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THE MODELLING OF THE DELAYED BEHAVIOUR OF KENAF BAST FIBRE BY THE CONTINUOUS SPECTRUM OF RELAXATION TIMES

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Abstract

The present study investigates the viscoelastic behaviour of kenaf bast fibre, utilising a modelling approach grounded in the continuous spectrum of relaxation times. The objective of this study is to enhance the comprehension of the long-term mechanical response of this plant fibre, with a particular emphasis on the phenomenon of stress relaxation, in order to augment the capacity to predict its behaviour in bio-based composite applications. The analysis of the master curve of the relaxation modulus in the time domain allows for the identification of the continuous range of relaxation times, estimated between 0.2117s and 66.4523s. Viscoelasticity has been identified as a determining factor in the mechanical behaviour of plant fibres subjected to static or dynamic stresses. Consequently, reliable prediction of their mechanical response necessitates thorough characterisation of the continuous distribution of their relaxation times. In this study, a model based on a continuous spectrum of relaxation times is proposed to describe the viscoelastic behaviour of kenaf's bast fibre. The results indicate a significant time dependence of this fibre, characterised by a broad distribution of relaxation times ranging from 0.016296 s to 100.4266 s, associated with spectrum values between 37.736 GPa and 6.732 GPa. This distribution is indicative of a heterogeneous microstructure, thus suggesting the existence of multiple internal mechanisms of energy dissipation. The models developed reproduce experimental curves with great precision, both in the linear and nonlinear viscoelastic range. This study underscores the significance of the continuous spectrum of relaxation times in describing the delayed behaviour of natural fibres. In addition, the tool provides a relevant function in the design and sizing of bio-based composites reinforced with kenaf fibres for structural and semi-structural applications.

Keywords: *kenaf bast fibre; viscoelasticity; modelling; continuous spectrum.*

Introduction

The quest for sustainable and eco-friendly materials has given rise to a surge of interest in plant fibres as reinforcements in composite materials[1]. This has resulted in a resurgence of interest in plant fibres, which are now being examined in numerous academic disciplines [2]. However, due to the inherent viscoelasticity of these fibres, their utilisation in textiles and composites for long-term applications poses a substantial challenge for researchers [3]. In this group, kenaf bast fibre (Hibiscus cannabinus L.) is particularly noteworthy for its low density, low cost, biodegradability, and high specific mechanical properties. These characteristics render it a promising candidate for applications in the automotive, construction, bio-based materials and textile sectors[4], [5]. However, as is the case with the majority of lignocellulosic fibres, kenaf fibres demonstrate a highly time-dependent mechanical behaviour, a consequence of their complex hierarchical structure, which consists primarily of cellulose, hemicellulose and lignin.

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This time dependence is manifested by creep, stress relaxation and viscoelasticity phenomena that directly influence the durability and reliability of composite structures subjected to prolonged loading [6].

In recent years, there has been a growing focus on the characterisation of the viscoelastic behaviour of natural fibres and bio-based composites, as evidenced by the numerous studies in this area. As demonstrated by [7], experimental studies conducted on kenaf fibres have shown that their delayed mechanical response is significant even at room temperature. Empirical models such as the Kohlrausch-Williams-Watts (KWW) function, Nutting's law, and Prony series have been successfully utilised to describe stress relaxation assays and predict the relaxation modulus of kenaf fibres [7–9]. However, it should be noted that these approaches are usually based on a limited number of parameters or discrete relaxation time spectra, which may limit their ability to accurately represent the diversity of dissipation mechanisms present in the plant microstructure.

Concurrently, recent advancements in viscoelastic modelling underscore the significance of representations based on a continuous spectrum of relaxation times. In contrast to the utilisation of a discrete set of viscoelastic elements in conventional Maxwell models, the continuous spectrum facilitates the description of an uninterrupted distribution of the relaxation mechanisms associated with the various structural scales of the material [10]. This approach provides a more realistic representation of heterogeneous and multi-scale materials, such as natural fibres. Recent research has demonstrated that direct identification of continuous spectra enhances the accuracy of the prediction of relaxation functions and facilitates a more profound comprehension of the physical mechanisms governing viscoelastic behavior [11]. We can cite, for example, the research work carried out on the elastic recovery and viscoelastic behaviour of Agave americana L. fibres. The fibre Agave americana L. has been observed to exhibit viscoelastic behaviour, due to the presence of crystalline regions, which serve as a source of reversible elastic deformations, and amorphous regions, which result in viscous deformations [12]. In a similar manner, research on the viscoelastic properties of kenaf bast fibre in relation to stem age indicates an increase in fibre stiffness with advancing maturity, accompanied by an increase in viscous response [13]. Furthermore, the viscoelastic behaviour of flax fibres at the micro and nano scale was analysed. This analysis demonstrated the deformation mechanisms of non-cellulosic polymers and amorphous cellulose, which are responsible for the viscoelastic behaviour of flax fibres [14]. Finally, a micromechanical analysis of bio-based polyurethane composites reinforced with flax fibres was conducted by [15]. The results obtained demonstrated a linear viscoelastic behaviour of flax fibres during stress relaxation tests, even under low levels of deformation.

