Exploring dynamic mechanical properties in fibre-reinforced composites using spatial mapping techniques

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Abstract
The damping characteristics of composites containing plant fibres have attracted significant attention, primarily through macroscopic analysis methods. However, due to their intricate and diverse microstructures, exploring their damping properties at a microscopic level becomes imperative for a comprehensive understanding of the underlying dissipation mechanisms. This paper proposes a methodology for characterizing the dynamic mechanical attributes of various composites at the micro-scale, employing Brillouin light spectroscopy a non-invasive and contactless dynamic method. By using this methodology, it is possible to get access to the rigidity and damping of different fibres inside a polymer composite. The main observation made in this paper is that the glass fibre presents a damping varying from 1.9% to 2.4%, compared to the flax fibre which shows higher damping with 5.6 to 7.4 %. In addition, the PA11 fibre highlights he highest damping with 18 to 21.7%.

1. Introduction

Over the years, metallic structures have progressively been replaced by structures made of composite materials, primarily due to their remarkable properties in terms of specific strength and rigidity, fatigue resistance, and corrosion resistance [1] but also to their lower density, which ultimately results in lighter-weight structures. This shift towards lightweight structures holds significant implications for reducing fuel consumption, particularly in critical sectors such as transportation, aerospace, and space industries. This transition is of main importance from economic, social, and environmental perspectives. While these composite materials can compete with metallic counterparts in terms of mechanical strength, their primary drawback lies in their environmental impact. Traditional composites, typically reinforced with glass or carbon fibres, need high energy consumption and subsequent CO_2 emissions for their production, underscoring the need for alternative solutions. These materials are generally not recyclable, leading to unsustainable end-of-life scenarios. In response to these environmental concerns, plant fibres are emerging as viable alternatives due to their abundant availability, renewable nature, biodegradability, low cost, lightweight characteristics and especially their ability to sequester CO_2 during plant growth. Prior research indicates that the damping capacity of tested plant fibre-reinforced composites (PFRC) surpasses those of synthetic fibre-reinforced composites (SFRC) [2,3]. However, the exploitation of this damping capacity remains confined to the domains of sports and leisure, where product and structure design is predominantly empirical. A better understanding and characterisation of the damping sources and the mechanisms underlying this behaviour are necessary to access more technological applications. While composite rigidity typically follows a simple mixture law, the prediction of damping is more complex. Duc et al [3] illustrated this point while studying unidirectional

composites with different matrices, observing that the addition of fibres to the matrix could either increase, decrease or maintain damping levels depending on the matrix type. This complexity cannot be explained and described by a simple mixture law. Plant fibres are inherently more complex than synthetic ones. They present intricate hierarchical microstructures and complex morphologies, along with a sensitivity to moisture [1]. These aspects can directly affect their dynamic behaviour, encompassing nonlinearities, couplings, and moisture-induced effects. This highlights the need to have a multiscale understanding, characterization and modelling of the composite's damping, to better assess and discriminate the energy dissipation resulting from the matrix, the fibre itself, the interfaces (matrix-fibre, fibre-fibre) and even from defects or porosity. To address this need, it is crucial to spatially map the viscoelastic properties at the microscopic scale.

To do so, different methods, sweeping across different frequency ranges, can be used. Grid nanoindentation represents a powerful technique to investigate the properties of heterogeneous materials like composite material samples made of micrometre-sized fibres in a polymeric matrix [4]. This method has been effectively employed to map the static mechanical properties in plant fibre composites [5-7]. Lately, Liu et al. [2] also used grid nanoindentation, but in dynamic mode using a constant amplitude measurement method, to map the viscoelastic properties (storage modulus and loss factor) in plant fibre composites (see Figure 1. a). The proposed method enables the refinement of loss factor determination and minimizes the contribution of plastic deformations commonly induced by the widely used continuous stiffness measurement (CSM) method in dynamic nanoindentation. However, this method is not without its limitations. Concerns arise regarding the influence of edge effects on results, as well as the complex interplay of inelastic and multiaxial contributions, raising questions about the method's scope and accuracy.

The need to get a mapping of the composite's damping without damaging it, getting a directional, non-invasive and contactless method is of main importance. To achieve this goal, optical methods present a promising avenue. Among these, Brillouin light spectroscopy (BLS) emerges as a particularly interesting option. Notably, BLS is non-contact thus non-invasive, offering localized measurements crucial for comprehensive composite analysis.

