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# Nonlinear Viscoelastic Viscoplastic Material Model Including Stiffness Degradation for Hemp/Lignin Composites

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## Abstract

In repeating tensile tests with increasing maximum strain for every loading cycle the hemp/lignin composites clearly showed a nonlinear behavior and hysteresis loops in loading and unloading. The explanation for this behavior is the inherent viscoelastic nature for this type of material, but also noticeable stiffness degradation with increasing strain level. Creep tests performed at different stress levels revealed a nonlinear viscoelastic response and after recovery viscoplastic strain was detected for high stress levels. It is demonstrated that Schapery's model is suitable to model nonlinear viscoelasticity whereas viscoplastic strain may be described by a nonlinear functional presented by Zapas and Crissman. In a creep test this functional leads to a power law with respect to time and stress.

In order to include stiffness reduction due to damage Schapery's model has been modified by incorporating a maximum strain-state dependent function reflecting the elastic modulus reduction with increasing strain measured in tensile tests. A generalized incremental model of the constitutive equation for viscoelastic case has been used to validate the developed material model in a linear stress controlled loading and unloading ramp. The model successfully

describes the main features for the investigated material and shows good agreement with test data within the considered stress range.

## **Keywords**

Polymer-matrix composites; Creep; Non-linear behavior; Modeling; Viscoelasticity

## **1. Introduction**

Plant fibers as reinforcement in composites have received much attention over the recent years [1-4]. Natural fibers, such as hemp, flax, jute and sisal are renewable and biodegradable cellulosic materials which offer relatively good specific mechanical properties. Drawbacks are high moisture absorption and poor adhesion with non-polar polymer matrices. Natural fiber composites with thermoplastic matrix are extensively used these days in the automotive industry where the fiber acts mainly as filler material in non-structural interior panels. Natural fiber composites for structural purposes do exist, but then usually with oil-based synthetic thermoset matrices. The future goal will be to develop environmental friendly high performance composites which are recyclable and come from renewable resources. A complete biodegradable system may be obtained if the matrix material also comes from a renewable resource. Examples of such matrix materials are lignophenolics, starch and polylactic acid (PLA). Some of these systems are showing encouraging results. For example, Oksman et al. [5] and Bodros et al. [6] have reported that flax fiber composites with PLA matrix outperform both flax/polypropylene and glass/polypropylene composites in terms of stiffness and strength.

Lignin is a complex non-crystalline aromatic macromolecule readily extracted in vast quantities from the paper industry. Studies have suggested that it is possible to replace part of phenol by lignin in phenolic thermoset matrices without loss of mechanical properties [7].

However, there seems to be little work done on mechanical performance of the hemp/lignin composite system in particular.

A very important feature of natural fiber composites is that the mechanical properties of both fibers and the polymer matrix are time dependent. Therefore, natural fiber composites experience complex time dependent stress-strain behavior with loading rate effects and hysteresis loops. This behavior is due to viscoelastic effects of both constituents and may also include micro damage evolution resulting in stiffness degradation and development of irreversible viscoplastic strains. Mechanical properties of natural fiber composites have been studied previously by several authors. For example, composites made from wood fibers and thermoset matrices have been studied [8,9]. Flax/polypropylene composites were characterized in terms of viscoelastic behavior in [10].

Tensile loading – unloading tests to different maxima performed on the hemp/lignin composites showed large increase in size of the hysteresis loops, permanent strains after unloading and stiffness reduction. Hence, the composite has to be described as a nonlinear viscoelastic viscoplastic material that also experience stiffness degradation.

A general thermodynamically consistent theory of nonlinear viscoelastic and nonlinear viscoplastic materials was developed by Schapery [11] and it has been used in simulations by several authors [12,13]. In an earlier study [14] Lou and Schapery presented a constitutive equation for nonlinear viscoelasticity in uniaxial loading. The model contains three stress dependent functions which characterize the nonlinearity. Actually these functions also depend on temperature and humidity [11], but under fixed environmental conditions they are functions of stress only [14]. A methodology to determine the nonlinearity parameters for materials which obey power law time dependence was described. However, not all materials obey the linear viscoelastic power law and a better fit to experimental data is often achieved if the viscoelastic creep compliance in form of Prony series is used [10,15]. In the range of

linear viscoelasticity and in fixed environmental conditions the stress dependent functions are equal to 1 and the data reduction scheme is significantly simplified. The optimal set of experiments needed to determine the stress dependent functions in the material model and development of reliable methodology for data reduction is still a debatable issue.

The development of viscoplastic strains will be described by a nonlinear functional presented by Zapas and Crissman [16].

The objective of the presented paper is to identify the material model for hemp/lignin composites which accounts for the observed nonlinear viscoelasticity, viscoplasticity and damage. The experimental part includes (i) determination of the damage related stiffness degradation with strain in tensile loading –unloading tests and incorporating the obtained maximum strain dependent degradation function in the material model., (ii) analysis of the viscoelastic and viscoplastic properties of this material in creep tests at several stress levels and measuring of the permanent strains after strain recovery .

The developed model will be validated in a test with a linear stress controlled loading and unloading ramp using an incremental form of the constitutive equation.

This paper address the modeling of macroscale material properties and thus details regarding material microarchitecture and morphology are not pertinent to the outcome in this work. It will be shown that the material can be characterized using only a few specimens and that the stress dependent nonlinearity functions may be expressed via simple polynomial functions.

## **2. Theory**

### **2.1. Constitutive model**

Lou and Schapery [14] presented a general nonlinear constitutive equation of viscoelasticity in the case of uniaxial loading. The same constitutive equation with an additional term for viscoplastic strain accumulation  $\varepsilon_{pl}(t, \sigma)$  was used previously by Marklund et al. [10] where flax/polypropylene composites were characterized using different forms of the viscoelastic creep compliance. No stiffness degradation was detected in [10] and thus nonlinear viscoelasticity and viscoplasticity were the mechanisms responsible for the observed behavior.