Current research on plant fibres also underscores the necessity to establish a correlation between microstructural characteristics and macroscopic viscoelastic properties[16], [17]. Multi-scale models recently developed for natural fibres demonstrate that relaxation phenomena result from the complex interaction between cell constituents and the different layers of the cell wall [18]. The findings of this study serve to reinforce the notion that theoretical frameworks grounded in a continuous spectrum of relaxation time are particularly well-suited for the description of the delayed behaviour of lignocellulosic fibres.

In this context, the present work aims to develop a model of the delayed behaviour of kenaf bast fibres based on the concept of a continuous spectrum of relaxation times. The objective of this study is to identify a representation capable of accurately reproducing experimental stress relaxation curves while providing a physical interpretation of the viscoelastic mechanisms involved. This approach will facilitate enhanced prediction of the long-term behaviour of composites reinforced with kenaf fibres, thereby promoting their utilisation in sustainable structural applications.

It is from this perspective that the spectrum of relaxation times is identified from the master curve of the relaxation modulus in the time domain. Subsequently, a presentation of the models of linear and nonlinear fibre behaviour is provided. Subsequently, the continuous spectra of the relaxation times are validated in the linear domain, as in the nonlinear domain. A comparison is finally made between the experimental results of stress relaxation tests at different deformations and analytical predictions.

Empirical equation of the relaxation spectrum

The sigmoid function employed by [19] to represent the modulus of stress relaxation, E(t), is as follows:

$$E(t) = E_e + \frac{E_g + E_e}{1+\left(\frac{t}{t_0}\right)^n} \tag{1}$$

In the given model, *Ee* and *Eg* are, respectively, the long-term or relaxed and instantaneous or short-term modulus, t_0 and n are adjustable parameters of the model. The relationship that enables the relaxation modulus to be expressed as a function of the continuous spectrum of relaxation times is elucidated by[20].

$$E(t) = E_e + \int_{-\infty}^{+\infty} H(\tau) e^{-t/\tau} \frac{d\tau}{\tau}\ ;\ t > 0 \tag{2}$$

By posing $x = \frac{1}{\tau}\ \ et\ f(x) = \frac{H(1/x)}{x}$;

We obtain:

$$E(t) - E_e = \int_0^{+\infty} f(x)\, e^{-tx} dx \tag{3}$$

It is asserted that the function f(x) is indicative of the Laplace transform of the relaxation modulus, in accordance with the expression proposed by [21].

$$E(t) - E_e = \mathcal{L}(f(x)) \tag{4}$$

Replacing equation (1) in equation (4), we get:

$$\mathcal{L}\big(f(x)\big) = \frac{E_g - E_e}{1+\left(\frac{t}{t_0}\right)^n} \tag{5}$$

The application of the inverse Laplace transforms results in the following equation being obtained:

$$f(x) = \frac{E_g - E_e}{\Gamma(n)}\ t_0^n x^{n-1} e^{-x t_0} \tag{6}$$

Where $\Gamma(n)$ is the Euler gamma function of n.

The transformation of variables results in the subsequent analytic expression for the spectrum of relaxation times:

$$H(\tau) = \frac{(E_g - E_e)}{\Gamma(n)} \left(\frac{\tau}{t_0}\right)^{-n} e^{-t_0/\tau} \tag{7}$$

Materials and Methods

Materials

Kenaf fibres

The experimental results of stress relaxation tests on kenaf fibre were modelled using a continuous spectrum of relaxation times in order to analyse the influence of delayed effects on this fibre. [7].

Methods

Determination the expression of E(t) in terms of H(τ)

The relationship between the modulus of relaxation and the continuous spectrum of relaxation times is expressed in equation (2). It is possible to expand the expression of the continuous spectrum of relaxation times (7) to obtain the expression of the relaxation modulus.

