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Characterisation of the Transverse Thermoelastic Properties of Natural Fibres used in Composites

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Abstract

To predict the properties of a composite, it is necessary to identify the properties of the constituent materials, especially those of the fibre. Mechanical properties of natural fibres (NF) are anisotropic and cannot be characterised in the same way as isotropic materials. Therefore further characterisation of the natural fibres is needed to determine their transverse thermo-mechanical behaviour. An understanding of the thermoelastic anisotropy of natural fibres is important for defining their performance in potential composite applications. In this study, thermoelastic properties of flax and sisal fibres where determined through a combination of experimental measurements and micromechanical modelling. Dynamic mechanical thermal analysis and thermal mechanical analysis techniques were employed to characterise model unidirectional NF- epoxy composites over a range off-axis loading angles. These results were input into a number of micromechanical and semi-empirical models to determine the transverse and longitudinal thermoelastic properties of the fibre. The results confirm the high degree of anisotropy in properties of the flax and sisal fibres.

1 Introduction

Natural fibre composites eco-profile has increased due to the growing concerns about the environment, depletion and price of oil and the new regulations for a cleaner and safer environment. This has lead to a great deal of scientific research being focused into natural fibre also known as ‘eco – composite’[1-5]. Several researchers argue that natural fibres can successfully compete with glass fibres in today’s market because of their eco profile and attractive properties. These properties include low cost, low density, good specific strength properties, renewable and produce neutral CO2 emissions when burned and there is an unlimited supply. Natural fibres are less abrasive than conventional fibres and involve little energy to produce. This provides advantages in material processing as it will help the tooling life and recycling [1, 3, 6].

To effectively predict the properties of a composite, it is essential to know the constituent materials. Simple relationships that characterise isotropic materials are not sufficient, due to the significant structural anisotropy of natural fibres. Therefore further investigation is needed to effectively measure their thermo mechanical behaviour. Unfortunately the majority of research to date has focused on longitudinal properties and not on the degree of anisotropy of natural fibres [1-4]. Although the longitudinal modulus of the natural fibres can be directly measured, measuring the transverse properties of the fibre is more challenging due to the complex structure of the fibre. The transverse fibre properties can be estimated by using a range of micro-mechanical and semi-empirical models.

A number of researchers have recently reported measuring the thermo-mechanical behaviour and degree of anisotropy of natural fibres in composites to help establish their true potential [7-9]. Cichocki and Thomason [7] explored the five thermo-elastic constants of jute fibres using a specified volume of unidirectional composites and micro-mechanical models. It was noted that the longitudinal Young's modulus of the jute fibre exceeded the transverse modulus by a factor of five. Ntenga et al [9] used the inverse method developed by Levenberg-Marquardt to approximate the elastic anisotropy of sisal fibre and found that the sisal fibre was also highly anisotropic [10]. Baley at al [5] used micromechanical models to estimate the transverse Young's modulus of the flax fibre. It was found that the transverse modulus was 8GPa and compared to the longitudinal modulus of 59GPa. These results emphasizes how anisotropic natural fibres are and this can be caused by their highly complex structure. The structure and natural composition of natural fibres can be influenced by global location and climate. The highly complex structure is made up from hollow cells and microtubules generally orientated along the fibre direction. The microtubules are made up from various substances; these substances include cellulose, hemicelluloses and lignin [11]. The fibre stiffness is reported to be influenced by the spiral angle of the crystalline micro fibrils and the concentration of non cellulosic substances [5, 11]. Natural fibres contain pores that are essential for the plant to live as it allows them to breath and grow but can be regarded as a defects when mechanical modelling a single fibre.

In this study, thermoelastic properties of flax and sisal fibres where determined through a combination of experimental measurements and micromechanical modelling. Dynamic mechanical thermal analysis (DMA) and thermal mechanical analysis (TMA) techniques were employed to model unidirectional natural fibre - epoxy composites over a range off-axis loading angles. These results could be input into a number of micromechanical and semi-empirical models to determine the transverse and longitudinal thermoelastic properties of the fibre.

