

## MODELLING OF MECHANICAL BEHAVIOUR OF POLYMERIC COMPOSITES WITH NONLINEAR CONSTITUENTS

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### Abstract

*There is need to model behaviour of time-dependent non-linear material in stress or/and strain controlled experiments. This study explores possibility to apply model in both forms developed by Schapery for viscoelastic materials. Viscoelasticity has been analysed using experimental data from creep and relaxation tests. Incremental simulation procedure, which inverts model, where strains are expressed through stresses, is used to simulate relaxation curves for bio-based polymer. Comparison of viscoelastic parameters obtained from simulated and experimental relaxation curves has been performed.*

### 1. Introduction

Composite materials have shown to exhibit time-dependent mechanical properties. Even more pronounced nonlinearity can be observed in bio-based composites. Therefore there is growing need for models that can predict behaviour of such materials. Nonlinear viscoelastic material model developed by Schapery [1,2] complemented with Zapa's model [3] for viscoplasticity and Varna's [4] model for damage has been successfully used in several studies to simulate stress controlled tests [5-7]. All parameters needed for this model are obtained by performing creep, strain recovery, tensile test as well as stiffness degradation due to damage experiment.

However, there is great need for simulation of strain controlled tests, since most of the codes for numerical structural analysis, analytical micromechanics models (rule of mixture, concentric cylinder assembly model), classical laminate theory requires constitutive model where stresses are expressed as a function of strain and time and also most often experiments are performed in displacement (strain) controlled mode. Similarly to the model described above there is thermodynamics based nonlinear model developed by Schapery where strain is used as an input variable [8]. In order to obtain all parameters needed for this type of model, relaxation tests, where viscoelastic strains are kept constant, must be performed. Since most of the materials have also viscoplastic strain component, these tests can not be performed straight forward as it is in case with creep tests. Another possibility is to rewrite the model, that has stress as a variable, in incremental form and then invert by expressing the stress increment through the increment of the applied strain. However, as analysed by Schapery [8], for non-linear materials the two described formulations (strain and stress controlled) are not exactly invertible and the inaccuracy introduced by inversion cannot be *a priori* estimated.

The long-term objective of this work is to use the inverted incremental models for fibre and resin to develop micromechanics based model for composites. Such model would significantly reduce number of experiments needed to characterize composites with different types of reinforcement, various matrix materials and laminate lay-ups. The current study is initial part of the development of the model for composites and focuses on unreinforced polymer. Viscoelastic parameters are identified using creep data and Schapery's model in stress formulation. Then incremental model is used to simulate relaxation tests ("simulated samples"). The accuracy of the incremental inversion technique is assessed characterizing the same material in relaxation test ("test samples"). The viscoelastic non-linearity parameters, obtained using Schapery's model in strain formulation and relaxation curves for test samples and simulated samples, are compared.

## 2. Material model

The nonlinear viscoelastic strain response in one-dimensional case [1-2] can be described as:

$$\varepsilon(\sigma) = \varepsilon_{el} + g_1 \int_0^t \Delta S(\psi - \psi') \frac{d(g_2 \sigma)}{d\tau} d\tau \quad (1)$$

In (1)  $\varepsilon_{el}$  represents elastic strain in undamaged composite which generally speaking, may be nonlinear function of stress. "Reduced time"  $\psi$  is introduced in (1) as:

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

Parameters  $g_1$ ,  $g_2$  and the shift factor  $a_\sigma$  are stress invariant dependent. They are also affected by temperature and humidity. The transient part of the viscoelastic response is characterized by  $\Delta S(\psi)$  according to the [2] and does not depend on stress; it has a form of Prony series.

$$\Delta S(\psi) = \sum_i C_i \left( 1 - \exp\left(-\frac{\psi}{\tau_i}\right) \right) \quad (3)$$

In (3)  $C_i$  are constants and  $\tau_i$  are retardation times. For some materials a region can be found where  $g_1 = g_2 = a_\sigma = 1$ , and (1) turns into the strain-stress relationship for linear viscoelastic material.

