MODELING THE INFLUENCE OF ENVIRONMENTAL RELATIVE HUMIDITY ON THE TENSILE PROPERTIES OF UNITARIAN HEMP FIBRES

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Abstract

In view of the production of high performance composites reinforced with plant-based fibres, it is essential to account for the high variability and the dependence of the fibre's mechanical properties to moisture conditions in the design tools and models.
This study proposes several empirical formula with probability distribution functions determined using experimental data sets to predict the evolution and the scattering of the tensile properties of elementary hemp fibres as a function of environmental relative humidity. In the present work, we also compare the experimentally determined median values of elastic modulus to the predictions of state-of-the-art micromechanical models. The comparison shows that the available models are unable to predict the tensile modulus as a function of humidity in good agreement with experimental measurements. This discrepancy is due to the lack of knowledge of the ultrastructure and arrangement of the cell wall components, their interaction with water and their evolution under mechanical loading and by this way to the relatively far-reaching hypotheses formulated to derive the micromechanical model.

1 Introduction

In recent years, composite industries and researchers have shown a keen interest in eco-friendly materials such natural fibres and biopolymers. With growing environmental awareness, plant based fibres represent, in particular, an interesting alternative to man-made fibres in composite applications, and can be competitive with glass fibres. However, before such fibres can be used for the reinforcement of organic matrices in high performance applications, an accurate understanding of their micro-mechanical behaviour and the development of theoretical tools able to integrate the large scattering of the mechanical properties are required. Unlike classical man-made fibres, the behaviour of plant-based fibres depends strongly on their humidity. Unfortunately, only a limited number of studies have been conducted on the influence of water on the mechanical behaviour of natural fibres [1-6] and only a few models are able to predict their hygroelastic behaviour [7-9].
Using a large set of experimental data obtained using tensile tests on elementary hemp fibres, the purpose of this work is to establish an empirical model to predict the tensile properties and their scattering as a function of humidity and evaluate the relevance of the predictions of state-of-the-art micromechanical models.

2 Experimental data

Isolated elementary fibres of hemp (*Cannabis sativa* L.) were tested in tension at a controlled temperature of approximately 23°C and several values of relative humidity (RH) between 10 and 85%. A total of 60 elementary fibres were tested for each RH level. The apparent Young's modulus, ultimate tensile strengths and strain to failure were determined individually and a statistical approach was used to analyse the influence of the environmental RH on the tensile properties. The apparent Young's modulus was computed from the last linear section of the stress-strain curve. The data, their analysis and interpretation at the ultrastructural level are synthesised in a previous paper [6]. The main conclusions are presented just below.