2 Materials

2.1 Fibre and Matrix

Natural fibres that was used for the investigation was untreated flax and sisal with an epoxy matrix. The epoxy matrix was Araldite 506 and a room temperature curing agent (triethylenetetramine) sourced from Sigma Aldrich were used to manufacture the natural fibre composite samples. The ratio of resin to curing agent was approximately 176grams to 24 grams respectively to make the matrix for the composite. The resin and curing agent mixture was left to cure overnight at room temperature before the mixture was post cured at 120^{o}C for 2 hours. The flax fibre that was used in the composite was sourced from Wigglesworth fibres and was grown in Germany and the Sisal fibres were also sourced from the same company but were grown in Brazil.

2.2 Composite Manufacturing

The natural fibre composites were made by a vacuum infusion process. The process manufactured unidirectional composite plates that were then cut and prepared for testing. A

volume fraction of 70% can be achieved through vacuum infusion as it reduces the voids in the composites. The process of vacuum infusion of natural fibre composite is discussed further by Symington [12]. Fibre mats were layered by hand, attempting to keep the fibres as unidirectional as possible. Vacuum to the rig allows smooth adjustable filling of resin into a mould and is achieved by using a vacuum pump via a pressure pot. The ideal infusion rate of approximately 5mm per second can be controlled by adjusting the inlet and outlet vales. A slow infusion rate ensures a reduction in voids. High compaction of the fibres without significant damage to the microtubules of each fibre is achieved through the sandwiching of aluminium and acrylic outer plates using a number of bolt fixings and G-clamps [12]. The infused composite is then left to cure at room temperature in the mould for 12 hours and then post cured at 120^{o}C for 12 hours. Post curing was done as natural fibres are highly hydrophilic so the composites were dried to achieve accurate results. A Struers Accutom 5 high speed precision cutting wheel with an aluminium oxide blade was used to accurately machine samples for thermal analysis. Samples for DMA and TMA were cut to approximately 55x14x2mm and 4x4x1mm respectively. The TGA sample size was cut the same size as TMA, 4x4x1mm.

3 Experimental

Thermal analysis was used on the natural fibre composites to find the weight change in relation to the temperature change, transverse modulus, glass transition temperature and coefficient of thermal expansion. The three pieces of equipment that were used were the thermogravimetric analysis, thermomechanical analysis and the dynamic mechanical analysis.

3.1 Thermogravimetric Analysis (TGA)

A Thermomechanical analysis machine (Q50 TA Instruments) was used to determine amount of weight change in relation to the temperature change and was also used to remove moisture from the sample as natural fibres are highly hydrophilic. The procedure used was to ramp the temperature of the sample from room temperature (20^{o}C) to 120^{o}C at a rate of 10^{o}C per minute and then hold at this temperature for 60minutes. This allowed the sample to be dried off. The sample was then put into a desiccator so that further tests could be run on the dry sample.

3.2 Thermomechanical Analysis (TMA)

A thermomechanical analysis machine (Q400 TA Instruments) was used to determine changes in the material properties. The TMA was used to determine the glass transition temperature and the expansion coefficient of the natural fibre composite. A macro expansion probe was used to allow a large contact area of the sample to be tested. Therefore more accurate readings could be obtained as the natural fibre composite surface is uneven. The composites were tested at different fibre orientations. The orientations of the fibre were 0^{o}, 25^{o}, 45^{o}, 65^{o}, 90^{o}. The TMA samples used were the samples that had be left on the desiccator from the TGA. The start temperature for the samples was -50^{o}C and increased at a rate of 3^{o}C/minute until the temperature of 150^{o}C had been reached. This allowed the glass transition temperature to be passed (approximately 90^{o}C) as the physical properties of the composite such as the coefficient of thermal expansion can be affected.

3.3 Dynamic Mechanical Analysis (DMA)

The Dynamic Mechanical Analysis machine (Q800 TA Instrument) was used to determine the transverse modulus of the composite. The samples were dried in the oven at 120°C for 2 hours and then stored in a desiccator with silca crystals to avoid the composites absorbing water until they are required for testing. The input parameters for the DMA were that the starting temperature was -50°C and ramped to 150°C at a rate of 3°C/minute with a preload of 0.1N and a frequency of 1Hz. The fibre orientation of the flax was 0°, 10°, 20°, 30°, 40°, 50°, 60°, 70°, 90° and the sisal orientation was 0 °, 10°, 20°, 30°, 40°, 50°, 70°, 80°, 90°.