In the present study however, tensile tests revealed that the hemp/lignin specimens do indeed experience stiffness degradation. The constitutive equation in this case has therefore been modified by incorporation of a maximum strain-state dependent function  $d(\varepsilon_{\max})$  which reflects the elastic modulus reduction with increasing strain.

$$\varepsilon = d(\varepsilon_{\max}) \cdot \left( \varepsilon_0 + g_1 \int_0^t \Delta S(\psi - \psi') \frac{d(g_2 \sigma)}{d\tau} d\tau + \varepsilon_{pl}(t, \sigma) \right) \quad (1)$$

In Eq. (1) integration is over “reduced time” according to,

$$\psi = \int_0^t \frac{dt'}{a_\sigma} \quad \text{and} \quad \psi' = \int_0^\tau \frac{dt'}{a_\sigma} \quad (2)$$

$\varepsilon_0$  represents the initial strain which may be nonlinear with respect to stress.  $\Delta S(\psi)$  is the transient component of the linear viscoelastic creep compliance.  $g_1$  and  $g_2$  are stress dependent material properties.  $a_\sigma$  is the shift factor, which in fixed conditions is a function of stress only. For sufficiently small stresses  $g_1 = g_2 = a_\sigma = 1$ , and thus Eq. (1) turns into the strain-stress relationship for linear viscoelastic viscoplastic materials. In the thermodynamic analysis presented by Schapery [11] it was shown that the viscoelastic creep compliance does not depend on the applied stress level and it may therefore be determined using loads in the linear region. Viscoelastic creep compliance in the form of Prony series was obtained,

$$\Delta S(\psi) = \sum_m C_m \left( 1 - \exp\left(-\frac{\psi}{\tau_m}\right) \right) \quad (3)$$

$C_m$  are constants and  $\tau_m$  are called retardation times. The retardation times are chosen arbitrary, but the highest  $\tau_m$  should at least cover the time for the conducted creep test. A good approximation to experimental data may be achieved if the retardation times are spread uniformly over the logarithmic time scale, typically with a factor of ten between them.

In a creep test the stress is constant until some time instant  $t_1$  whereby the stress is removed and the recovery period begins according to  $\sigma = \sigma[H(t) - H(t - t_1)]$  where  $H(t)$  is the Heaviside step function. The expression (1) may therefore be divided into creep strain and recovery strain in a creep test. Together with the creep compliance from Eq. (3) we obtain the following form of creep strain and recovery strain respectively:

$$\varepsilon_c = d(\varepsilon_{\max}) \cdot \left( \varepsilon_0 + g_1 g_2 \sigma \sum_m C_m \left( 1 - \exp\left(-\frac{t}{a_\sigma \tau_m}\right) \right) + \varepsilon_{pl}(t, \sigma) \right) \quad (4)$$

$$\varepsilon_r = d(\varepsilon_{\max}) \cdot \left( g_2 \sigma \sum_m C_m \left( 1 - \exp\left(-\frac{t_1}{a_\sigma \tau_m}\right) \right) \exp\left(-\frac{t - t_1}{\tau_m}\right) + \varepsilon_{pl}(t_1, \sigma) \right) \quad (5)$$

## 2.2. Viscoplastic strain

Assuming that the viscoplastic strain may be described via a nonlinear functional presented by Zapas and Crissman [16] we have,

$$\varepsilon_{pl} = C_{pl} \left\{ \int_0^t \sigma(\tau)^M d\tau \right\}^m \quad (6)$$

$C_{pl}$ ,  $M$  and  $m$  are constants and must be determined experimentally. The following discussion is a short summary of the procedure needed for the parameter identification which is described in detail in [10,15]. First the *time dependence* of viscoplastic strains is determined by performing creep tests at a fixed stress level so that integration of Eq. (6) is trivial. After



strain recovery the remaining irreversible strain corresponding to the loading period is measured. Several creep tests with different lengths are performed and the developed viscoplastic strains are summed. The viscoplastic strain after  $k$  steps of creep loading at the *same stress level*  $\sigma_0$  will be,

$$\varepsilon_{pl}^{1+2+\dots+k} = C_{pl} \sigma_0^{M \cdot m} (t_1 + t_2 + \dots + t_k)^m \quad (7)$$

The development of viscoplastic strains at fixed stress should thus follow a power law in time with coefficient  $B = C_{pl} \sigma_0^{M \cdot m}$  and constant  $m$  which are determined as the best fit in logarithmic axes. Furthermore, only one specimen is necessary to obtain the time dependence of viscoplastic strains at a certain fixed stress level.

The *stress dependence* of viscoplastic strains has to be obtained performing creep tests of the same length at several stress levels. In result  $B$  is obtained as a function of stress and the best fit in logarithmic axes yields the required constants  $C_{pl}$  and  $M$ .

### 2.3. Incremental form of the constitutive equation

In structural analysis with nonuniform and complex stress state the material model has to be implemented in FE codes which require an incremental form of Eq. (1). Substitution of Eq. (3) in (1) and integration gives,

$$\varepsilon(t) = d(\varepsilon_{\max}) \cdot \left( \varepsilon_0(\sigma) + g_1(\sigma) g_2(\sigma) \sigma \sum_m C_m - g_1(\sigma) \sum_m \varepsilon^m(\psi) + \varepsilon_{pl}(\sigma, t) \right) \quad (8)$$

where

$$\varepsilon^m = \int_0^\psi C_m e^{-\frac{\psi - \psi'}{\tau_m}} \frac{d[g_2(\sigma) \sigma]}{d\psi'} d\psi' \quad (9)$$

