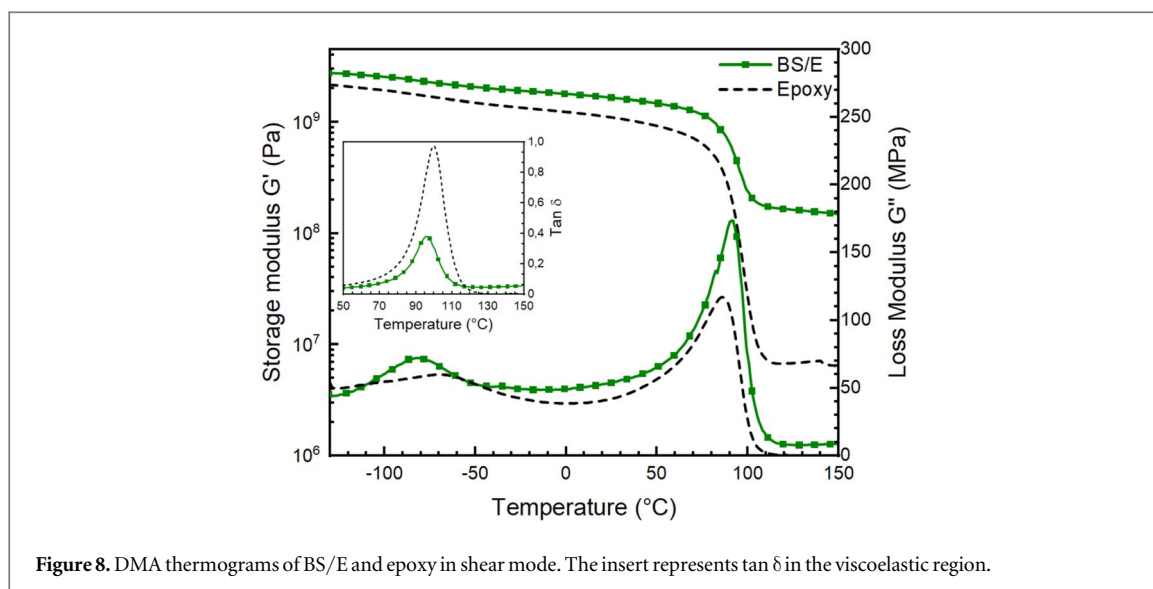
Loss modulus  $G''$  thermograms of the pristine samples show two distinct relaxation modes:

- At higher temperature, a primary relaxation, designated as the  $\alpha$  mode.
- At lower temperature a secondary relaxation, designated as the  $\beta$  mode.

The  $\alpha$  relaxation is the anelastic manifestation of the glass transition of the epoxy matrix. The  $T_{\alpha}$  temperature of the  $\alpha$  mode is defined as the maximum of the loss modulus peak. The bio-based matrix displays a  $T_{\alpha}$  of  $89^\circ\text{C}$  and  $87^\circ\text{C}$  respectively for the first and the second run.

The  $T_{\alpha}$  values are consistent with the  $T_g$  glass transition temperature previously determined by DSC at  $92^\circ\text{C}$  for the second run. Accordingly, the  $\alpha$  mode may be attributed to the anelastic manifestation of the glass transition of the epoxy matrix.

At lower temperature, the  $\beta$  relaxation is observed as a broad peak located between  $-130^\circ\text{C}$  to  $-50^\circ\text{C}$ . This secondary relaxation is associated with a localised molecular mobility of short molecular sequences of the polymeric network. This mode was observed in petroleum-based epoxy and its origin has been largely discussed. By studying the dynamic properties of epoxy-amine networks, several authors have concluded that this relaxation is related to the sum of the localised mobility of glyceryl units and the diphenylpropane units ( $\text{CHOH}-\text{CH}_2-\text{O}$ ) [47, 48]. Figure 7 shows that the  $\beta$  relaxation is more intense for the matrix in the hydrated state. This observation suggests interactions of polar groups with water molecules which significantly increased the magnitude of the relaxation. On the second scan, this relaxation is shifted to higher temperatures with a smaller amplitude confirming a dependence towards hydration [49]. It is interesting to mention that the existence of a double relaxation visible at low frequencies, related to IPDA cycle has been previously mentioned in the literature [50].