Let us set once more: $x = \frac{1}{\tau}$ and $g(x) = \frac{H(1/x)}{x}$

It comes:

$$E(t) = E_e + \int_0^{+\infty} g(x) e^{-tx}\, dx \tag{8}$$

The expression of g(x) becomes:

$$g(x) = \frac{1}{x}\left[\frac{E_i - E_e}{\Gamma(n_i)} \left(\frac{1}{xt_i}\right)^{-n_i} exp^{(-xt_i)}\right] \tag{9}$$

$$g(x) = \frac{E_i - E_e}{\Gamma(n_i)} \frac{1}{t_i^{-n_i}} \left(\frac{1}{x}\right)^{1-n_i} exp^{-xt_i} \tag{10}$$

Replacing g(x) with the expression of E(t), we obtain:

$$E(t) = E_e + \frac{(E_i - E_e)}{\Gamma(n_i)} t_i^{n_i} \int_0^{+\infty} \left(\frac{1}{x}\right)^{1-n_i} exp^{(-x(t_i+t))} dx \tag{11}$$

The function E(t) is expressed as the Laplace transform of the function g(x).

$$E(t) = E_e + \frac{(E_i - E_e)}{\Gamma(n_i)} t_i^{n_i} \mathcal{L}(f(x))(t + t_i) \tag{12}$$

Where, $f(x) = \frac{1}{x^{1-n_i}}$

Let us set $\alpha = n_i - 1$

$$\mathcal{L}(x^\alpha) = \frac{\Gamma(1+\alpha)}{t^{1+\alpha}} = \frac{\Gamma(n_i)}{t^{n_i}} \tag{13}$$

$$\mathcal{L}(f(x))(t + t_i) = \frac{\Gamma(n_i)}{(t+t_i)^{n_i}} \tag{14}$$

Following the inverse Laplace transform, the relaxation modulus is obtained:

$$E(t) = E_e + \frac{(E_i - E_e)}{\Gamma(n_i)} t_i^{n_i} \times \frac{\Gamma(n_i)}{(t+t_i)^{n_i}} \tag{15}$$

Subsequent to the arrangement process, the following expression is obtained:

$$E(t) = E_e + (E_i - E_e)\left(\frac{t_i}{t+t_i}\right)^{n_i} \tag{16}$$

Linear modelling of the viscoelastic behaviour of kenaf fibre

Most of viscoelastic models contain linear and nonlinear viscoelastic parameters and the linear viscoelastic parameters correspond to relaxation time spectra of materials [22]. The spectrum of relaxation times will be identified from the master curve of the relaxation modulus in the time domain. It is an established fact that a spectrum of relaxation times is a fundamental feature of a linear viscoelastic material, from which other viscoelastic functions and responses can be obtained [23]. The master curve presented hereinafter was derived from the research conducted by [7]. The evolution of the master curve of the relaxation modulus can be delineated into three zones, as illustrated in Figure 1 below. The three zones are composed of a light plateau with brief periods, a zone of rapid relaxation (viscoelastic), and a final zone with extended periods, which is represented by the relaxed module. It can thus be concluded that two slopes (n1 and n2) provide a connection between the three zones. The sigmoid function equation (1) facilitates the identification of the spectrum of relaxation times through the modelling of the modulus of relaxation. This function enables the reproduction of a single evolution of the slope, as outlined by [21].

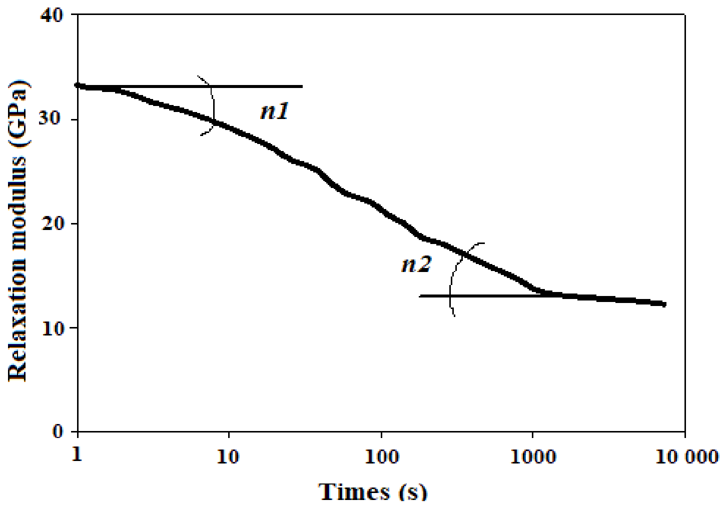

Fig. 1. Representation of slopes on the relaxation master curve

Nonetheless, the master curve of the relaxation modulus in this instance exhibits two slopes (see Figure 1). It is imperative to model the evolution of the relaxation modulus by a combination of two functions, as outlined below:

$$E(t) = E_e + \frac{E_{g1} - E_e}{1 + \left(\frac{t}{t_1}\right)^{n_1}} + E_e + \frac{E_{g2} - E_e}{1 + \left(\frac{t}{t_2}\right)^{n_2}} \tag{17}$$