The integral in Eq. (9) may be calculated in time instant  $t_k$  using the previously determined value at time  $t_{k-1}$  where  $t_k = t_{k-1} + \Delta t$ . Equation (2) gives the relation between the time increment and  $\Delta\psi$  according to,

$$\Delta\psi = \frac{1}{a_\sigma} \Delta t \quad \text{and} \quad \psi_{k+1} = \psi_k + \Delta\psi \quad (10)$$

The recursive expression for Eq. (9) becomes,

$$\varepsilon^m(t_k) = e^{-\frac{\Delta\psi}{\tau_m}} \varepsilon^m(t_{k-1}) + C_m \left( 1 - e^{-\frac{\Delta\psi}{\tau_m}} \right) R(t_{k-1}) \tau_m \quad (11)$$

In (11)

$$R(t_{k-1}) = \frac{d[g_2(\sigma)\sigma]}{d\sigma} \Big|_{\sigma_{k-1}} \cdot a_\sigma \Big|_{\sigma_{k-1}} \cdot \frac{d\sigma}{dt} \Big|_{t_{k-1}} \quad (12)$$

The routine for simulation of  $\varepsilon(t)$  is as follows; (i) choose the time increment  $\Delta t$ ; (ii) for the time instant  $t_{k-1}$  find  $\Delta\psi$ ,  $R(t_{k-1})$  and calculate all  $\varepsilon^m(t_k)$  using  $\varepsilon^m(t_{k-1})$  from the previous step (they are zero in the 0-step); (iii) now use Eq. (8) to find  $\varepsilon(t)$  and then repeat the steps.

### 3. Experimental

Lignin is a very brittle material at RT and therefore the composites were manufactured containing a large fraction of plasticizer. In addition to this flame retardant was also added. Weight fractions were hemp fiber 30%, flame retardant 20%, plasticizer 30% and finally the matrix material lignin 20%. Further detail regarding constituent morphology is irrelevant for the model development and thus omitted in the following discussion.

The reduction in elastic modulus was measured using an Instron 4411 tensile testing machine with a 5 kN load cell. Strain was measured by an Instron 2630-100 series extensometer with 50 mm gauge length. Cross-head speed was set to 5mm/min. All tests were performed at RT and with relative humidity of 25 – 35%. The stiffness reduction was

measured repeatedly applying a load-unload ramp with increasing maximum to introduce damage followed by low stress load-unload ramp (after each cycle) to measure the elastic modulus. Due to viscoelastic effects the specimens after introducing damage had to recover for some short period of time before the elastic modulus was measured. The specimens were not removed from the grips during the recovery. The maximum strain in the first cycle was 0.2% with increment of approximately 0.1% in each following cycle until failure. In the non-damaging stiffness determination ramps maximum strain was 0.2%. A few examples of stress-strain curves are shown in Figure 1.

In the data reduction the elastic modulus was estimated in the stress region corresponding to 0.05 and 0.15% in strain for the undamaged specimen. The sampling rate was 20 points per second which gave about 25 points for establishing the modulus. The modulus was calculated for the loading part as well as for the unloading part of the curve and then taken as the average.

The creep tests were performed by hanging of dead-weights and measuring strain with standard extensometers. One specimen was first subjected to 10, 20 and 30 min of creep loading at 9 MPa with following recovery in order to estimate the viscoplastic time dependence at a fixed stress level. The viscoelastic properties were later determined from the same specimen by subjecting it to 1 hour of creep loading at stress levels 9, 6 and 3 MPa respectively. Another specimen was subjected to 10, 20 and 30 min of creep loading at 6 MPa for estimation of the viscoplastic stress dependence. A small degradation in stiffness was detected afterwards which indicates that viscoplastic strains due to damage might have developed. Unfortunately we were unable to measure these very small viscoplastic strains with sufficient accuracy. The limit for viscoplastic strain development was therefore set to 6 MPa. The same specimen was also tested in creep at 10 MPa (10, 20 and 30 min). A third

specimen was tested in creep at 11 MPa and experienced creep rupture after less than 8 min of loading. The fracture surface revealed a dry region and many voids.

#### 4. Results and discussion

##### 4.1. Stiffness degradation

The initial modulus  $E_0$  was determined from the first loading cycle corresponding to 0.20% in strain. The initial Young's modulus was in the range 2.3 – 2.8 GPa. Tensile strength was 14 – 16 MPa. Figure 2 shows the reduced modulus normalized with respect to the initial modulus. No stiffness degradation could be seen for strain values lower than 0.3% and consequently this strain level was therefore set as limit for stiffness reduction. At a strain level of 0.9% the stiffness reduction was roughly 7-8%. The regression line in Figure 2 determines the maximum strain-state dependent function required in Eq. (1),

$$d(\epsilon_{\max}) = \begin{cases} 1 & \text{never loaded above 0.3 \%} \\ \frac{1}{-0.116\epsilon_{\max} + 1.033} & \text{otherwise} \end{cases} \quad (13)$$

Stiffness degradation is the manifestation of damage evolution. In Figure 3 in situ micrographs of poorly impregnated regions in one of the tested hemp/lignin specimens are shown. The load is in horizontal direction. The micrographs give an understanding of the condition of the edge surface. There are many small randomly oriented surface cracks (a) with an opening almost independent on the load. Their contribution to the stiffness reduction is most likely negligible. The large crack (b) opens more with increasing load which means that it is more a volume crack than a surface crack. This indicates that the large crack is one of the contributors to the stiffness reduction.

##### 4.2. Viscoplastic strain

The viscoplastic strains in tension were measured as the remaining recovery strains after 6 times the loading period in creep. The viscoplastic strains accumulated in all steps were summed and plotted against time in log-log scale according to the description in section 2.2. The slope of the trendline was straight which means that the development of viscoplastic strains at fixed stress indeed follows a power law with high accuracy. However, the power law which is a consequence of assumption Eq. (6) states that the exponent  $m$  is constant for all stress levels and certainly that was not true in the present case. At 10 MPa the exponent is higher than at 9 MPa. In light of this result the decision was to use an average value of  $m$  in the forthcoming calculations. Experimental results and model predictions for viscoplastic time dependence at fixed stress using an averaged  $m$  value is shown in Figure 4. The corresponding values of  $B$  are also shown.