**Figure 8.** DMA thermograms of BS/E and epoxy in shear mode. The insert represents  $\tan \delta$  in the viscoelastic region.

Below  $T_{\alpha}$ , in the vitreous state, the  $G'$  storage modulus is on the glassy plateau; it represents the elastic energy stored in the material. For the bio-based matrix, the storage modulus at room temperature is  $G_g' = 1.1 \pm 0.1$  GPa for both runs. Above  $T_{\alpha}$ , in the rubbery state, the rubbery modulus  $G_r'$  is stable ( $7.1 \pm 0.6$  MPa): there is no evolution of the network confirming that the material is totally cured.

### 3.4. Thermomechanical analysis of bamboo strips/epoxy composites

To evaluate the role of bamboo reinforcements in bio-based epoxy matrix, composites were characterized using the same protocol than for neat epoxy. For this section, thermograms recorded by DMA corresponds to the dehydrated state (second run) to get rid from hydration.

#### 3.4.1. Dynamic mechanical analysis of bamboo strips/epoxy in shear mode

Figure 8 presents the thermograms of the epoxy matrix reinforced with bamboo strips, with as reference, the epoxy matrix.

The storage modulus  $G'$  of BS/E is higher than the one of epoxy matrix in the whole temperature range. The introduction of strips significantly stiffens the matrix. On the vitreous plateau, at room temperature  $G_g' = 1.7 \pm 0.1$  GPa, which is almost 1.5 times the one of the epoxy matrix. This behaviour can be explained by the existence of bamboo/epoxy interactions due to static hydrogen bonds and confirms the observations from SEM images. At higher temperature, in the viscoelastic region, the drop of modulus of the composite is lower than the one of the matrix, indicating that the large scale motions involved in the epoxy are restricted by bamboo strips. Following the viscoelastic transition, the rubbery plateau of BS/E material is  $G_r' = 149.5 \pm 20.1$  MPa i.e., 21 times the rubbery modulus of epoxy. This behaviour can be explained by bamboo/epoxy entanglements.