Consequently, an analytic expression of the continuous spectrum of relaxation times, which can be used to represent the linear behaviour of the kenaf bast fibre, can be obtained from two contributions as follows:

$$H(\tau) = \frac{(E_1 - E_e)}{\Gamma(n_1)} \left(\frac{\tau}{t_1}\right)^{-n_1} exp^{\frac{-t_1}{\tau}} + \frac{(E_2 - E_e)}{\Gamma(n_2)} \left(\frac{\tau}{t_2}\right)^{-n_2} exp^{\frac{-t_2}{\tau}} \tag{18}$$

Nonlinear modelling of the viscoelastic behaviour of kenaf fibre

In the course of mechanical testing, the nonlinear behaviour of the plant fibre is contingent on the imposed deformation [14];[24];[25];[26];[27]. This non-linearity is perceived by these authors as a viscoelastic behaviour of the plant fibre. However, the analysis of the master stress-relaxation curve reveals that the rapid relaxation zone appears to be subject to modification in the presence of deformation. Therefore, the nonlinear behaviour of the plant fibre must be modelled. [28] model is the same as resetting the n_2. Consequently, the parameter in question is contingent on the deformation that is applied to the fibre. Consequently, the following assertion can be made.

$$n_2 = n_2(\varepsilon) \tag{19}$$

The expression of the parameter n_2 as a function of the deformation can be obtained from the following decreasing function:

$$n_2(\varepsilon) = \frac{n_2(0)}{1+\left(\frac{\varepsilon}{\varepsilon_r}\right)^{\gamma}} \tag{20}$$

In the context of the aforementioned equation, the initial value, designated as n_2 *(0)*, is obtained from the sigmoid function equation (1). The term ε denotes the imposed strain, while ε_r and γ represent two parameters that are calculated through the utilization of the 'NonLinearModelFit' command within the MATLAB environment.

Results and Discussions

Linear modelling

Table 1 presents the mean values of the parameters that were estimated by the MATLAB "NonLinearModelFit" command. The model identified from the master curve of the relaxation modulus is illustrated in Figure 2. It is evident that the resultant parameters are not unique; therefore, the set of parameters that is selected is the one that best minimizes the deviation between the model and the experimental points in the designated time interval. This is achieved with a root mean square error of 0.025. In a similar vein, a strong correlation is observed between the model and the experimental results, thereby validating the necessity to decompose the function into two contributions.

Table 1. Key features and mechanisms of generative deep learning models for molymer design

Ei (GPa)			*ti* (s)		*ni* [adim]	
Ee	$\boldsymbol{E_1}$	$\boldsymbol{E_2}$	$\boldsymbol{t_1}$	$\boldsymbol{t_2}$	$\boldsymbol{n_1}$	$\boldsymbol{n_2}$
5.52	32.6	28.89	0.2117	66.4523	12.991	0.6617

As illustrated in Figure 3, the spectrum of relaxation times of plant fibres is shown as a function of the logarithm of time. However, since the spectrum is also divided into two contributions, each contribution reaches a local maximum at a well-defined characteristic time. It can thus be concluded that a primary characteristic time, τ, is obtained, which is equal to t_1/n_1 (0.016296 s). This is linked to the initial contribution, which could be perceived as the signature of the molecular mobility of the long chains of cellulose. This is evidenced by the observation of some possible movements, such as the vibration of molecules, the bending or twisting of chains of molecules at the atomic scale. A secondary temporal characteristic, designated as τ, is expressed as t_2/n_2, with a recorded duration of 100.4266 seconds. This particular temporal value is attributed to the movement of all the long cellulose chains over an extended time period. The maximum values of the spectra at the different characteristic times are, respectively, 37.736 GPa for the first peak and 6.732 GPa for the second peak, sufficiently showing the viscoelastic character of kenaf bast fibres. Recent literature indicates a paucity of studies dedicated to identifying the continuous spectrum of relaxation times of plant fibres. The methods developed are essentially validated on generic viscoelastic materials (i.e. polymers, biomaterials and elastomers)[28], [29]. It is evident that the identification of the continuous spectrum of relaxation times of the Kenaf bast fiber aligns seamlessly with this logic.