The stress dependence for development of viscoplastic strains was obtained by assuming a strain value of 0.001% at 6 MPa in order to have three values of  $B$  (remember that we were unable to measure the very small viscoplastic strains at this stress level and at 11 MPa the specimen experienced creep rupture). Consequently, values  $M = 20.7$  and  $C_{pl} = 1.65 \times 10^{-13}$  (for time in s, stress in MPa and strain in %) were determined.

#### 4.3. Nonlinear viscoelastic model with Prony series

We suggest that each specimen in the viscoelastic analysis is analyzed separately which is a preferable strategy since the data reduction procedure otherwise easily becomes both tedious and impractical and may contain some artificial trends when averages are used.

After recovery in the 9 MPa creep test a small irreversible strain was detected. Thus viscoplastic strains are developing during the test and the creep strain is a sum of viscoelastic and viscoplastic strains. In order to obtain a pure nonlinear viscoelastic response the viscoplastic strains that develop must therefore be subtracted from experimental data.

Following the work in [15] the development of viscoplastic strains in the current creep test is expressed by,

$$\varepsilon_{pl}(t) = \left[ \left( \varepsilon_{pl}^{1+2+\dots+k} \right)^{1/m} \frac{t}{t_k} + \left( \varepsilon_{pl}^{1+2+\dots+(k-1)} \right)^{1/m} \left( 1 - \frac{t}{t_k} \right) \right]^m - \varepsilon_{pl}^{1+2+\dots+(k-1)} \quad (14)$$

$t_k$  is the length of the creep period in question and  $\varepsilon_{pl}^{1+2+\dots+k}$  is the total viscoplastic strain development in  $k$  creep tests.  $m = 0.49$  was determined in the previous section. The specimen had experienced creep loading three times at 9 MPa in the viscoplastic characterization procedure earlier with a total duration of one hour so  $\varepsilon_{pl}^{1+2+3} = 0.043\%$  was known from the previous creep tests. After the 9 MPa creep test in the viscoelastic characterization procedure  $\varepsilon_{pl}^{1+2+3+4} = 0.055\%$  was measured and Eq. (14) could be used calculating the pure nonlinear viscoelastic response for this stress level. No viscoplastic strains developed during the 6- and 3 MPa tests. Tensile tests before and after the viscoplastic characterization procedure showed that the stiffness of the specimen was reduced by approximately 8% and measurements after 9-, 6- and 3 MPa in the viscoelastic characterization creep tests showed no further stiffness degradation. Thus  $d(\varepsilon_{\max}) = 1.08$  was used in the following calculations.

The first step in the determination of the parameters in the nonlinear viscoelastic model is to determine the parameters in the linear viscoelastic range, i.e. to determine  $\tau_m$  and  $C_m$ . The retardation times  $\tau_m$  were chosen to be uniformly spread over the logarithmic time scale and with the highest value covering the time for the creep tests in question. The coefficients  $C_m$  were determined from experimental recovery data for the 3 MPa creep test with the assumption that we are within the region of linear response (all stress dependent nonlinearity functions are thus equal to 1) and the result is shown in Table 1. This procedure, and also the forthcoming calculations of the stress dependent nonlinearity functions were performed using the method of least squares written in MATLAB code.

Next step will be to determine the stress dependent nonlinearity functions from recovery and creep data and the methodology will be the same for all stress levels:  $a_\sigma$  is altered in the *recovery* strain expression (5) in time interval  $t_1 < t < 2t_1$  and for every value of  $a_\sigma$  we obtain the corresponding value of  $g_2$  directly via method of least squares. This procedure is continued until we find the value of  $a_\sigma$  which gives a nice visual fit to experimental data. Equation (4) is then used to find  $\varepsilon_0$  and  $g_1 \cdot g_2$  from experimental *creep* data for the same stress level. When all the nonlinearity parameters have been determined for all stress levels they may be approximated with simple analytical functions. Figure 5a) shows the initial strain  $\varepsilon_0$  as a function of stress and b) the nonlinearity values for the creep tests of 3-, 6- and 9 MPa and their approximations as function of stress.

In order to check whether the approximations lead to acceptable results the model is compared to experimental creep and recovery data as shown in Figure 6. Clearly, the accuracy of the model based on Prony series and polynomial approximation of the stress dependent functions is good for all stress levels in the considered stress range.

The developed model may be used to predict the nonlinear viscoelastic behavior for this type of composite (with similar properties) for any stress from 0-9 MPa. However, we must emphasize that all the viscoelastic time dependences have been obtained from creep tests on one single specimen and so far the model is applied to that particular specimen. It is therefore crucial for the characterization that the specimen is representative for the analyzed material. The specimen was singled out on the basis that its elastic properties were intermediate in this group of specimens. The accuracy of the developed model is best understood comparing the simulated creep curves with other specimens not used in the data reduction and it has been checked that the variation of properties between different specimens is larger than the small deviation shown in Figure 6a).

#### 4.4. Model validation in linear loading and unloading ramp

The developed viscoelastic viscoplastic material model including stiffness degradation was used to simulate the composite behavior in a linear loading and unloading scheme: constant load rate of 0.01 MPa/s up to 9 MPa, unloading with 0.01 MPa/s and finally loading with 0.02 MPa/s up to 13 MPa according to the solid line in Figure 7. The dashed line in Figure 7 shows the shift in time which is necessary when the viscoplastic strains are calculated. According to Eq. (6) the integration is over a continuous function from time  $t = 0$ . However, in this case we have no viscoplastic strains developing for stresses lower than 6 MPa (between  $0 < t < t_1$  and  $t_3 < t < t_4$ ). The stress as a function of time (with time coordinate  $t'$ ) for the different parts of the loading ramp which needs to be integrated is also shown in Figure 7.