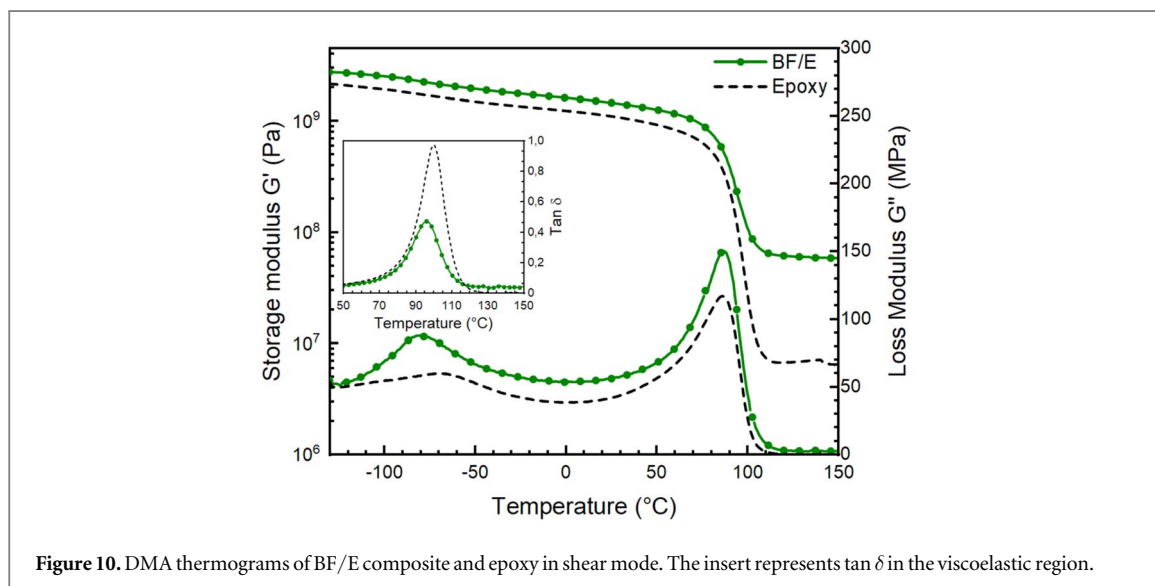
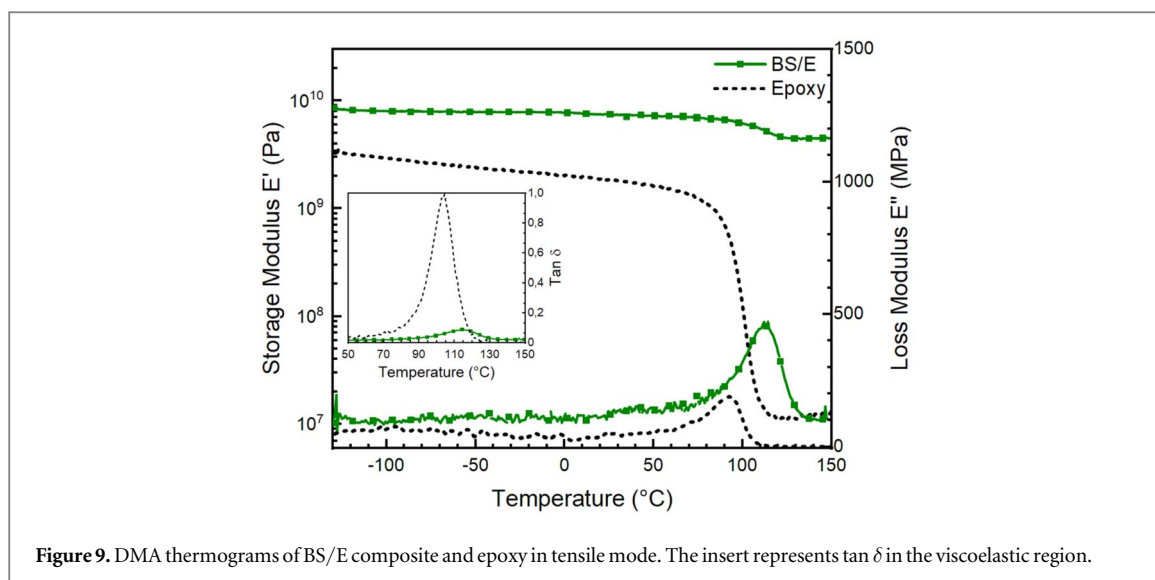
$\tan \delta$  curves for BS/E and epoxy matrix as a function of temperature in the viscoelastic region are shown in the insert of figure 8. The lower damping ratio in composites is linked to the higher rubbery storage modulus.

The  $G''$  loss modulus thermogram of BS/E shows two relaxation modes as for the matrix. This modulus referred to the ability of the material to dissipate energy. Figure 8 shows that the  $\beta$  relaxation of BS/E composite at  $-80^\circ\text{C}$  is broader than the one of the matrix. This dissipative effect might be attributed to interactions between bamboo and epoxy polar groups. It is interesting to note that the  $\alpha$  relaxation peak of BS/E is identical to the one of epoxy indicating that the incorporation of bamboo doesn't modify the network of epoxy.

#### 3.4.2. Dynamic mechanical analysis of bamboo strips/epoxy in tensile mode

Thermograms obtained in tensile mode for BS/E are presented in the figure 9 with, as reference, the one of epoxy matrix. It is important to note that the composites were processed with the same protocol than for the shear mode. In the glassy zone of the matrix, at room temperature, composites show a glassy modulus of  $7.7 \pm 0.8$  GPa. This value is particularly high compared with the glassy modulus of the matrix ( $1.8 \pm 0.1$  GPa). In a previous work, the Young's modulus of the same bamboo strips than the ones used in composites was measured:  $10 \pm 2.4$  GPa [34]. The major point on figure 9 is that, in the elongation mode, the contribution to the Young's modulus of the bamboo reinforcement is predominant.