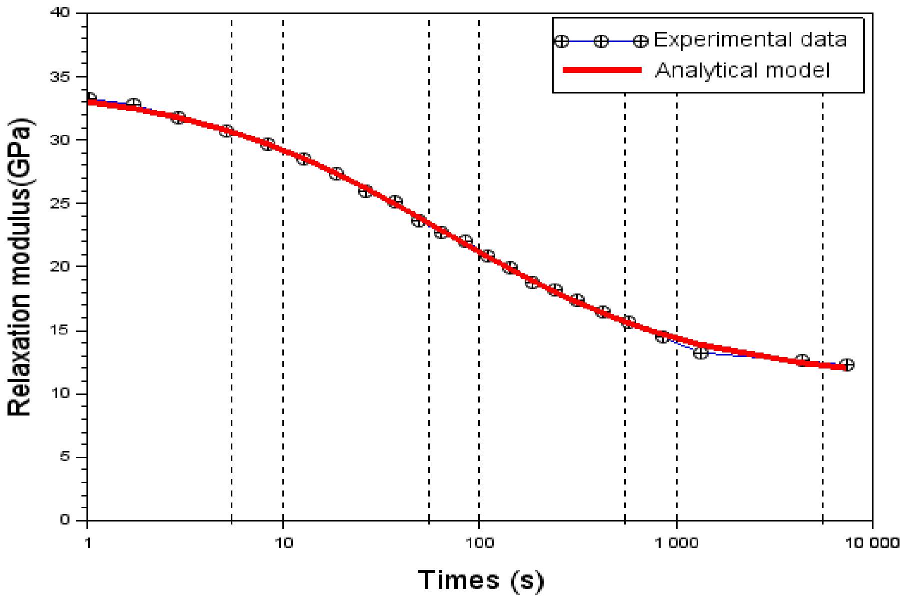

Fig. 2. comparison the experimental master curve of the relaxation modulus and the sigmoidal function

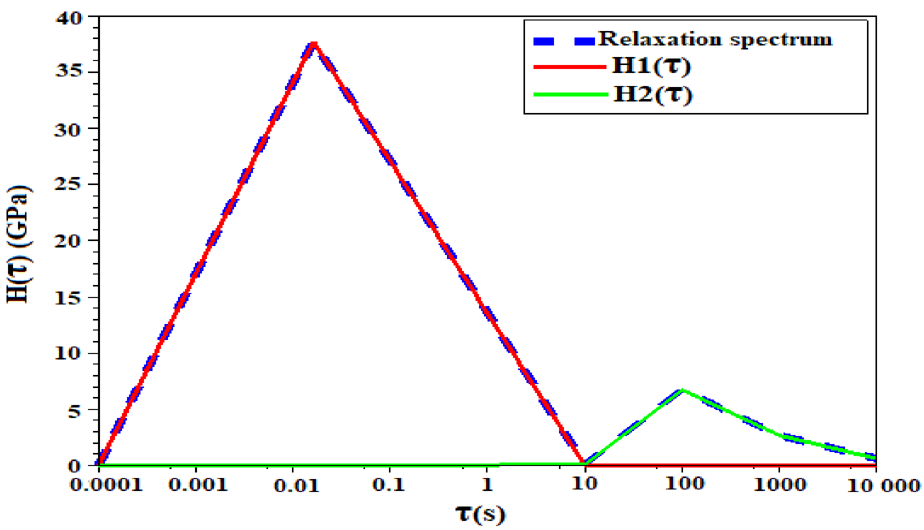

Fig. 3. Evolution of the continuous spectrum as a function of the logarithm of the relaxation times

The relaxation time spectrum, in itself, has the capacity to describe the viscoelasticity of a given material. Furthermore, the shape of the continuous spectrum of relaxation times, which vary in narrowness (see figure 3), would indicate a significant contribution of viscous behaviour during the stress deformation of the plant fibre. The validation of the continuous spectrum of relaxation times in the linear domain is achieved through the utilisation of equation (16), which has been developed for this purpose. Accordingly, the expression of the relaxation modulus can be obtained in the following manner:

$$E(t) = E_e + (E_i - E_e)\left(\frac{t_i}{t+t_i}\right)^{n_i} \tag{21}$$

In the event of i = 2, this process enables the combination of two equations, thus facilitating the validation of the model illustrated in Figure 4. This figure demonstrates the evolution of the relaxation modulus, obtained with respect to the experimental points. A strong correlation is evident between the predicted data and the experimental points. The mean relative error between the analytic model's prediction and the experimental points is approximately 3.93%. However, this discrepancy could be linked to measurement errors, the construction of the master relaxation curve, as well as the non-uniqueness of the parameter identification response.