3.4 Fibre Volume Fraction Measurements

Volume fraction of the constituent material to effectively predict the purpose of a composite therefore an accurate measurement of volume fraction is extremely important. A technique based on the tendency of natural fibres to absorb high levels of moisture in relation to their environment was utilised to measure the volume fraction of fibre in composite samples. The composite moisture absorption is dominated by the higher hydrophilic nature of the natural fibre compared to the matrix material therefore a higher fibre content leads to a higher amount of absorbed moisture. Monitoring the uptake of moisture in of moisture in fibre, composite and resin samples under strict environmental conditions allows us to determine the volume fraction of the constituent materials using the percentage weight gain. Using the thermogravimetric analysis (TGA) the percentage weight loss due to desorption of moisture from the constituent material can be monitor therefore volume fraction can be determined.

6 Background on Micro-Mechanical and Semi- Empirical Models

The longitudinal mechanical properties of composites and their constitutive materials can be determined experimentally however the transverse properties of the natural fibres of the composites can be determined using a number of micromechanical and semi-empirical models. Using these complex mathematical models can reduce costly and time consuming testing but these models are used for ideal composites therefore the values that are calculated are treated as estimates.

In order to predict the properties of the composite fundamental quantities must be know such as the elastic modulus, Poison's ratio, densities and volume fractions of the constituent materials. The relationship between off-axis Young's modulus, E_Θ, and the principal properties of a 2-D composite ply is shown below [7]:

$$E_\theta = \frac{1}{\frac{1}{E_1}.cos^4\theta + \frac{1}{E_2}.sin^4\theta + \left[\frac{1}{G_{12}} - \frac{2\nu_{12}}{E_1}\right].cos^2\theta.sin^2\theta} \quad (1)$$

Composite samples may be tested at three different fibre orientations, Θ, to determine the values of E_1, E_2 and $(1/G_{12} - 2\nu_{12}/E_1)$. Where E_Θ, E_1 and E_2 are the off-axis, longitudinal composite and transverse composite modulus respectively, G_{12} is the composite shear

modulus and v_{12} is the longitudinal composite Poisson's ratio. This data may then replace with either Poisson's ratio or shear modulus data to determine all the relevant longitudinal elastic properties of the composite ply.

Voigt's rule of mixtures is one of the simplest and most popular models used to predict the elastic properties of a composite material. The rule of mixtures is the relationship between E_1, E_{1f} and E_m and is illustrated below:

$$E_1 = E_{1f}.V_f + E_m.V_m \quad (2)$$

E_{1f}, E_m, V_f, V_m are the elastic modulus and volume fraction of the fibre and the matrix respectively. A similar expression for Poisson's ratio can also be derived, where the values of modulus are substituted for the equivalent Poisson's ratio gives:

$$v_{12f} = \frac{(v_{12} - v_m V_m)}{V_f} \quad (3)$$

This expression is most effective with uni-directional continuous fibres, where a basic assumption of equal strain in the two material constituents holds true [13].

A straightforward equation to predict the transverse modulus of the fibre, E_{2f} is the rule of mixtures. The equation below can be rearranged to get the fibre modulus.

$$E_2 = \frac{v_f}{E_f} + \frac{v_m}{E_m} \quad (4)$$

Halpin and Tsai developed semi – empirical equations which are used to predict the elastic properties, particularly the transverse and shear moduli of the fibre reinforced composites [10].The Halpin-Tsai equation can be used to solve for the transverse modulus of the fibre, E_{2f} [7]:

$$E_{2f} = \frac{E_m(\eta.\beta + 1)}{(1 - \beta)} \quad (5)$$

$$\beta = \frac{\left(\frac{E_2}{E_m} - 1\right)}{\left(\eta V_f + \frac{E_2}{E_m} V_f\right)} \quad (6)$$

Where η is a curve fitting parameter that takes into consideration the distribution of the reinforcing fibre. Several researchers have adopted η=2 when applying the Halpin-Tsai relationship and with insufficient experimental information to accurately identify this curve fitting parameter for the natural fibre composites, a value of η=2 has likewise been adopted in this study [7].