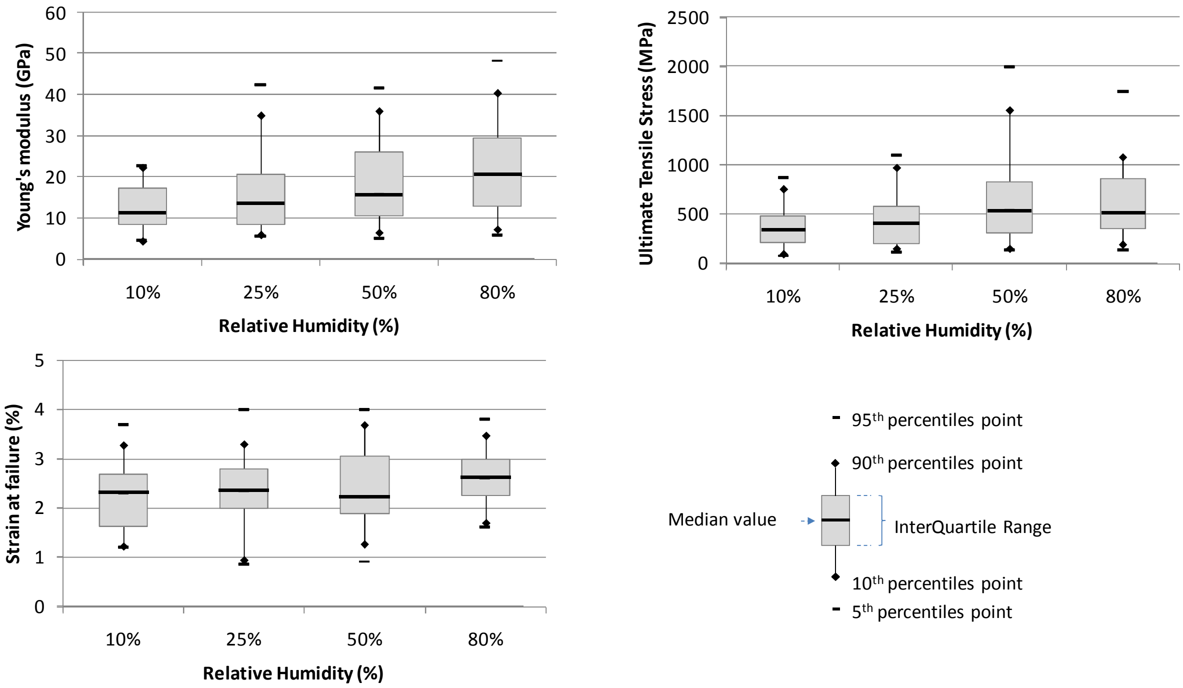

Figure 1: Variation in apparent Young's modulus, Ultimate Tensile Stress (UTS) and strain at failure of isolated elementary hemp fibres as a function of the environmental relative humidity [6]

E-modulus. Contrary to all expectation, an increase in the mean value of the apparent E-modulus of approximately 0.114 GPa/%RH was observed between 10% and 80% of relative humidity. As shown on Fig. 1, using the median value, the apparent E-modulus increases up to a certain point, corresponding to a relative humidity ranging between 50% and 70%, as the humidity exposure is increased, with a slope of approximately 0.105 GPa/%RH. Beyond this point, the increase in E-modulus seems to speed up with a rate of about 0.168 GPa/%RH.

UTS. Drying of the fibre appears to significantly lower its strength. An increase in UTS up to 50-60% RH, and a stabilization or a slight decrease in UTS beyond this threshold are observed. This evolution is particularly apparent when considering the upper bound of the experimental points (95^{th} percentile points, Fig. 1).

Strain at failure. Considering the mean value, the strain at failure increases with RH (from 2.34% to 2.67% between 10% and 80% of RH). This conclusion has to be restrained because the evolution of the median value (Fig. 1) shows that this difference is not really statistically significant.

3 Empirical model for the prediction of the tensile properties of hemp fibres and their scattering as a function of RH

To consider the highly skewed distribution of the tensile properties, several families of distributions were tested (normal, lognormal, exponential, gamma, beta and Weibull) using

the EXTREMES® 1.1 software. The Anderson-Darling and Cramer-Von Mises tests were used for each family to estimate the best parameters of the distribution law.
Gamma (G(α,β)), Weibull (W(η,β)) and lognormal (Logn(m,σ)) distributions are found to be more realistic and are preferred to the other distributions. These distributions are two parameters family of continous probability distributions. For the gamma and Weibull distributions, θ and η are scale parameters and α and β shape parameters. For the lognormal distribution, m and σ are the mean and standard deviation.

$$G(\alpha,\theta): f(x) = x^{\alpha-1} e^{-\frac{x}{\theta}} \frac{1}{\theta^{\alpha}\Gamma(\alpha)} \quad (1)$$

where the gamma function is defined as : $\Gamma(\alpha) = \int_0^{\infty} t^{\alpha-1} e^{-t} dt$ (2)

$$W(\eta,\beta): f(x) = 1 - e^{-\left(\frac{x}{\eta}\right)^{\beta}} \quad (3)$$

$$Logn(m,\sigma): f(x) = \frac{1}{x\sigma\sqrt{2\pi}} e^{\left(-\frac{(\ln(x)-m)^2}{2\sigma^2}\right)} \quad (4)$$

Several criterion of distances were used to evaluate the fit and the validity of the prediction. The three functions were found to be relevant to predict the UTS distribution. Only the gamma distribution was acceptable for the Young's modulus and the strain at failure. Figure 2 shows the distribution of the apparent E-modulus, UTS and strain at failure at 50% of RH and the best fit using the three distribution laws.