We observe a slight increase of the  $T_{\alpha}$  of BS/E at  $112^\circ\text{C}$  in comparison with the epoxy one. The introduction of bamboo reinforcements increases the density of the epoxy network. Accordingly, the  $\beta$  relaxation cannot be



identified in this configuration. Due to the increase of the conservative modulus, the  $\beta$  relaxation cannot be observed in this configuration.

The damping factor of BS/E and the pristine epoxy are plotted in function of the temperature in figure 9. It is clear that the incorporation of bamboo strips has an influence on the damping properties of epoxy. The lower damping in BS/E composites is due to the much higher value of its tensile conservative modulus in the whole temperature range.

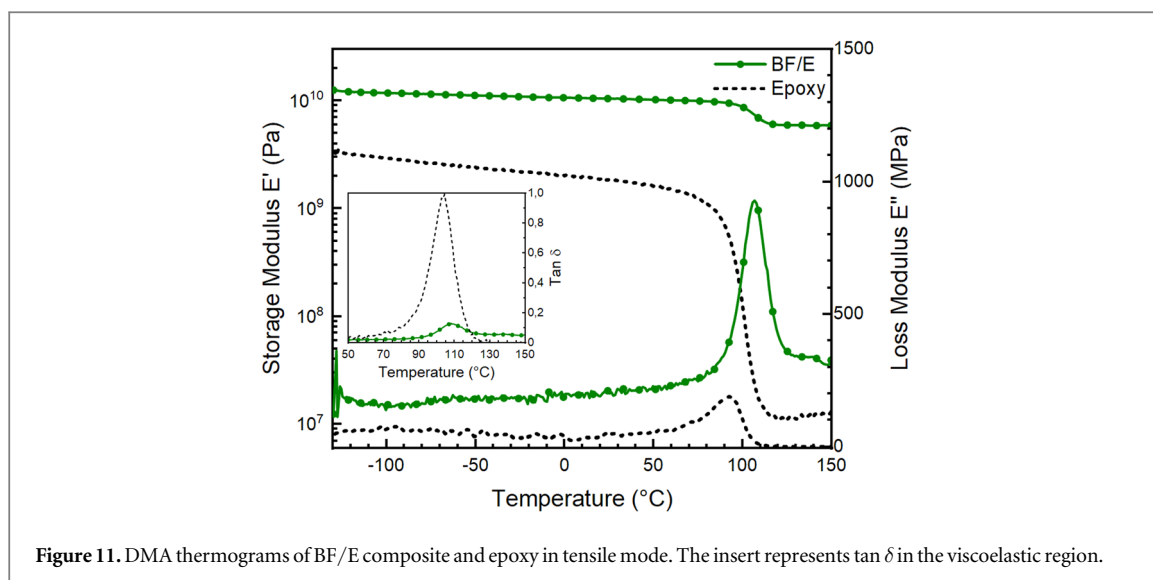
### 3.5. Thermo mechanical analysis of bamboo fibres/epoxy composites

#### 3.5.1. Dynamic mechanical analysis of bamboo fibres/epoxy composites in shear mode

The epoxy matrix was also reinforced with bamboo fibres. The thermograms recorded in the shear mode on BF/E composites are presented in figure 10.

In the whole temperature range, the  $G'$  storage modulus of BF/E composite ( $G_g' = 1.5 \pm 0.1$  GPa) is higher than the one of the matrix ( $G_g' = 1.1 \pm 0.1$  GPa). In the vitreous state of the matrix, the increase is analogous with the one observed in BS/E composite. This effect is independent from the morphology of reinforcement and confirms the good compatibility between bamboo reinforcements and the bio-based matrix as observed by SEM. For the rubbery state of the matrix, the increase of rubbery modulus for BF/E composite is much lower for the fiber reinforcement than for strip reinforcement.