Another semi–empirical method for determining the transverse modulus of the fibre in a composite was developed by Tsai and Hahn. The method assumes that the transverse stress in

the matrix is less than the stress carried by the fibre and thus a stress partitioning factor, η was developed to derive the expression [7]:

$$E_{2f} = \frac{V_f}{\left(\frac{V_f + \eta . V_m}{E_2} - \eta . \frac{V_m}{E_m}\right)} \quad (7)$$

Where the symbols have their usual meanings. Several researchers have adopted a value η = 0.5 [7] for natural fibres therefore this value has been adopted for this study. Like many of these micromechanical and semi – empirical models, many of the properties can be interchanged. An example of this is the fibre longitudinal shear modulus can be obtained by substituting G_{12f}, G_{12} and G_m for E_{2f}, E_2 and E_m.

5 Results and Discussion

5.1 Elastic Anisotropy

The volume fraction of the flax and sisal composite was found to be 68% and 41% respectively. Poisson's ratio was found by tensile testing of the sample using a bi-axial strain gauge. The ratio of the resin, flax and sisal composite were found to be 0.36, 0.49 and 0.38 respectively.

The off axis Young's modulus, E_Θ (equation 1) is related to the longitudinal modulus, E1, the transverse modulus, E2, the longitudinal Poisson's ratio v_{12} and the shear modulus, G_{12} of the composite. E_Θ was measured by the DMA and v_{12} was measured by the composite ply. E_1 and E_2 could be approximated as E_0 and E_{90}. The longitudinal shear G_{12} has been used to curve fit equation 1.

The experimental and calculated data for the off axis Young's modulus plotted as a function of temperature can be seen for Flax and Sisal in Figures 1 and 2. It can be seen that the relationship between the calculated and experimental values are reasonable, therefore the micromechanical models can be employed to determine the elastic properties of the fibre. It can be seen that the relationship between the calculated and the experimental values are reasonable except for the sisal 20º and flax 10º orientation experimental data. This may be explained by the volume fraction of the fibre and/or the void content being different to the other composite samples.

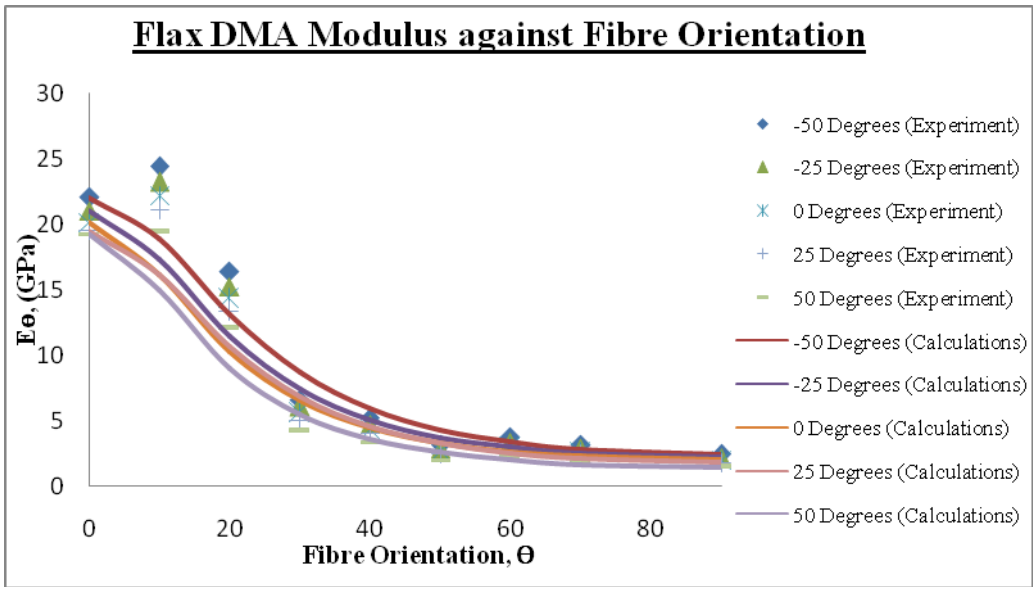

Figure 1: Modulus of flax composite plotted as a function of fibre orientation angle. Experimental Data has been curve fit with Equation 1 by adjusting the Value G_{12}.