Similarly to stress dependent model described above in one-dimensional case stresses can be expressed through strains [8]:

$$\sigma = \sigma_{el} + h_1 \int_0^t \Delta E(\xi - \xi') \frac{d(h_2 \varepsilon)}{d\tau} d\tau \quad (4)$$

where  $\sigma_{el}$  is the elastic part of the model. The time-dependent part can be described

$$\Delta E(\xi) = -\sum_m E_m \left( 1 - \exp\left(-\frac{\xi}{\tau_m}\right) \right) \quad (5)$$

In (5)  $E_m$  and  $\tau_m$  are stress independent constants. The “reduced time”  $\xi$  in this case is expressed through:

$$\xi = \int_0^t \frac{dt'}{a_\varepsilon} \quad \text{and} \quad \xi' = \int_0^\tau \frac{dt'}{a_\varepsilon} \quad (6)$$

Parameters  $h_1$ ,  $h_2$  and  $a_\varepsilon$  are strain invariant dependent functions. As in case of first model (1)-(3) these parameters are also affected by temperature and humidity, but in fixed environment they are just strain dependent. Within the interval where material has linear viscoelastic behaviour, these parameters can be assumed equal to 1. For linear viscoelastic materials the form (1)–(3) can be inverted to form (4)–(6). For non-linear materials both forms are, generally speaking, [8] not compatible.

### 3. Experimental procedures

#### 3.1. Materials and manufacturing

The epoxidized pine oil based resin (~75% bio-based) EpoBioX (Amroy, Finland) with Amroy CA35Tg curing agent (100:27 weight ratio) is used. Plates were manufactured using resin transfer moulding at room temperature and at low flow speed (cured for 2h at 80°C).

After curing resin plates were cut into rectangular pieces and sample edges were grinded in order to remove defects from cutting and obtain uniform cross-section. Approximate sample dimensions are 160x10x2 mm. Samples were conditioned in desiccator with relative humidity 41%. Mass of samples was constantly monitored in order to ensure saturation of moisture has been reached. If humidity in testing area was lower than 41%, humidifier was used and/or sample was wrapped in thin plastic film.

#### 3.2. Creep and relaxation tensile tests

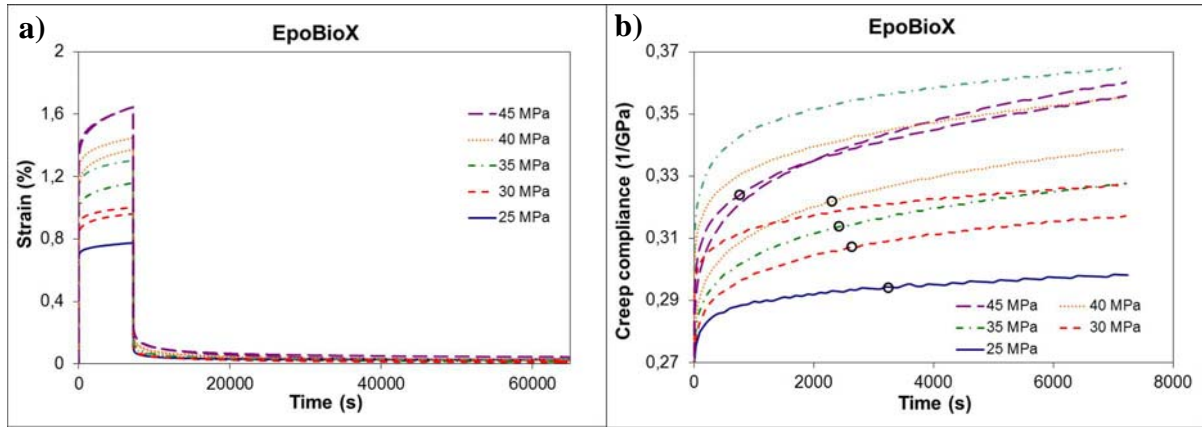
During previous studies it was observed that elastic modulus varies from sample to sample, which significantly influences instant elastic response in creep and relaxation tests. Therefore prior to creep or relaxation tests, elastic modulus of every specimen was measured. Tensile creep tests were performed using creep rig with dead weight. Load was applied instantly and kept for 2h then followed by strain recovery for 16h. Creep and recovery strains were measured using extensometer with gage length of 50 mm. It was observed that after recovery irreversible strains were negligibly small (highest recorded strain was 0.04 %) or not present at all, therefore we can assume that this material in the analysed stress and time range does not exhibit viscoplastic behaviour. Initially for each stress level new sample was used in creep test, but after observing high scatter between experimental results and realizing that the stress-strain response is reversible, one specimen was tested at each stress level.