This non-invasive technique relies on laser spectroscopy coupled with interferometry (see Figure 1. b). This method has been widely developed over the last decades, especially in crystals but recently tends to be more adapted in the biomedical field [8]. So far, it has not been used extensively to determine damping in composite materials. However, recently, Elsayad et al. [9], focused on the fibre scale and identified successfully both rigidity and damping on cellulose fibres.

The present study proposes to map the elastic and damping properties in plant fibre composites using the BLS technique. The paper is structured as follows: an introduction in the first section, a detailed explanation of materials and methods in the second section, covering sample creation to characterization, and finally, a presentation of results using BLS in the third section.

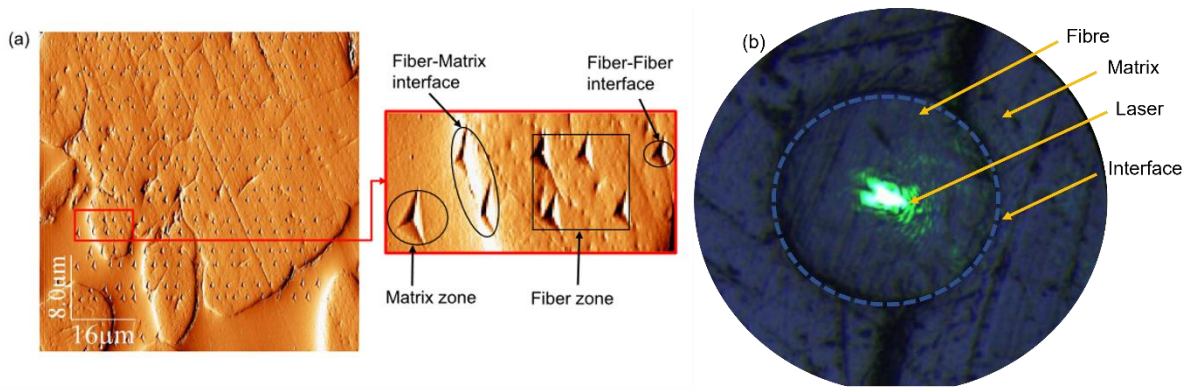

Figure 1: (a) Flax fibre reinforced composite after grid nanoindentation test [2], (b) composite subjected to local measurement via laser spot for BLS test

2. Materials and method

2.1. Materials

The presented work employs four different types of fibres, covering a wide range of fibre categories. Flax represents vegetal fibres, silk stands for animal fibres, polyamide PA11 represents synthetic fibres, and E-glass fibre is used for inorganic fibres. Table 1 presents their mechanical and optical properties. The resin used for the composite manufacturing is an epoxy (SR GREENPOXY 56) from SICOMIN coupled with a hardener (SD 7561) also coming from SICOMIN cured for 48 hours at ambient temperature.

Table 1. E longitudinal modulus, ρ density, n refractive index and ν Poisson ratio of tested fibres and matrix.

Specimen Type	E (GPa)	ρ (kg/m^3)	n [16]	ν
Flax	30 – 110 [15]	1450 – 1560 [15]	1.56 – 1.59	0.25-0.35 [15]
E-glass	76 [1]	2550 [1]	1.51	0.25 [14]
PA11	1 – 1.3[12]	1030 – 1040[12]	1.52	0.3-0.4 [14-19]
Silk	3 – 3.5[18]	1320 – 1400[18]	1.49 – 1.52	0.3-0.5 [13]
Greenpoxy	3 – 3.4[17]	1100 – 1200[17]	1.51	0.33 [14]

2.2. Sample preparation

Mini composites were manufactured by embedding one row of five elementary fibres in a Greenpoxy matrix. The main objective of such a sample is to represent a minimal volume of a composite, to get its fundamental components, i.e. the fibre, the interface and the matrix. One row of each fibre type was placed inside the Greenpoxy. Flax fibres were individually isolated from bundles, while others were extracted from unidirectional fabrics. These fibres were then carefully arranged within a paper frame, awaiting embedding. A Greenpoxy resin (100/34 g) was prepared and poured into a mould. After allowing 48 hours for drying, the fibres were embedded in the matrix. Subsequently, the composite was removed from the mould and subjected to polishing. The protocol uses a polishing machine (Presi Minitech 233) and consists of 5 minutes using polishing disks ranging from P400 to P4000 polishing disk at 100 turn/s without water to avoid flax swelling. Surface quality was monitored using a microscope KEYENCE (VHX-5000) after each disk and the time of polishing can be adjusted according to the observations. This process facilitates accurate analysis of the samples.