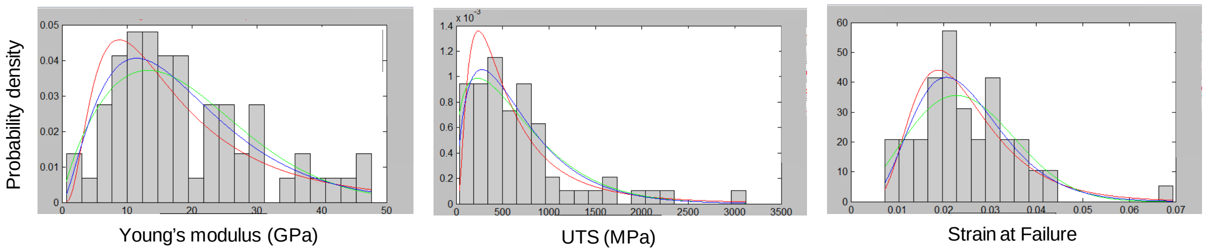

Figure 2. Tensile properties distribution for a relative humidity of 50% (red line: lognormal law, blue line: gamma law, green line: Weibull law)

Table 1 gives the values of the identified parameters for the gamma distribution and figure 3 plots the distributions in a 3D graph. X_L is the lower bound and X_U the upper bound of the distribution (with X being E, σ_R or ε_R). The 5th percentile and 95th percentile points are preferred to the extreme points to define these bounds.

Tensile properties	Distribution parameters	RH = 10%	RH = 25%	RH = 50%	RH = 80%
E [GPa]	α_E	3.28	2.25	2.53	2.46
	θ_E	4.18	8.12	7.52	9.23
	E_L	5	5	5	5
	E_U	23	42	42	48
σ_R [MPa]	α_σ	2.33	1.94	1.69	2.38
	θ_σ	175.5	250.3	406	273.5
	σ_L	70	110	130	130
	σ_U	865	1100	2000	1750
ε_R [%]	α_ε	8.48	7	5.81	15.33
	θ_ε	$2.76\ 10^{-3}$	$3.36\ 10^{-3}$	$4.3\ 10^{-3}$	$1.74\ 10^{-3}$
	ε_L	1.2	0.85	0.9	1.6
	ε_U	3.7	4	4	3.8

Table 1. Parameter values of the gamma distribution of the tensile properties at 4 values of RH

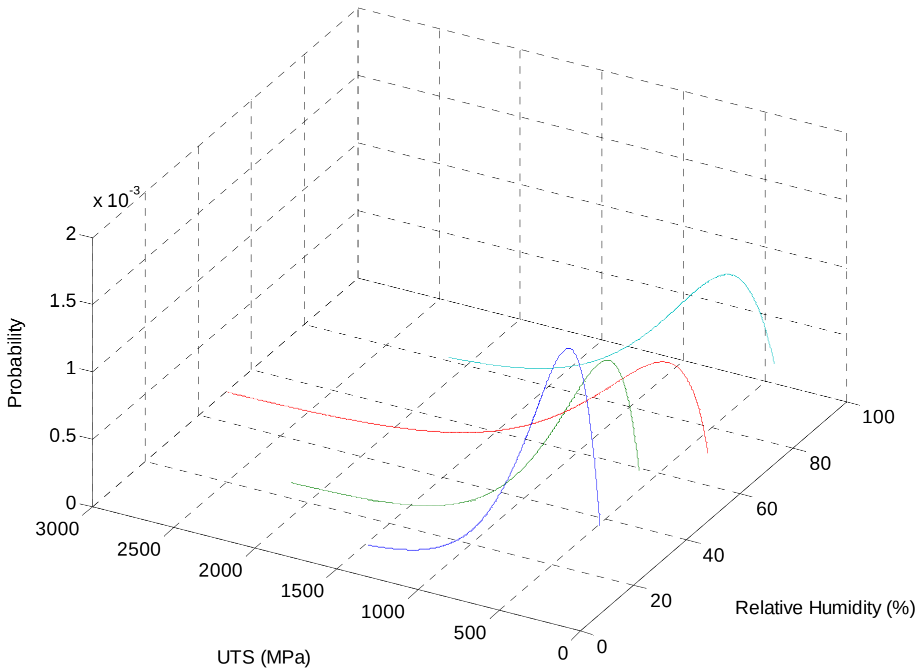

Figure 3. Gamma distribution of the UTS at the four values of tested relative humidity