Since material is purely viscoelastic, relaxation tests where viscoelastic strains are kept constant are simple to perform. Relaxation tests were done on Instron 3366 equipped with 10 kN load cell and pneumatic grips. Standard Instron extensometer 2620-601 (50 mm base) was used to measure longitudinal strain. Sample was loaded with strain rate 3 %/min and close to

target strain level strain rate was decreased to 0.1 %/min to avoid overloading. Duration of relaxation test was 2h. Relaxation tests at several strain levels were performed only on two samples EB-32 and EB-34 with elastic modulus respectively 3.49 GPa and 3.34 GPa.

#### 4. Results and discussions

Experimental results from creep tests are presented in fig. 1a) and viscoelastic creep compliance is shown in fig. 1b). No clear trend can be observed if results from all specimens are pooled together, see fig.1b). However if results from the same sample are analysed separately (see curves marked with circles in fig. 1b) ) it can be concluded that the compliance increases with higher stress level.



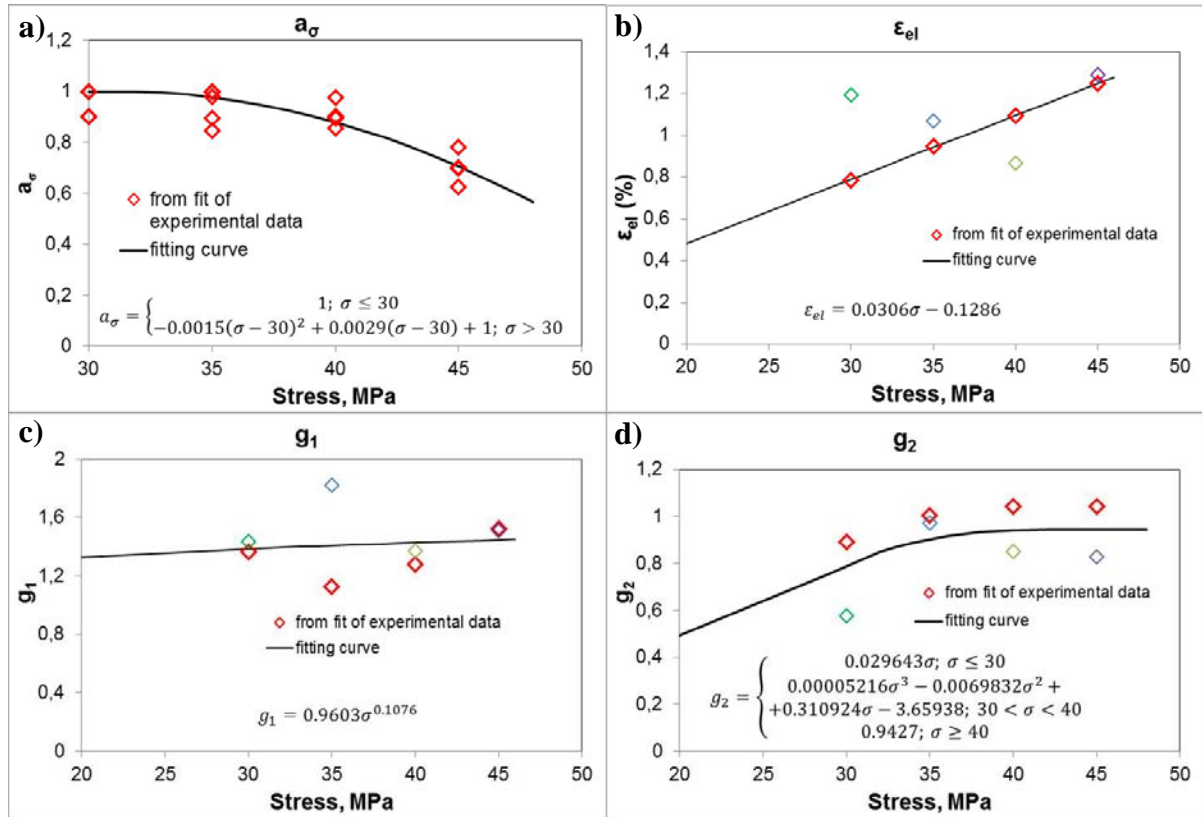
**Figure 1. a)** Creep and recovery strain; **b)** creep compliance (curves from same sample are marked with circle).