2.3. Brillouin light scattering fundamentals

BLS is a spectroscopic technique that is used to study the interactions between light and acoustic waves (phonons) in materials. When light interacts with phonons, it undergoes a change in frequency known as the Brillouin frequency shift (inelastic scattering). By measuring this frequency shift, BLS provides valuable information about the mechanical properties of materials, such as their elastic constants, sound velocities, and damping constants. The output of such a method is a frequency shift related to the velocity of phonons, and the bandwidth of the transmission peak. Figure 2 presents an example of the obtained spectrum in the case of the PA11 fibres, with the central peak corresponding to the laser line broadened by elastic (Rayleigh) scattering. In the case of isotropic material, the determination of the elastic modulus is done via the identification of the C_{11} constant of the 4th-order elasticity tensor. C_{11} is linked to the phase velocity V of the longitudinal waves by the relation in Equation 1.

$$V = \left(\frac{C_{11}}{\rho}\right)^{\frac{-1}{2}}, \quad (1)$$

with ρ the material density. The Brillouin frequency shift w_b is linked to this phase velocity and thus to C_{11}. Its expression is given in Equation 2.

$$w_b = \frac{2nV}{\lambda_0} sin\left(\frac{\theta}{2}\right), \tag{2}$$

with n the refractive index, λ_0 the wavelength of the laser and θ is the angle of scattering. For isotropic material, the relation linking C_{11} and the longitudinal modulus is as follow:

$$C_{11} = E\,(1-\nu)/[(1+\nu)(1-2\nu)]. \tag{3}$$

With the relation in Eq.3, the modulus can be identified directly by knowing the Poisson's ratio (refer to Table 1).
For damping, the loss factor can be identified via the half bandwidth method. A sharper Brillouin's peak typically indicates lower damping in the material, which implies lower energy dissipation and less vibrational attenuation. This relationship is often utilized in identifying the loss factor through the half-bandwidth method. In this method, the width of the Brillouin peak at half of its maximum intensity is measured. Therefore, by analyzing the width of the Brillouin peak, the damping characteristics of the material are identified. The BLS response is fitted against a Lorentzian model [9].

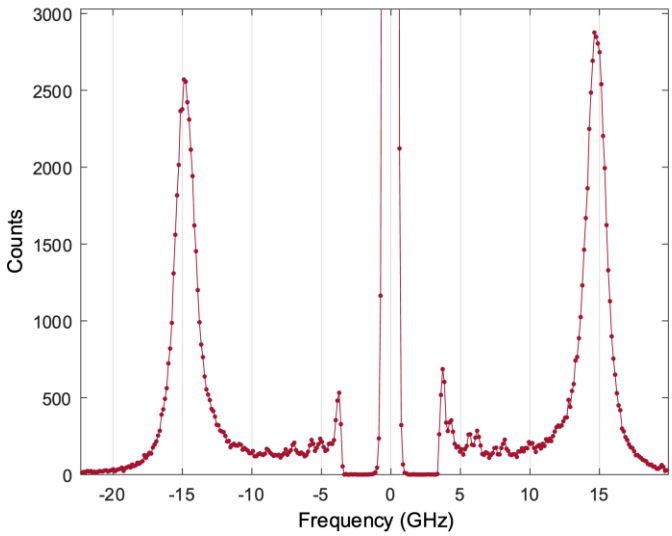

Figure 2: Brillouin spectrum for glass fibre

2.2. Brillouin light scattering experimental setup

The used setup is presented thoroughly in the paper by Ugarak et al. [9]. It consists of a laser (Nd-YAG laser) emitting at 532 nm of wavelength and 20 mW of power that passes through the composite sample. A fraction of the laser beam is directly oriented to a Fabry-Pérot interferometer by a beam splitter to generate a reference peak. The rest of the laser is directed to the surface of the sample of interest. Backscattered light is focused into a tandem Fabry–Pérot interferometer (TFP-2 HC) for filtering and detection. The position of the laser spot (around 3µm in radius) is monitored via a camera combined with a microscope. The sample is fixed onto an XY micrometric stage to control the position and the tilting of the sample. As shown in Figure 1, the laser beam is perpendicularly oriented to the transverse section of the fibre and the measurements are performed.