The evolution of the UTS' distribution function can be expressed as a function of the relative humidity. The same approach was used with E and ε_R, only the results obtained for UTS are presented in this paper. Figure 4.a. shows for example that the median values of UTS can be fitted using a polynomial regression.

Considering the results of van Voorn *et al.* [4], showing for flax fibres a maximum strength at 70% RH, with a strength decreasing when the RH was either increased or decreased with respect to this point, the identified regression with a maximum at approximately 65% seems relevant. Nevertheless to confirm the validity of this regression, additional tests have to be performed at 65% of RH. The above equations (Eq. 5-8) describe the evolution of the gamma distribution parameters and of lower and uppers bounds.

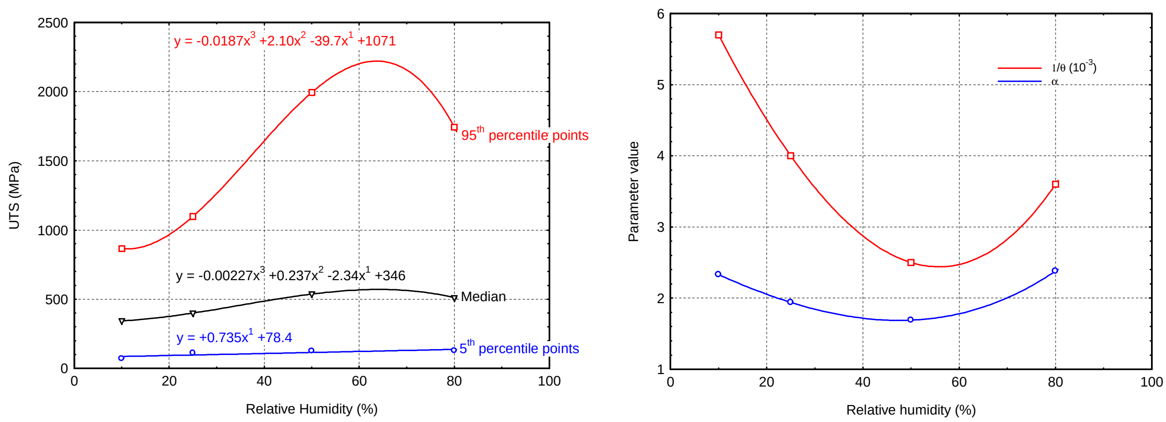

Figure 4. Evolution of the median, parameters, lower and upper bounds of the UTS' gamma distribution as a function of RH

$$\alpha_\sigma = 2.857rh^3 + 1.571rh^2 - 3.429rh + 2.654 \tag{5}$$

$$\frac{1}{\theta_\sigma} = (6.061rh^3 + 8.182rh^2 - 14{,}788rh + 7.091) \times 10^{-3} \tag{6}$$

$$\sigma_L = 0.735rh + 78.4 \tag{7}$$

$$\sigma_U = -0.019rh^3 + 2.099rh^2 - 39.687rh + 1071 \tag{8}$$

with $rh \in [10{,}80]$

It is also very often noted in literature that the mechanical properties, and more precisely the tensile strength and rigidity of such fibres, are highly dependent on their diameter. If the origins of the diameter-dependence of the apparent E-modulus still remain partly unresolved [10], the strength of natural fibres appears to be in agreement with Griffith's theory, developed in the field of the mechanics of fracture in brittle materials. So, we propose here to fit the distribution of the UTS as a function of the fibre diameter, as proposed by Virk [11], using a lognormal function and also as a function of humidity. Figure 5 shows the fibre strength decreases with increasing fibre diameter, for the four values of RH. In agreement with the work of Virk *et al*. [12] on jute fibres, we confirm that the relationship between the fibre strength and the fibre diameter show a logarithmic trend. The logarithmic red line on figure 5b describes the general variation σ(d), identified using the median value of the diameter clusters.

$$\sigma(d) = \sigma_0 - m\ln(d)) \tag{9}$$

The logarithmic evolution is also apparent when considering the upper bound of the experimental points, while the lower bound describes an almost constant dwell (black lines on Fig. 5.b).