In the creep test the stress is applied at  $t = 0$  and is kept constant until time instant  $t_1$ . Then the stress is suddenly removed and the strain recovery period begins. The loading dependence on time can be written as  $\sigma = \sigma[H(t) - H(t - t_1)]$  where  $H(t)$  is the Heaviside step function. The material model eq. (1) may therefore be applied separately to the creep  $t \in [0, t_1]$  and to the strain recovery interval  $t > t_1$ . In [4,6] following expressions were obtained for creep strain  $\varepsilon_{creep}$  and recovery strain  $\varepsilon_{rec}$  (since EpoBioX did not exhibit viscoplasticity, the viscoplastic term has been removed):

$$\varepsilon_{creep} = \varepsilon_0 + g_1 g_2 \sigma \sum_i C_i \left( 1 - \exp\left(-\frac{t}{a_\sigma \tau_i}\right) \right) \quad (7)$$

$$\varepsilon_{rec} = g_2 \sigma \sum_i C_i \left( 1 - \exp\left(-\frac{t}{a_\sigma \tau_i}\right) \right) \exp\left(-\frac{t-t_1}{\tau_i}\right) \quad (8)$$

Eq. (7) and (8) were then used to fit both viscoelastic creep and strain recovery simultaneously using method of least squares. The obtained viscoelastic parameters with their functions are presented in fig. 2. The coefficients in Prony series are given in table 1. It can be seen from fig. 2 b) that data scatter has great influence on fitting results. If only data points from one sample are considered, the fitting curve forms straight line. Parameter  $g_1$  shows small increase, but parameter  $g_2$  is increasing until reaching plateau value of 0.94. Function extrapolation at lower stresses cannot be confirmed due to unreliable strain measurements.



**Figure 2.** a) Shift factor , b) elastic response, c) parameter  $g_1$  and d)  $g_2$  as a function of applied stress. Different colour dots represents different sample (except fig. 1a)).

$\tau_i$ (s)	$C_i$ (%/MPa)
20	$7.5329 \cdot 10^{-4}$
200	$1.2991 \cdot 10^{-3}$
1500	$8.9368 \cdot 10^{-4}$
9000	$3.2590 \cdot 10^{-3}$

**Table 1.** Parameters in Prony series.

All viscoelastic parameters and functions were then used in incremental mode to simulate relaxation test. Simulations were done for both samples (the difference is in elastic response only). Comparison of experimental and modelled curves and relaxation modulus for one sample (EB-32) is presented in fig. 3. The results for second sample (EB-34) are very similar. Relaxation modulus in fig. 3 b) shows that material is linearly viscoelastic until 0.9 % strain and thus at lowest strain level we can assume that  $h_1 = h_2 = a_\epsilon = 1$ .

It was noted that relaxation curves for both test samples have the same shape and they are only shifted in vertical direction (along stress axis). In order to validate this observation, the curves were corrected for different elastic response so that they start at the same initial value. The shifted curves for both test samples at 0.9 % strain level are presented in fig. 4 (it should be noted that the results for other strain levels are very similar). The curves in fig. 4 show that experimental data coincide, whereas curves for simulated samples present different trend depending on which elastic modulus has been used. In simulated relaxation tests stress is decreasing faster if higher modulus is used. This leads to the conclusion that relaxation

according to the incremental model is elastic modulus sensitive, whereas relaxation data are not. This may be one of the consequences of using model (1)-(3) in strain controlled cases.