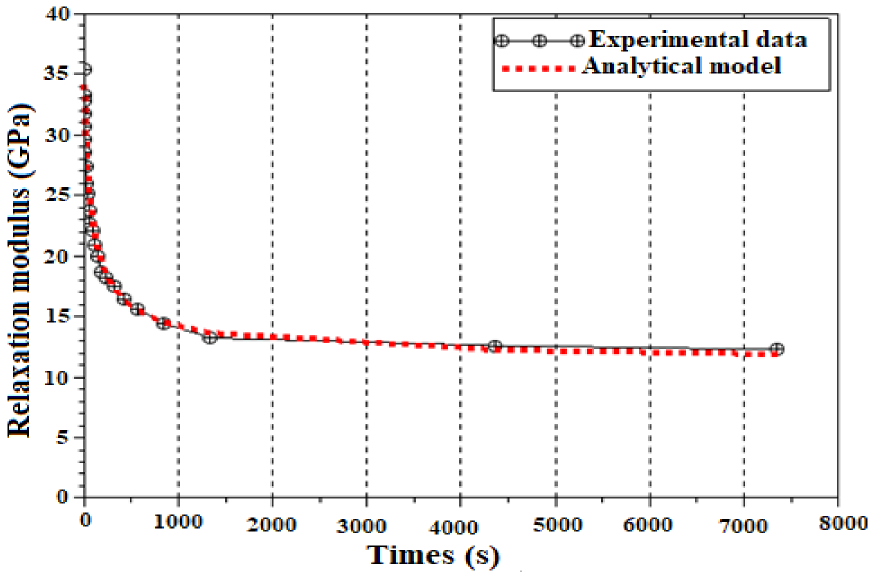

Fig. 4. Comparison between the master relaxation curve and the analytical model

Nonlinear modelling

Subsequent to the redefinition of the parameter n2 in accordance with the deformation and its introduction into the expression of the relaxation time spectrum, the figure 5 is obtained. It is evident that the extent of deformation correlates directly with the decrease in amplitude of the secondary peak, see figures 6 and 7. However, for large deformations, the second peak tends to zero. Cellulose is a natural polymer whose molecules are formed of long chains. Consequently, under substantial deformations and over extended periods, the fibres rigidity is known to increase, leading to a complete cessation of molecular movement within the cellulose chains. The spectrum of relaxation times once again demonstrates the influence of deformation on the viscoelastic behaviour of the plant fibre.

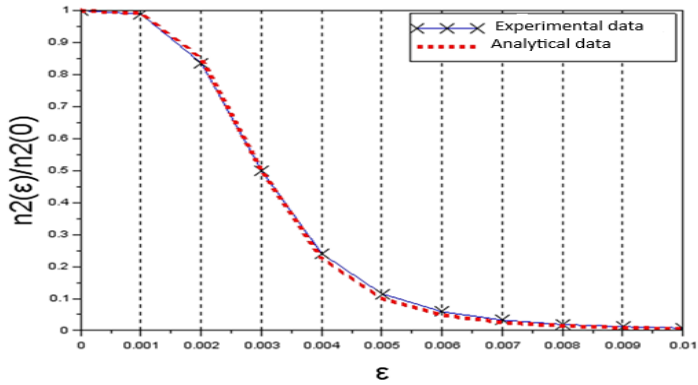

Fig. 5. The evolution of the parameter n2 in relation to the deformation

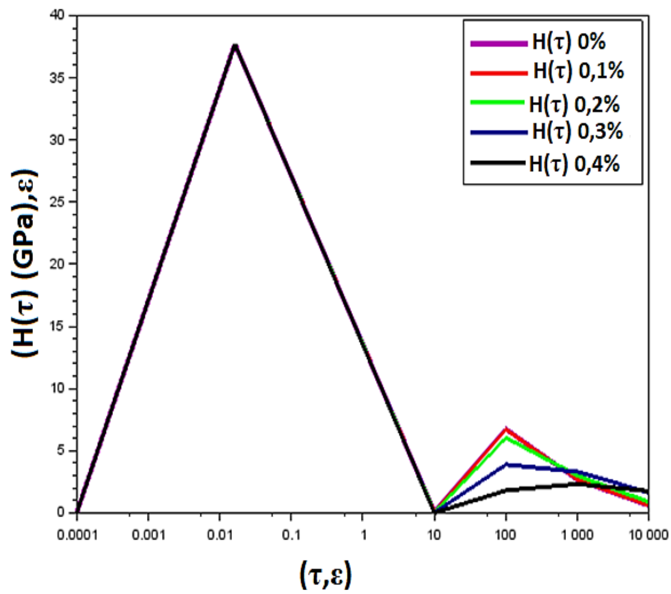

Fig. 6. Evolution of the spectrum as a function of logarithm of time and strain

In this study, a nonlinear viscoelastic model is derived from its continuous spectrum of relaxation times, incorporating the variation of deformation:

$$H(\tau) = \frac{(E_1-E_e)}{\Gamma(n_1)}\left(\frac{\tau}{t_1}\right)^{-n_1} exp^{\frac{-t_1}{\tau}} + \frac{(E_2-E_e)}{\Gamma(n_2)}\left(\frac{\tau}{t_2}\right)^{-n_2} exp^{\frac{-t_2}{\tau}} \quad (22)$$

$$n_2(\varepsilon) = \frac{n_2(0)}{1+\left(\frac{\varepsilon}{\varepsilon_r}\right)^{\gamma}} \quad (23)$$

$$E(t) = E_e + (E_1 - E_e)\left(\frac{t_1}{t+t_1}\right)^{n_1} + E_e + (E_2 - E_e)\left(\frac{t_2}{t+t_2}\right)^{n_2} \quad (24)$$

The nonlinear viscoelastic model obtained from its continuous spectrum of relaxation times has been verified, as previously demonstrated in equation (24). Initially, the model is compared to the experimental data of the master curve. Subsequently, analytical predictions are made at varying degrees of deformation based on the model's correlation with experimental data. Consequently, with each imposed deformation, a phenomenon of stiffening of the relaxation modulus is observed over time, which makes it possible to obtain the predicted relaxation curves illustrated in Figure 8.

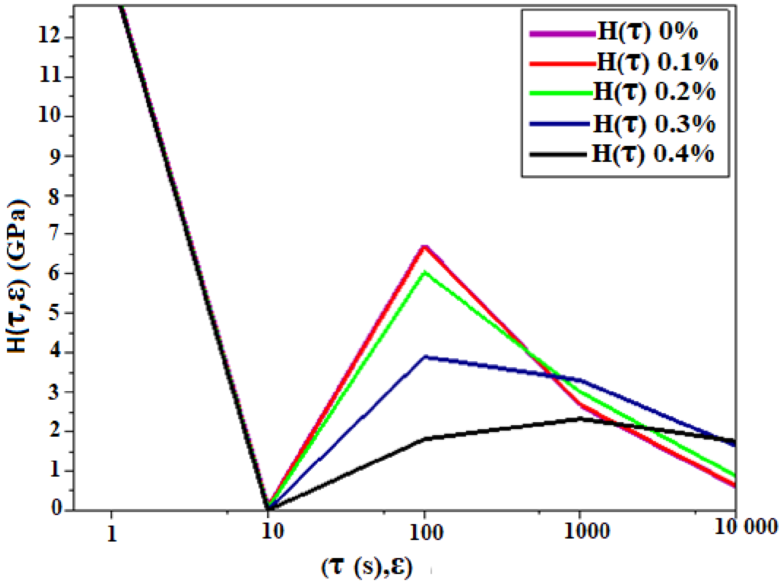

Fig. 7. Variation of the continuous spectrum as a function of strain

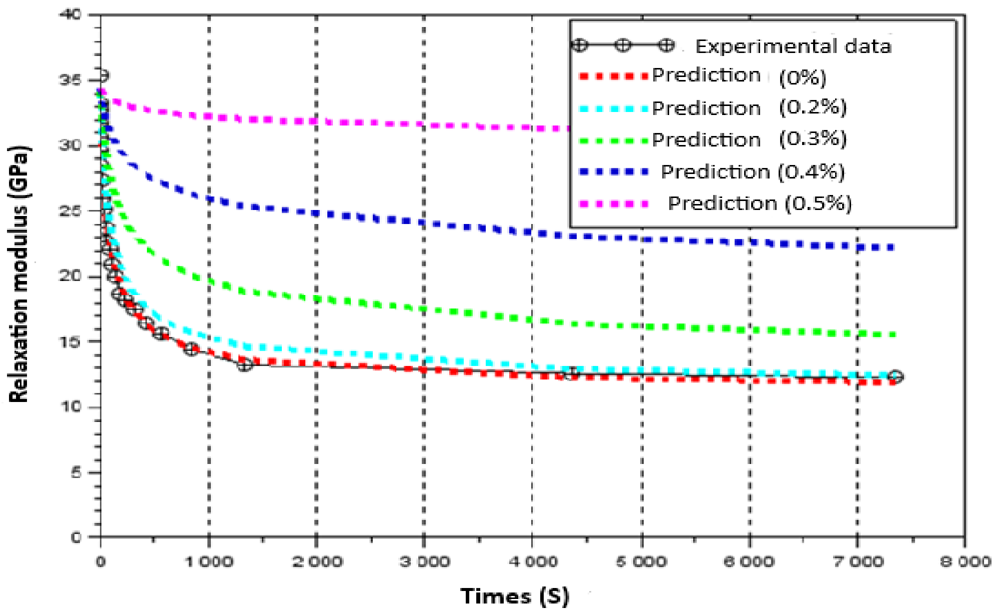

Fig. 8. Predictions analytical of the non-linear viscoelastic model of single-axial deformations

As illustrated in Figure 9, a comparison is presented between the experimental outcomes of stress relaxation tests at varying degrees of deformation and the analytical predictions derived from these tests.