The incremental form of the constitutive equation was programmed using MATLAB code following the routine described in section 2.3. A time step of 0.1 sec was used. Smaller steps were not considered since the difference in result from using a time step of 1 sec was less than 1%. The elastic (initial) strain, the viscoelastic- and viscoplastic strains are all calculated separately and then added. Finally the total strain due to stiffness degradation is calculated with the requirement that  $d(\epsilon_{\max}) = 1$  before  $\epsilon(t)$  passes 0.3% the first time and then  $d(\epsilon_{\max}) > 1$  following Eq. (13). Since  $d(\epsilon_{\max})$  is a function of the highest previously known strain state it will increase until  $t_2$  and then have a constant value of  $d(\epsilon(t_2))$  until next time it reaches a higher strain value (around  $t = 2250$  sec and  $\sigma = 8.92$  MPa). Figure 8 shows the result of the simulation. The largest part of the time dependent strain is clearly nonlinear viscoelastic. The viscoplasticity and stiffness reduction do not contribute much to the total strain except in the end of the loading ramp where we have high stresses and actually are very close to rupture for these composites.



The model and its strain response to the linear loading and unloading ramp was also validated by comparing it to experimental data for two specimens, see Figure 9. The accuracy of the model is remarkably good and it captures most features from the experimental curves except for a small discrepancy in the unloading part. The model also seems to predict the behavior of the composites rather well even for the very high stresses prior to rupture which is surprising since we are well outside the region for which it was designed. Figure 10 on the other hand illustrates the problem that might occur if we apply the model to a specimen tested in creep at 11 MPa. The specimen experienced secondary creep and ruptured after only 8 minutes. Clearly the creep mechanisms are different at this high stress level.

## 5. Conclusions

The analysis of time dependent properties for the hemp/lignin composites has been limited to tension case. The behavior in compression can certainly be different. Creep tests performed on the composites showed that they may be described as a nonlinear viscoelastic material for stresses higher than 3 MPa (for this particular time of creep loading). For stresses higher than 6 MPa the material may be described as nonlinear viscoelastic and viscoplastic. The material also showed micro damage evolution which resulted in reduction of elastic modulus for strain levels higher than 0.3%.

The largest part of the time dependent strain is viscoelastic and this material behavior has been modeled using the theory of nonlinear viscoelasticity developed by Schapery. The stress dependent nonlinearity functions in Schapery's expression was successfully described by simple polynomial functions. Prony series was used to describe viscoelastic creep compliance. The viscoplastic strain was described by a nonlinear functional originally presented by Zapas and Crissman. This functional contains three constants which must be determined from experiments. It was found that one of these "constants" was in fact not a

constant, but rather a function of stress. Therefore, this term was approximated using an average value in the considered stress range. A maximum strain-state dependent function reflecting the elastic modulus reduction with increasing strain has also been incorporated in the material model.

The developed model has been validated in a linear loading and unloading ramp using an incremental form of the constitutive equation. The accuracy of the model is remarkably good within the stress range for which it is designed and captures most features of the compared experimental curves. For very high stresses when we are well outside the model stress range (and actually close to rupture) the model description gets inaccurate.

It was shown that the time dependent properties of the material may be characterized using only a few specimens. It was therefore crucial for the characterization procedure that the specimens were representative for the analyzed material.

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### Table Captions

Table 1. Coefficients in Prony series.

m	$\tau_m$ (s)	$C_m$ (%/MPa)
1	1	0.00061
2	10	0.00198
3	100	0.00368
4	1000	0.00437
5	10000	0.00940

### Figure Captions

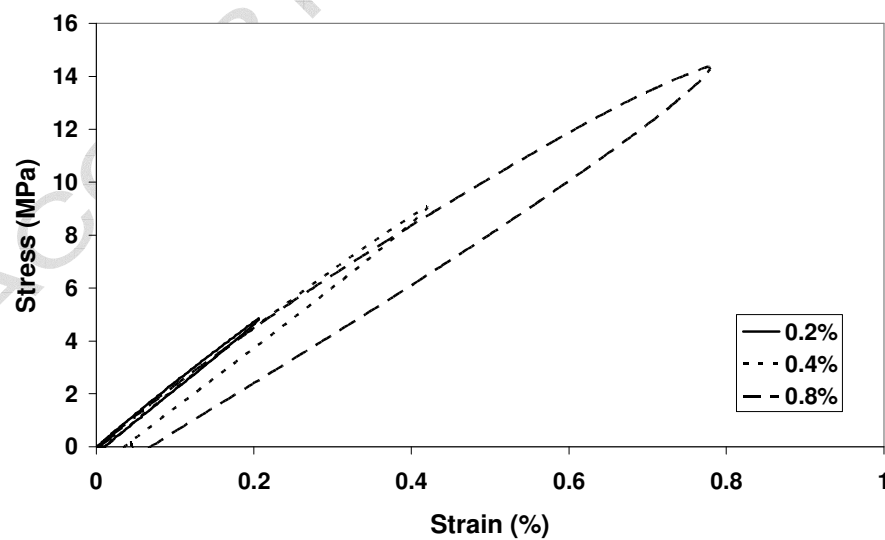


Figure 1. Tensile stress-strain curves.