3. Results and discussions

Figure 3 summarises the measurements conducted on the various mini composites. It gives the longitudinal elastic modulus against the loss factor for the various tested fibres (flax, silk, glass, and PA11) and the Greenpoxy matrix.
Notably, flax fibres exhibit the highest measured modulus, reaching 92 to 100 GPa, while glass fibres follow closely behind with a modulus of approximately 76 GPa. In contrast, silk fibres demonstrate significantly lower modulus values at 2.9 GPa, while PA11 fibres exhibit the lowest modulus between 0.6 and 0.8 GPa. Considering the resin, its modulus falls around 6.5 GPa, which is higher than the data provided by the suppliers.

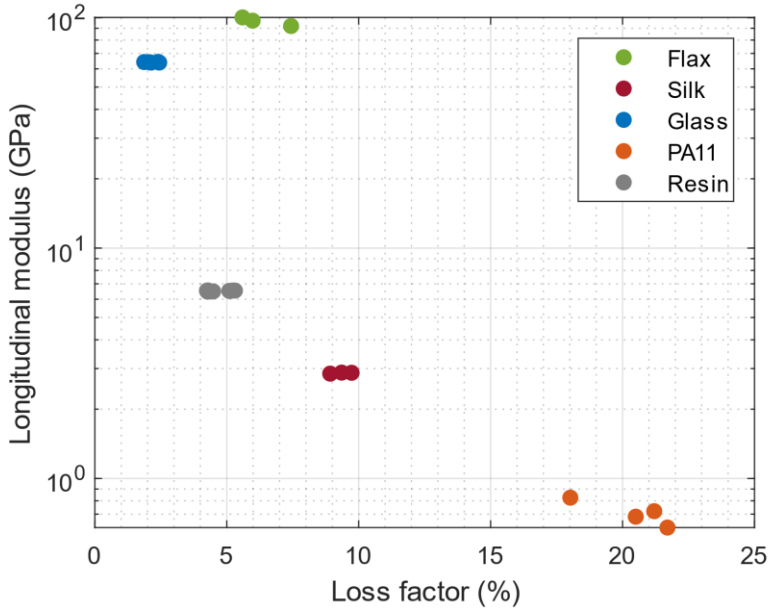

Figure 3: Longitudinal modulus against loss factor of different fibres inside Greenpoxy subjected to Brillouin spectroscopy

Shifting the focus to damping characteristics, figure 3 underscores a clear distinction: glass fibres exhibit lower damping compared to both their natural (flax and silk) and synthetic counterparts. Specifically, glass fibres present a damping level of around 1.8 - 2.4%, while flax fibres range between 5.5-7.5%. Silk fibres exhibit slightly higher damping levels, ranging from 8.9 – 9.7%, whereas PA11 fibres demonstrate the highest damping, with a loss factor between 18.2 - 21.7%. Finally, the resin presents a damping between 4.2 to 5.9 %. These findings provide valuable insights into both damping and modulus at the fibre scale within the resin. However, it is crucial to emphasise that the modulus identification is highly dependent on the Poisson's ratio. This implies that even a slight error in this parameter can lead to direct errors in the modulus determination. Therefore, the identified modulus for flax, silk and PA11 fibres can be discussed. Since the Poisson's ratio varies for these fibre types, the modulus is also influenced accordingly. Additionally, the refractive index is not easily measured and should be interpreted with caution. Indeed, the observed disparity is likely attributable to inherent uncertainties associated with the employed technique. Brillouin spectroscopy introduces several uncertainties and approximations, with the refractive index being a primary concern. While the refractive index value was sourced from the literature (refer to Table 1), it is important to note that it can vary with temperature and wavelength [8]. Additionally, morphological aspects such as anisotropy and changes in density in flax fibre are considered, introducing further uncertainties that need to be addressed for more consistent results.

However, it is noteworthy that the measured damping for flax fibre can be compared with values obtained using the nanoindentation method in the study by Liu et al [2]. Their research involved subjecting a flax-reinforced Greenpoxy composite to nanoindentation tests, with reported values within the fibre averaging at 4%. Although obtained at 5 Hz, this value closely aligns with the results presented in our study.

4. Conclusion

This paper underscores the dynamics of fibre-damping behaviour inside the composite using a Brillouin spectroscopy as a characterization method. Notably, flax, PA11, glass and silk fibre have significant differences in damping properties. While the technique used for modulus and damping characterization shows promising attributes, further exploration is needed. Indeed, the study's methodology reveals critical limitations, emphasizing the need for future investigations and methodological improvement.

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