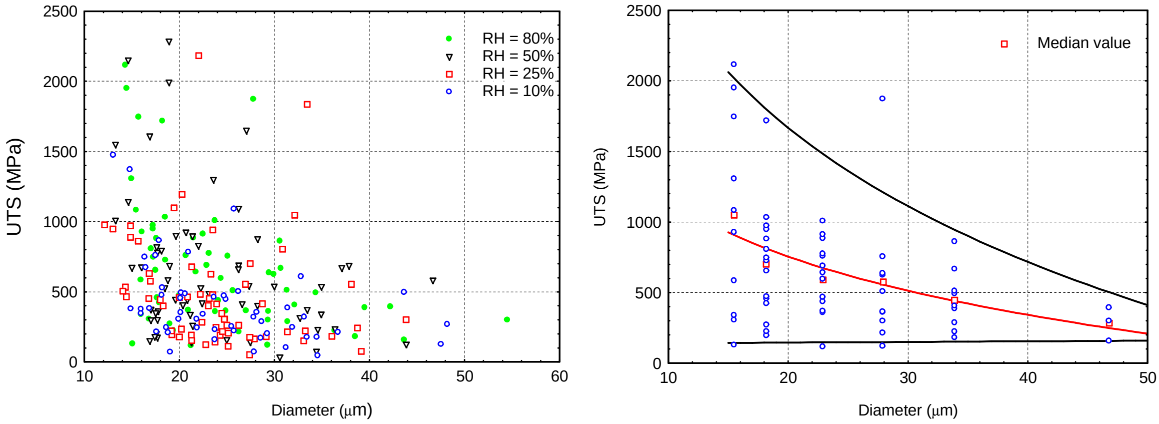

Figure 5. a. UTS as a function of the fibre diameter for the different values of RH. **b.** Evolution of the lognormal distribution of UTS as a function of fibre diameter at 80% of RH.

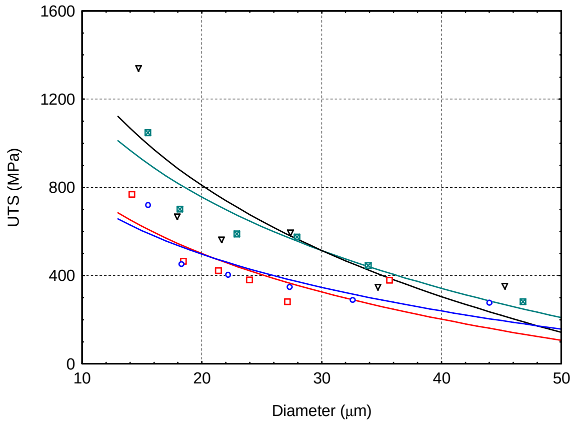

Figure 6. Comparison of the logarithmic regression for each RH (blue line: 10%, red line: 25%, black line: 50%, grey line: 80%)

The logarithmic function can be expressed as a function of the fibre diameter and of RH (Eq. 10). The values of the regression parameters are synthesised in table 2.

$$\sigma(d,rh)=\sigma_0(rh)-m(rh)\ln(d) \qquad (10)$$

RH [%]	σ_0 [MPa]	m	r^2
10	1610	370	0.76
25	1785	430	0.65
50	2990	725	0.7
80	2540	595	0.88

Table 2. Values of the regression parameters

To take into account the overestimation of the fibre cross-section area made by the diameter method (determination of the area considering the fibre as a perfect cylinder without lumen), the UTS can be corrected using a factor F_c. Virk [11] and Thomason *et al.* [13] suggested a value of approximately 1.5 for F_c. Eq. [6] can be written as followed:

$$\sigma(d,rh)=F_c(\sigma_0(rh)-m(rh)\ln(d)) \qquad (9)$$

This factor could also take into account the variation of the cross-section area due to the possible difference in humidity encountered when the diameter is measured and when the tensile test is performed.