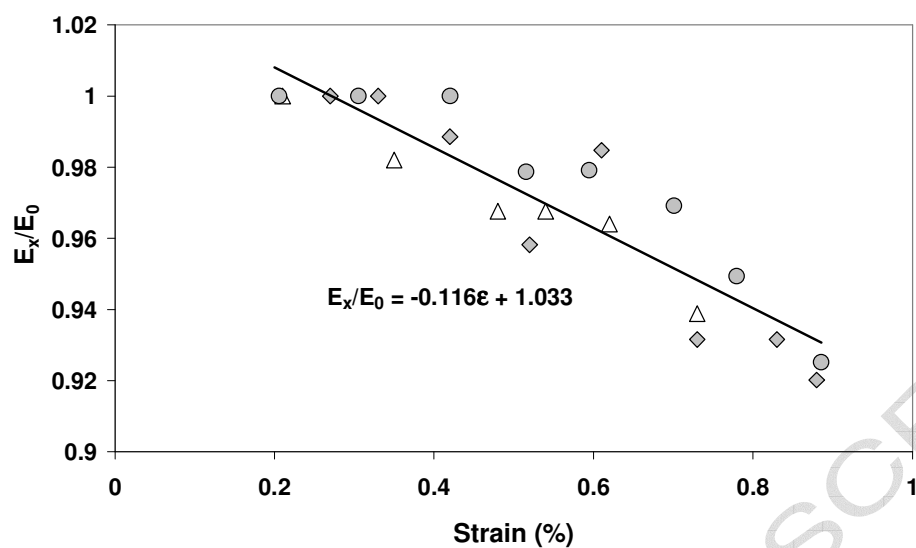
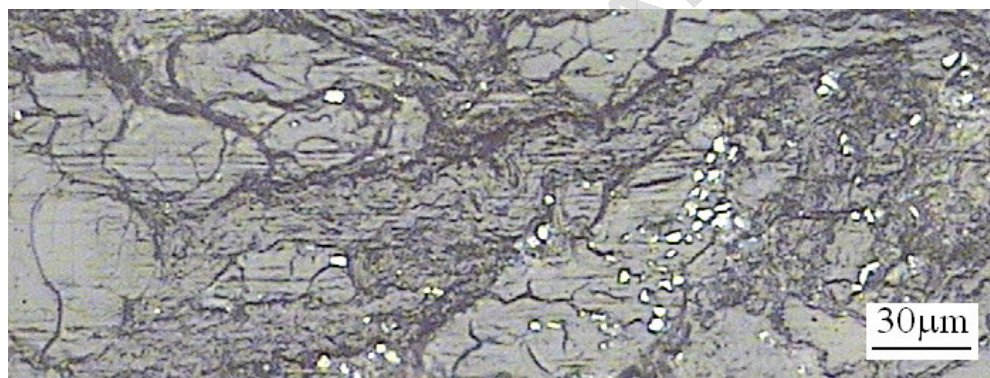
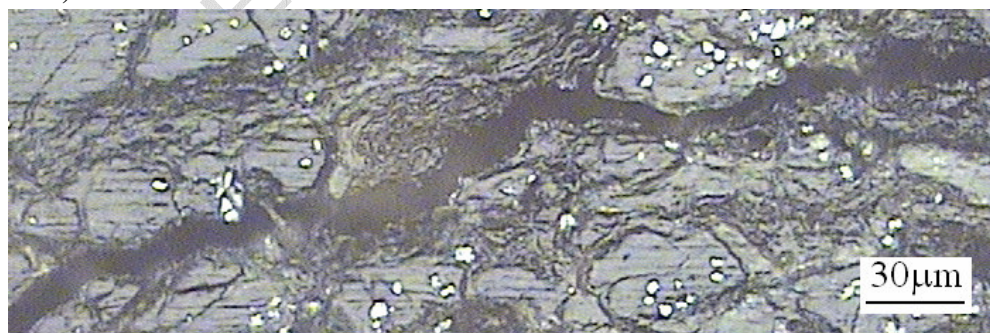


Figure 2. Stiffness reduction with increasing strain for three specimens from tensile tests and regression line that determines the function  $d(\epsilon_{\max})$ .



a)



b)

Figure 3. An in situ hemp/lignin specimen.

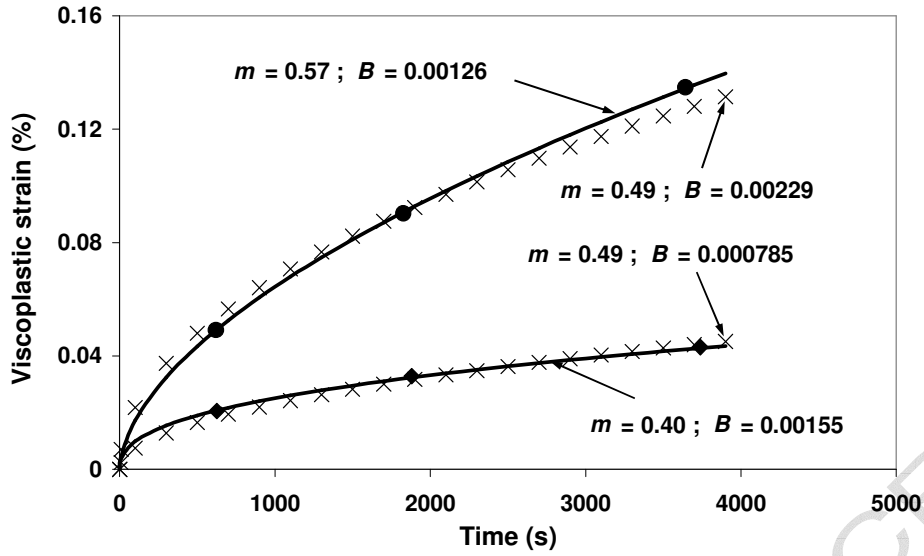
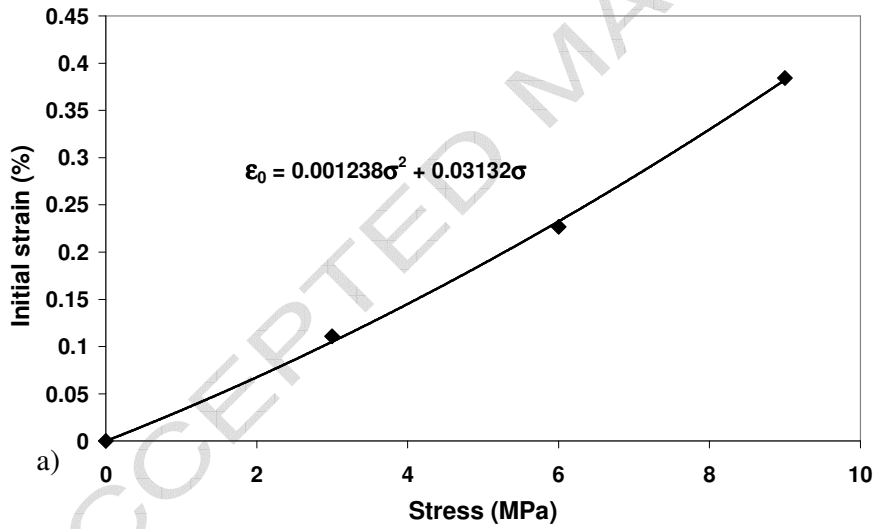