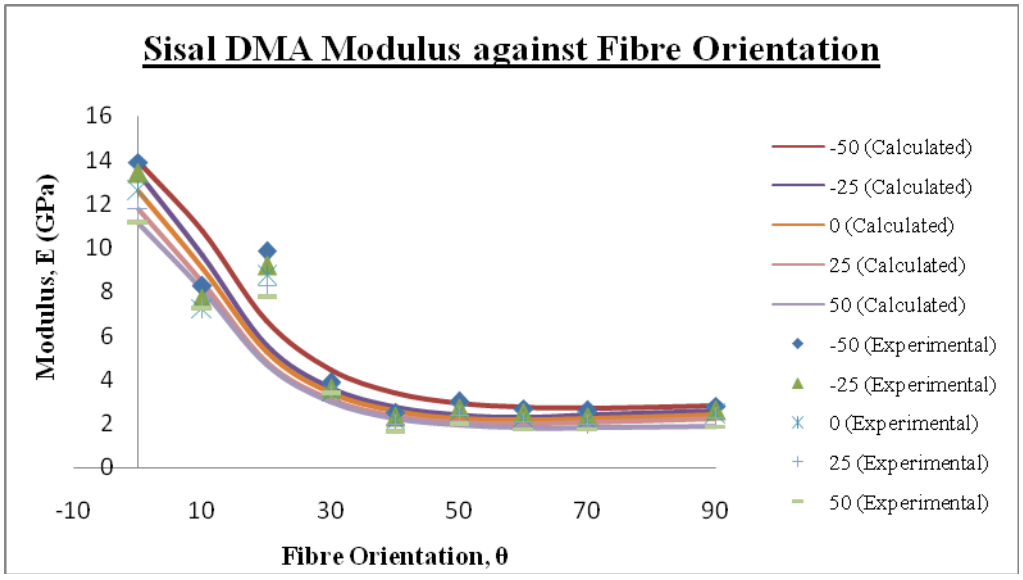

Figure 2: Modulus of flax composite plotted as a function of fibre orientation angle. Experimental Data has been curve fit with Equation 1 by adjusting the Value G_{12}.

Figure 3 illustrates the longitudinal Young's modulus of the fibre, E_{1f} against the transverse Young's modulus of the fibre, E_{2f}, presented as a function of temperature and highlights the degree of anisotropy evident with natural fibres. E_{1f} was calculated using the Voigt's rule (equation 2) and E_{2f} was calculated using an average of three Micromechanical models: Halpin and Tsai (equation 5), Inverse rule of mixtures (equation 4) and Tsai and Hahn (equation 7) and can been seen in Table 1 and 2. A comparison of the micromechanical and semi –empirical models used to predict the transverse Young's modulus of the fibre, E_{2f} are shown in figure 4.