4 Modelling the hygroelastic behaviour of hemp fibres using micromechanical model

Over the last decades several models of the structure and ultrastructure of elementary fibres have been attempted in order to make mathematical or numerical predictions of the mechanical properties and behaviour of fibres stemming from wood [7-9,14-15] and annual plants [10,16-18]. In the present study, we propose to use a 3D state-of the-art model for three-phase composites, together with an analytic approach to its solution, to investigate the influence of environmental RH on the elastic modulus of hemp fibres. This model, similar to the Marklund-Varna's model [8] is widely described in a previous paper [10]. This one was improved to take into account the hygro-expansion behaviour, using the extension proposed by Neagu and Gamstedt [7]. Humidity is considered as a swelling and softening factor for the hygroscopic components (hemicelluloses and lignin). The values of the hygroexpansion coefficients of the components and the evolution of the elastic properties as a function of relative humidity are determined using literature data and assumptions as proposed by Marklund [9].

The longitudinal elastic modulus of the fibre computed using this model is compared to experimental data collected on a range of relative humidity from 10% to 85%. The experimental E-modulus was corrected using a factor of 1.50, as suggested by Virk [11] and Thomason *et al*. [13], to account for the overestimation of the fibre cross-section area by the diameter method.

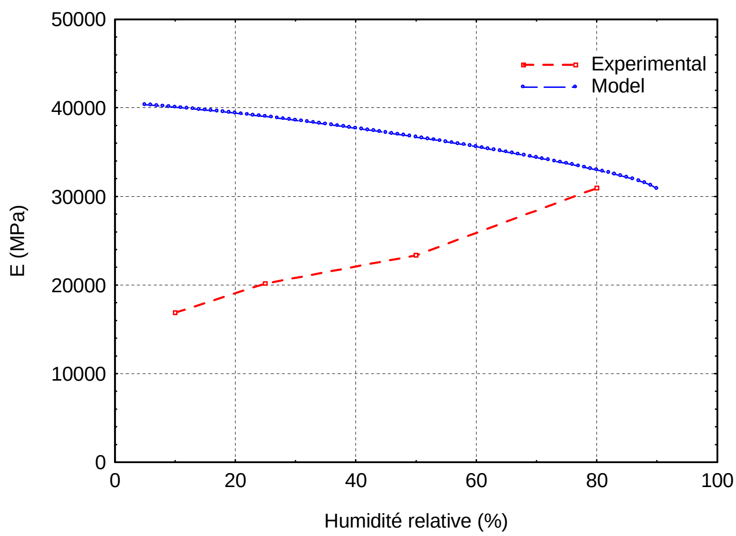

Figure 7. Comparison of the elastic modulus computed from the hygro-elastic model and obtained from experiments (median values).

Fig. 7 clearly shows the inability of the hygro-elastic model to predict the mechanical behaviour of elementary hemp fibres. If the prediction is in a pretty good agreement for the highest values of RH, a huge gap is observed for the lowest ones. Moreover, the model cannot predict the increase in E-modulus as the humidity exposure increases. It must be said that the upturn in apparent Young's modulus as the RH increases is quite surprising for these natural fibres derived from annual plants, considering the well-known plasticizing effect of water on heterogeneous amorphous phases in materials. In addition to this softening effect, the rearrangement of matrix molecules, accompanied by the absorption of bounded water, could enhance the flexibility of the polymer network and under axial mechanical loading, the cellulose microfibrils, or more exactly the crystalline cellulose, could be able to creep in the relaxed amorphous components and to realign up about the fibre axis, and then increase the fibre stiffness. To precisely predict the tensile behaviour of fibres, models have to integrate these ultrastrucural changes and at least take into account the MFA evolution according to tensile stress.