Figure 4. The development of viscoplastic strains at fixed stress levels: Experimental data at 9 MPa (♦), 10 MPa (●) creep tests and model predictions using the averaged  $m$  value (×) for both stress levels compared to "true" model predictions (solid lines).



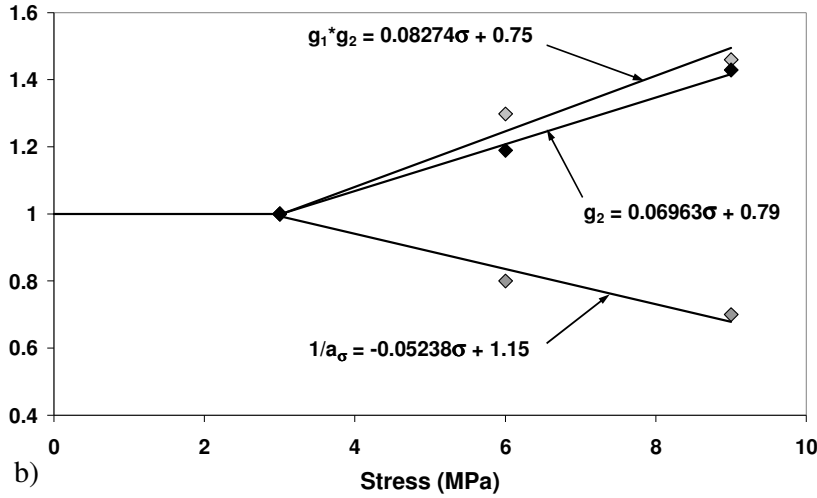


Figure 5. a) Initial strain  $\varepsilon_0$  as a function of stress b) stress dependence of the nonlinearity functions.

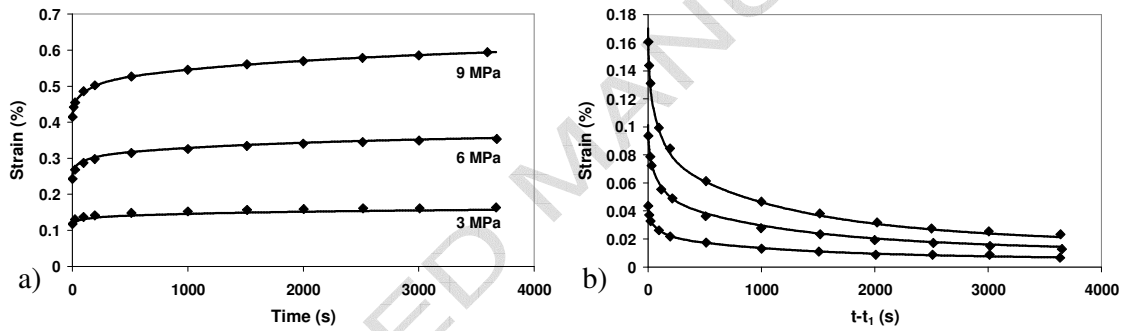


Figure 6. a) Viscoelastic strain response from creep tests, model prediction (solid line) vs. experimental data (dots) b) strain recovery from creep tests.

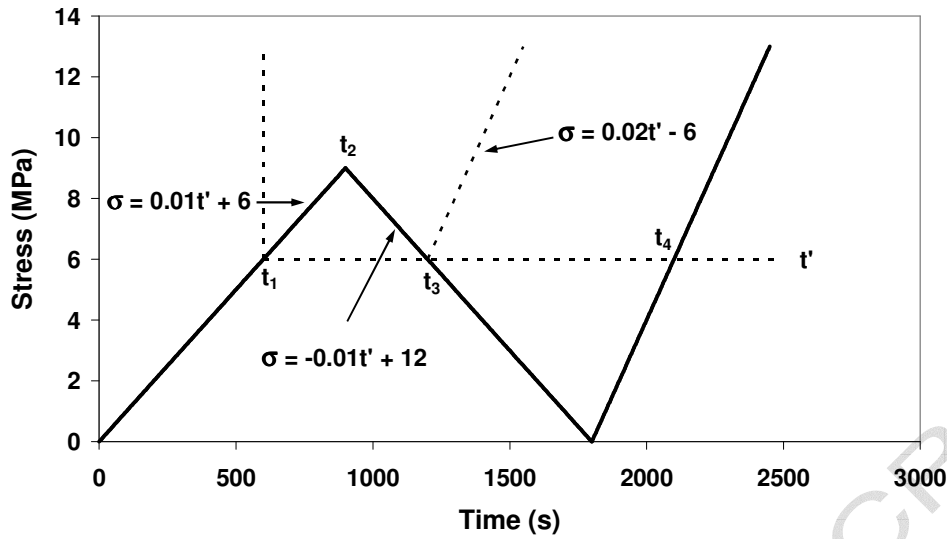


Figure 7. Linear loading and unloading ramps and the required time shift for viscoplastic strain calculation.