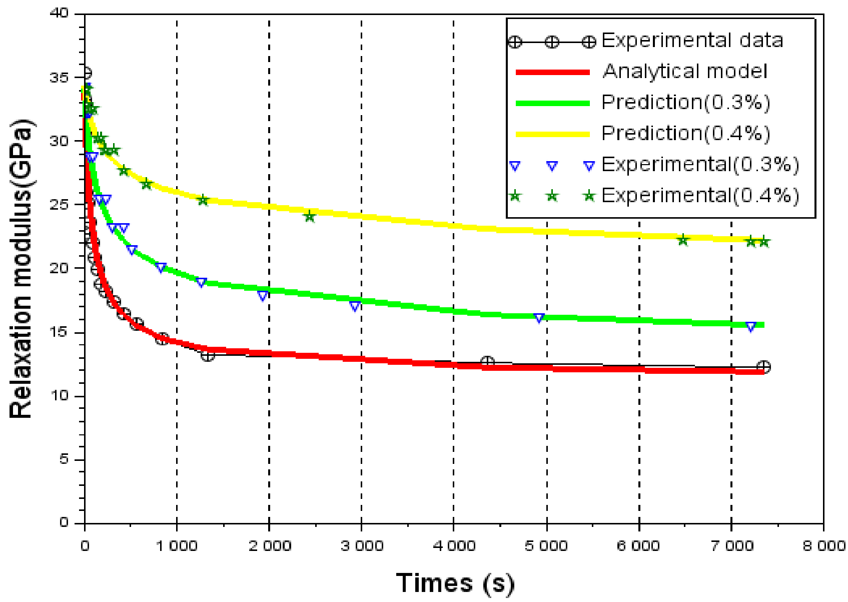

Fig. 9. Comparison between experimental points and analytical predictions of the nonlinear viscoelastic model

A strong correlation is evident between the model and the experimental points of the relaxation modulus over time as a function of the imposed deformation. This outcome serves to reinforce the validity of the nonlinear model and its capacity to simulate a relaxation test for variable deformations.

Conclusion

This study enabled the efficient modelling of the delayed behaviour of the kenaf bast fibre using the continuous spectrum of relaxation times. The employment of a relaxation function, comprising two contributions, facilitated the formulation of an expression for a continuous spectrum of relaxation times. Initially, this spectrum was utilized to model the linear viscoelastic behaviour of the fibres. Subsequently, the consideration of static deformation by modifying the second contribution enabled the modelling of the nonlinear viscoelastic behaviour of fibres. The findings demonstrate that the function employed for the identification of the continuous spectrum of relaxation times is capable of reproducing the master curve of the relaxation modulus. This finding serves to underscore the significance of the continuous spectrum in predicting the long-term behaviour of natural fibres. This work opens up important prospects for the optimization of the design of bio-based composite materials, in particular for applications requiring good dimensional stability over time. However, the validity of this model could be established through comparison of the prediction with the experimental results of other types of tests, such as the response to a more complex load.

Future developments will focus on extending the proposed methodology to coupled hygro-thermo-mechanical loading, where moisture diffusion and temperature significantly affect the viscoelastic response of natural fibres. Additional investigations will also address the evolution of the relaxation spectrum during ageing, fatigue and environmental degradation, as well as comparisons with fractional viscoelastic models and data-driven identification techniques. These perspectives will contribute to establishing reliable predictive tools for lifetime assessment and structural design of sustainable bio-composite materials.

Overall, this study demonstrates that the continuous spectrum of relaxation times is not merely an alternative mathematical representation of viscoelasticity but a powerful physical modelling framework capable of linking the hierarchical microstructure of kenaf bast fibres to their long-term mechanical behaviour. It therefore provides a significant step towards the development of robust, physically based constitutive models for natural fibres and their structural composite applications.

Credit authorship contribution statement

Saïdjo: was responsible for the experimental work, analysis of the data, interpretation of the results and preparation of the manuscript; Djoda Pagoré Frédéric: participated in the analysis of the data, interpretation of the results, preparation of the manuscript and Ntenga Richard: approved the final version of the document.

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