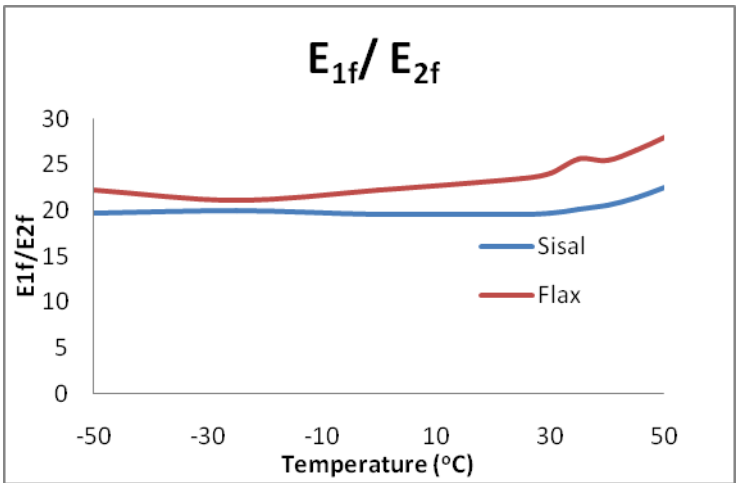

Figure 3: Ratio of E1f Vs E2f for Flax and Sisal

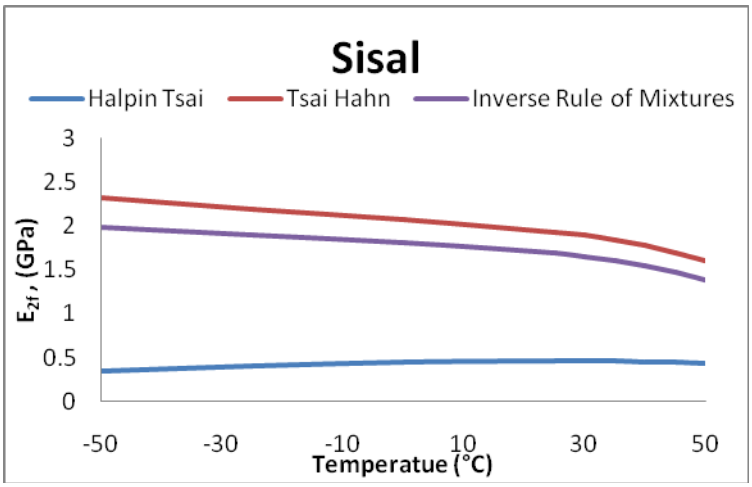

Figure 4: Comparison of models to determine the transverse fibre modulus for Sisal

7.2 Thermal Expansion Anisotropy

Figures 5 and 6 illustrate the thermal expansion coefficient of the sisal and flax fibre in the longitudinal α_{1f} and the transverse α_{2f} direction respectively.

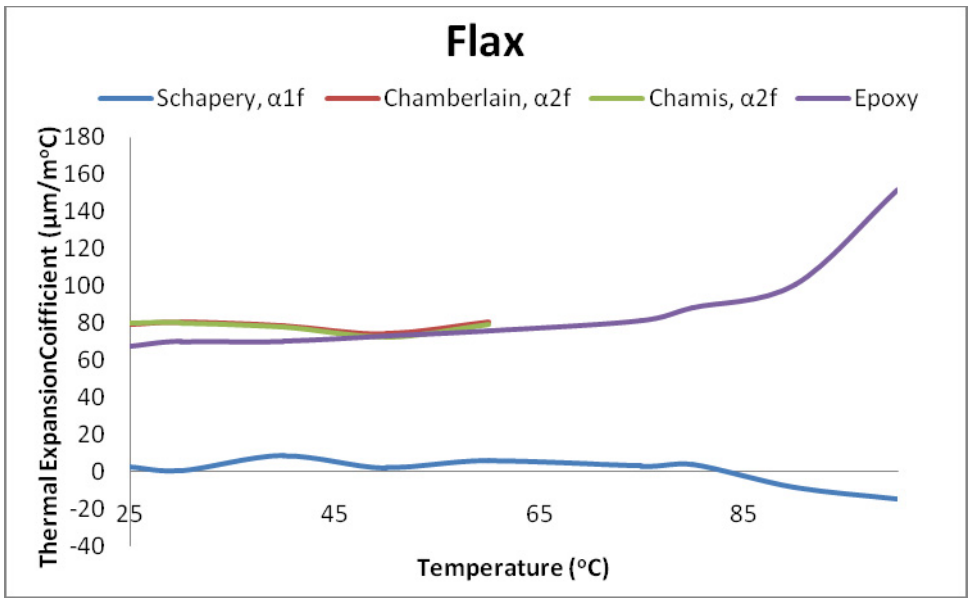

Figure 3: Flax fibre coefficient of Thermal expansion (CTE)