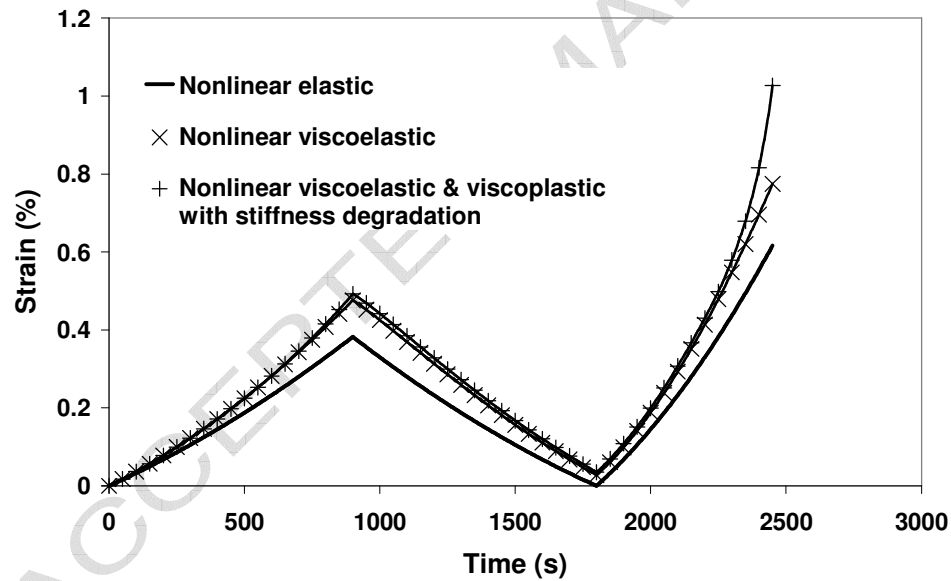


Figure 8. The contribution of nonlinear elastic strain and nonlinear viscoelastic strain compared to total strain (viscoelastic strain, viscoplastic strain and stiffness reduction) in the linear loading and unloading ramp.



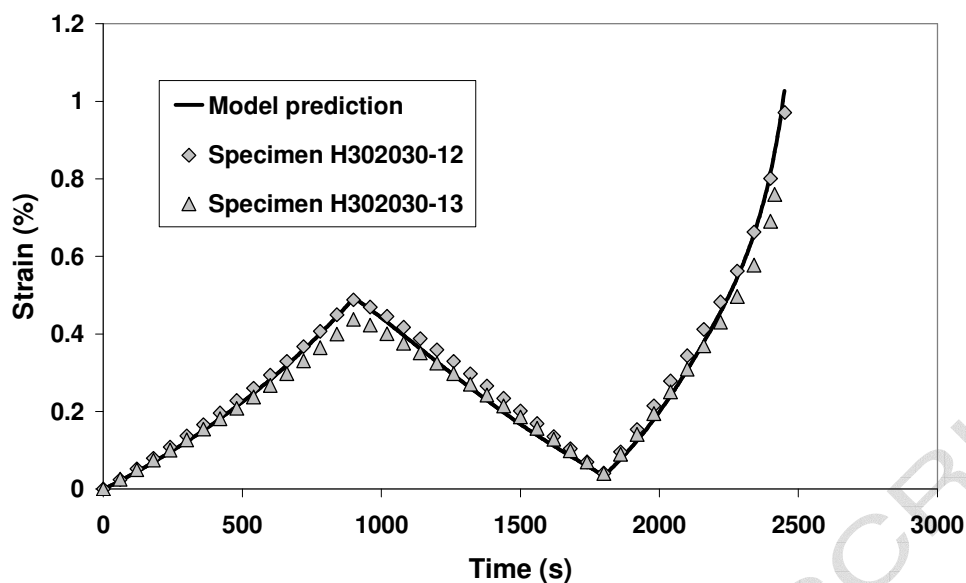


Figure 9. Strain response to the linear loading and unloading ramp, model prediction (solid line) and experimental values for two specimens (dots).

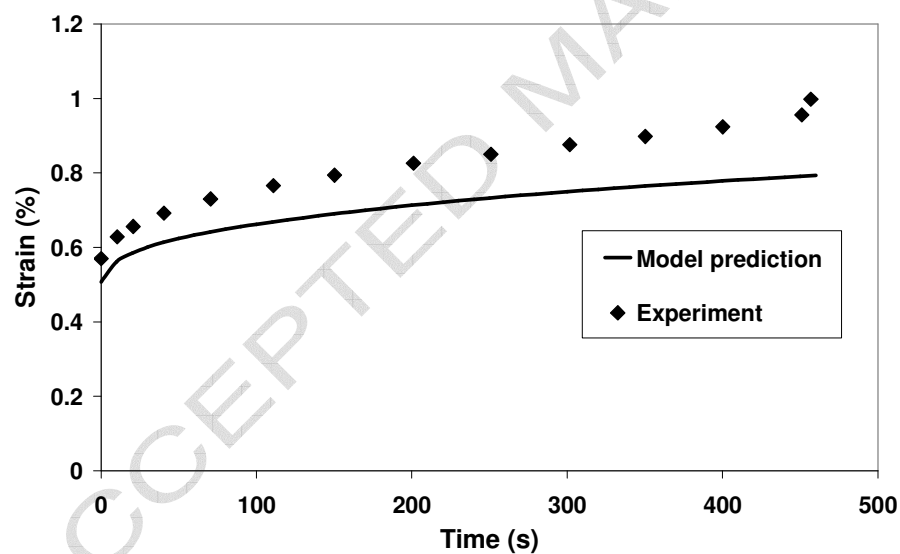


Figure 10. Creep strain at 11 MPa and model prediction.