For the  $G''$  loss modulus, as previously observed for BS/E, two relaxations modes are present. The primary relaxation of BF/E located at 86 °C is very close from the one of the matrix. This indicates that, at the scale of macro-Brownian molecular mobility, the polymeric network is not modified by the presence of fibres. The



**Figure 11.** DMA thermograms of BF/E composite and epoxy in tensile mode. The insert represents  $\tan \delta$  in the viscoelastic region.

secondary mode is shifted to lower temperature and more intense than in the case of epoxy matrix. At the scale of the localised molecular mobility, the effect of bamboo/epoxy interaction induces a higher localisation responsible for the shift towards higher frequencies.

The insert of figure 10 represents  $\tan \delta$  versus temperature in the viscoelastic region. Like for BS/E composites, the lower damping in composites is due to the higher value of the storage modulus in the whole temperature range.

### 3.5.2. Dynamic mechanical analysis of bamboo fibres/epoxy composites in tensile mode

As shown by the thermograms of BF/E in figure 11, the incorporation of bamboo fibres into epoxy matrix increases the tensile modulus in the whole temperature range. In the glassy state, the Young modulus at room temperature is  $E_g' = 9.6 \pm 0.7$  GPa. It corresponds to an improvement of the modulus of epoxy by a factor of 5. This increase indicates a good stress transfer between fibres and matrix due to the existence of static hydrogen bonds between hydrophilic fibres and polar groups of the epoxy matrix. Moreover, as for BS/E composites, the contribution to the Young's modulus of bamboo fibres is predominant. It is interesting to compare this value with data obtained in previous work on the bamboo fibres used for composites: We found that the Young's modulus was 25 GPa [39]. The difference between both values might be explained by the poor orientation of fibres due to the processing of composites.

Above the viscoelastic transition temperature, the value of the rubbery modulus of the matrix is  $E_r' = 12.6 \pm 2.5$  MPa and for BF/E is  $E_r' = 5.5 \pm 1.0$  GPa i.e. more than 400 times the value of the matrix at the same temperature. Entanglements of bamboo fibres through the polymeric network might explain such high values. The loss modulus shows a primary relaxation for BF/E composite at 107 °C i.e. a few degrees above the matrix. This slight shift may be attributed to orientation. In such environment, the secondary relaxations are not observed.

The insert in figure 11 represents  $\tan \delta$  versus temperature in the viscoelastic region. The lower damping in BF/E composite is due to the much higher value of its tensile modulus in the whole temperature range.

## 4. Conclusions

In this study, fully bio-based composites were successfully prepared with two different continuous reinforcements at 40 wt%: bamboo strips and bamboo fibres. The thermal and mechanical properties of unidirectional bamboo reinforced bio-based epoxy composites were presented in this work.

As shown by the DSC studies, the glass transition of composites is close from the one obtained on the neat epoxy. Accordingly; there is no chemical interactions between bamboo and epoxy matrix so that the introduction of bamboo does not modify the polymeric network.

The evolution of the mechanical moduli was analysed by DMA. In shear mode, the introduction of bamboo reinforcements increased the conservative moduli by approximately a factor of 1.5 at room temperature. This result suggests the existence of bamboo/epoxy interactions independently from the morphology of the reinforcement. We have assigned this effect to physical interactions as polar bonding. SEM images of the

composites showed good compatibility of the cardanol-based epoxy with both bamboo fibres and bamboo strips. Those observations are consistent with the DMA data.

In tensile mode, the improvement of the Young's modulus is specific to the morphology of the reinforcement. Indeed, for bamboo strips composites, the tensile modulus is  $7.7 \pm 0.8$  GPa while it reaches  $9.6 \pm 0.7$  GPa for bamboo fibres composites. This increase is explained, on one hand, by the higher tensile modulus of fibres and, on the other hand, by the optimisation of stress transfer inherent to fibrous morphology. The increase of modulus upon introduction of bamboo reinforcement is also observed for the rubbery modulus. This evolution reaches 3 decades for the fibre epoxy composites; it is due to specific entanglements for this morphology. Finally, it is important to emphasise that, in the shear mode, the mechanical behaviour of the matrix is optimised upon both bamboo reinforcements.

## Acknowledgments

This work was carried out within the framework of the **BAMboo long fibre reinforced bio-based Matrix COMposites (BAMCO)** program supported by BPI and the Conseil Régional d'Occitanie/France.

## Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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