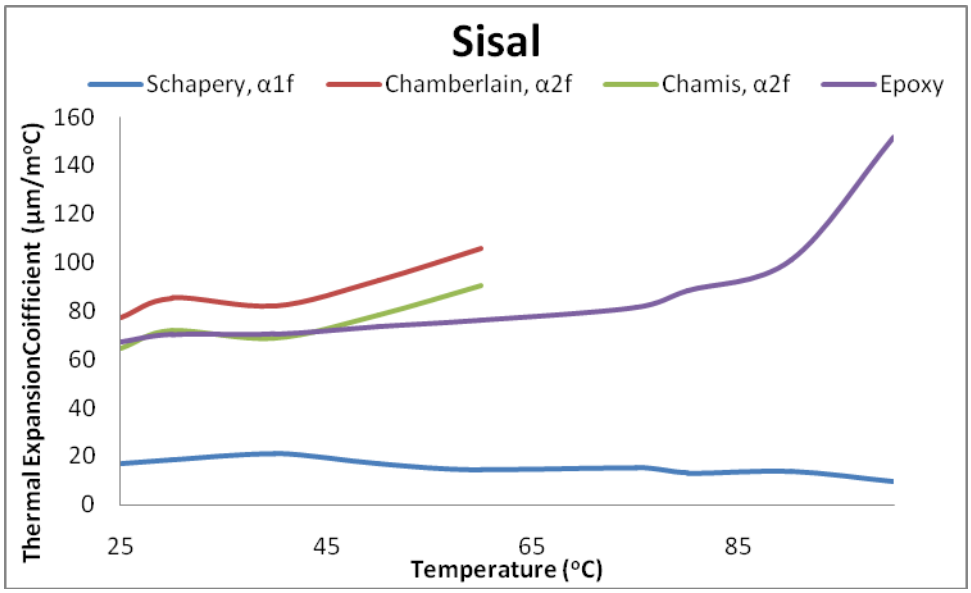

Figure 4: Sisal fibre coefficient of Thermal expansion (CTE)

It can be seen from the graphs that the micromechanical and semi-empirical models for α_{2f} are in good agreement with each other. The models used for finding the coefficient of thermal expansion can be found and discussed further by Karadeniz and Kumlumtas [12].

The flax and sisal longitudinal CTE is approximately 2.8µm/mºC and 16.8µm/mºC respectively at room temperature (25ºC). This behaviour is quite common as Cichocki and Thomason [7] reported that jute fibre longitudinal CTE was low at room temperature. Whereas the transverse CTE of the flax and sisal fibre is 79.8 µm/mºC and 70.8 µm/mºC which is completely different to the longitudinal CTE of the natural fibres.The transverse coefficient of thermal expansion for flax and sisal is very close to the CTE of the resin.

Summary of the micromechanical and semi empirical model of the Sisal and Flax are shown in Table 1 and 2.

	Sisal					**Flax**				
	-50°C	**-25°C**	**0°C**	**25°C**	**50°C**	**-50°C**	**-25°C**	**0°C**	**25°C**	**50°C**
E1f (GPa)	30.4	29.9	28.3	26.6	25.6	35.4	34.30	33.38	32.42	31.65
E2f (GPa)	1.6	1.5	1.5	1.4	1.2	1.6	1.6	1.5	1.4	1.1
G_{12f} (GPa)	1.16	0.80	0.85	0.82	0.99	5.6	4.4	3.8	5.0	1.3
α_{1f} (µm/m°C)				16.8	16.9				2.8	2.1
α_{2f} (µm/m°C)				70.8	84.9				79.8	80.5

Table 1: Thermoelastic Properties of Sisal Fibre

8 Conclusions

The thermoelastic behaviour of untreated flax and sisal composites has been investigated and quantified in this study. The offaxis properties of the unidirectional natural fibre composites were determined through a number of mechanical and thermo-mechanical experiments and were then incorporated into numerous micromechanical and semi empirical models to estimate the thermoelastic properties of the flax and sisal. The longitudinal and transverse properties of the fibres found to be significantly different, highlighting the highly anisotropic structure of the natural fibres. The longitudinal E_{1f} and transverse E_{2f} modulus of flax at room temperature (25ºC) is ~ 32.4 GPa and ~ 1.4 GPa respectively. The longitudinal E_{1f} and transverse E_{2f} modulus of sisal at room temperature (25ºC) is ~ 26.6 GPa and ~ 1.4 GPa respectively. The shear modulus, G_{12f} of both fibres is low with sisal 0.8GPa (25°C) and flax 5GPa (25°C). The coefficient of thermal expansion of the fibres exhibit low values in the longitudinal direction and high values in the transverse direction. The results of the study highlight flax and sisal having highly anisotropic properties. It is therefore likely that the use of only longitudinal fibre properties in composite mechanical property modelling will lead to inaccurate predictions of composite performance.

9 References

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