5 Conclusion

This study proposes several empirical formula based on a stochastic approach to predict the evolution of the tensile properties of elementary hemp fibres and their scattering as a function of humidity. This work also emphasis the inability of the state-of-the-art hygro-elastic models to predict the experimentally observed behaviour and the need to integrate the changes in the fibre ultrastructure and macromolecular arrangement under hygro-mechanical loading, such as microfibrils reorientation.

References

[1] Davies G.C., Bruce D.M. *Effect of Environmental Relative Humidity and Damage on the Tensile Properties of Flax and Nettle Fibers.* Text Res J, 68(9), pp. 623-629 (1998).

[2] Symington M.C., Banks W.M., West O.D., Pethrick R.A. *Tensile testing of cellulose based natural fibers for structural composite applications.* J Comp Mater, 43(9), pp. 1083-1108 (2009).

[3] Baley C., Morvan C., Grohens Y. *Influence of the Absorbed Water on the Tensile Strength of Flax fibers.* Macromol Symp, 222, pp. 195-201(2005).

[4] van Voorn B., Smit H.H.G., Sinke R.J., de Klerk B. *Natural fibre reinforced sheet moulding compound.* Compos: Part A, 32, pp. 1271-1279 (2001).

[5] Stamboulis A., Baillie C.A., Peijs T. *Effects of environmental conditions on mechanical and physical properties of flax fibers.* Compos: Part A, 32, pp. 1105-1115 (2001).

[6] Placet V., Cisse O., Boubakar L. *Influence of environmental relative humidity on the tensile and rotational behaviour of hemp fibres.* J Mater Sci, 47, pp. 3435-3446 (2012).

[7] Neagu R.C., Gamstedt E.K. *Modelling of effects of ultrastructural morphology of the hygroelastic properties of wood fibres.* J Mater Sci ,42, pp. 10254-10274 (2007).

[8] Marklund E., Varna J. *Modeling the effect of helical fiber structure on wood fiber composite elastic properties.* Appl Compos Mater ,16, pp. 245-262 (2009).

[9] Marklund E. *Modeling the mechanical performance of natural fiber composites.* Doctoral thesis, 2007.

[10] Placet V., Trivaudey F., Cisse O., Guicheret-Retel V., Boubakar L. *Diameter dependence of the apparent tensile modulus of hemp fibres: a morphological, structural or ultrastructural effect?* Compos: Part A, 43, pp. 275-287(2012).

[11] Virk A.S. *Numerical models for natural fibre composites with stochastic properties*, PhD thesis, 115 pp. (2010).

[12] Virk A.S., Hall W., Summurscales J. *Failure strain as the key design criterion for fracture of natural fibre composites.* Compos Sci Technol, 70, pp. 995-999 (2010).

[13] Thomason J.L., Carruthers J., Kelly J., Johnson G. *Fibre cross-section determination and variability in sisal and flax and its effects on fibre performance characterization*. Compos Sci Technol, 71, pp. 1008-1015 (2011).

[14] Gamstedt E.K., WilhelmssonD., Neagu R.C., Bardage S. *Finite element Modelling of Tensile Tests of Geometrically Well-Characterized Single Wood Fibres* in “Proceeding of *Workshop on Single Fiber Testing and Modelling”*, Stockholm, (2009).

[15] Sedighi-Gilani M., Navi P. *Experimental observations and micromechanical modeling of successive-damaging phenomenon in wood cells’ tensile behavior.* Wood Sci Technol, 41, pp. 69-85(2007).

[16] Baley C. *Analysis of the flax fibres tensile behaviour and analysis of the tensile stiffness increase.* Compos: Part A, 33, pp. 939-948(2002).

[17] Gassan J., Chate A., Bledzki A.K. *Calculation of elastic properties of natural fibers.* J Mater Sci, 36, pp. 3715-3720 (2001).

[18] Nilsson T., Gustafsson P.J. *Influence of dislocations and plasticity on the tensile behaviour of flax and hemp fibres.* Compos: Part A, 38, pp. 1722-1728(2007).