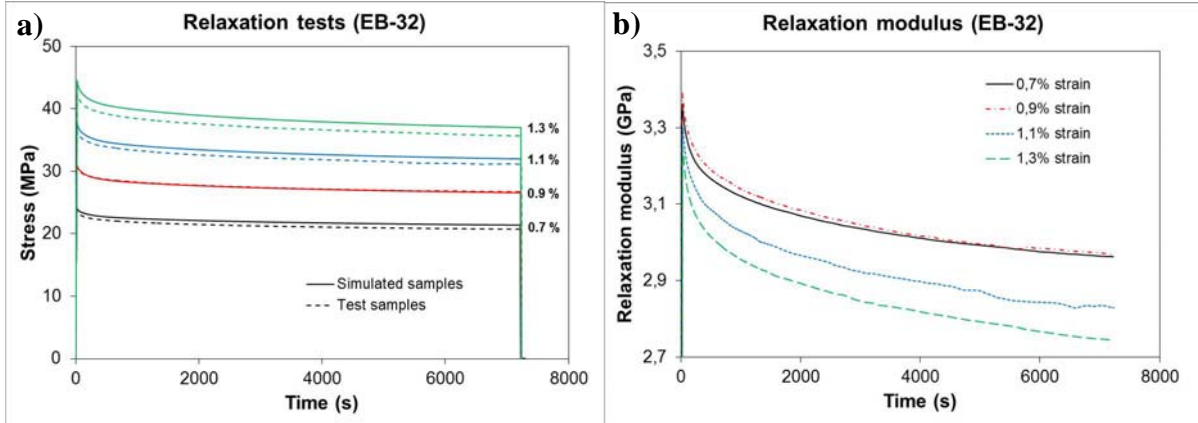


Figure 3. a) Relaxation curves (model and experiment) and b) relaxation modulus (experiment).

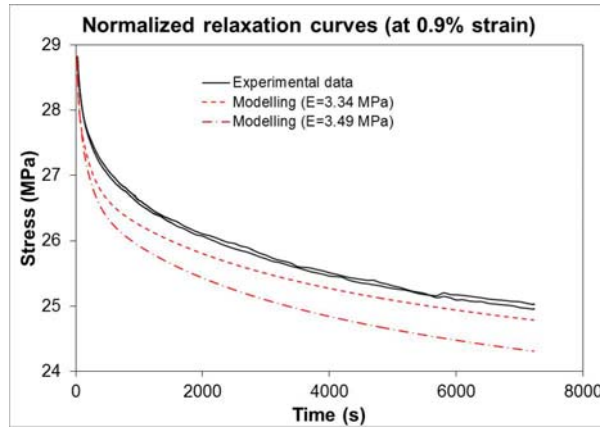


Figure 4. Shifted relaxation curves at strain level 0.9 %.

Model (4)-(6) was then applied to relaxation data for test samples and simulated samples. In relaxation tests constant strain is applied instantly  $t = 0$  and is kept constant until time instant  $t_1$ . Similarly to creep tests it is possible to derive expression for relaxation:

$$\sigma_{relax} = \sigma_{el} + h_1 h_2 \varepsilon \sum_m E_m \left[ 1 - \exp\left(-\frac{t}{a_\varepsilon \tau_m}\right) \right] \quad (9)$$

The applied load can be instantly removed in creep test to follow strain recovery, but in relaxation tests removing applied strain would lead to compressive stresses with unknown nonlinearity parameters and possible bending of the sample. Since this type of test sequence was not used, we could not find separately parameters  $h_1$ ,  $h_2$  and only product of  $h_1 h_2$  can be obtained from relaxation test. One way to resolve this issue would be to perform relaxation test at high stress first and then continue experiment at lower strain level within linear viscoelastic region, where  $h_1 = 1$  thus finding  $h_2$ . It is still uncertain if at the lower strain levels stress measurement would be reliable enough for precise parameter analysis.

Relaxation curves for test samples and for simulated samples were fitted to obtain viscoelastic parameters. At first parameters  $E_m$  and  $\tau_m$  were found at strain 0.7 % (linear viscoelastic

region, so  $h_1 = h_2 = a_\epsilon = 1$ ), and then using these parameters  $a_\epsilon$ ,  $\sigma_{el}$  and product  $h_1 h_2$  were obtained for all other strain levels. It was observed, that fit was much better if parameters  $E_m$  and  $\tau_m$  were chosen individually for each sample. This may indicate that finding nonlinearity parameters sample to sample variation should be respected not only for elastic modulus but also for parameters of linear viscoelasticity. Coefficients of Prony series for viscoelasticity are presented in table 2 and viscoelastic parameters are presented in fig. 5.

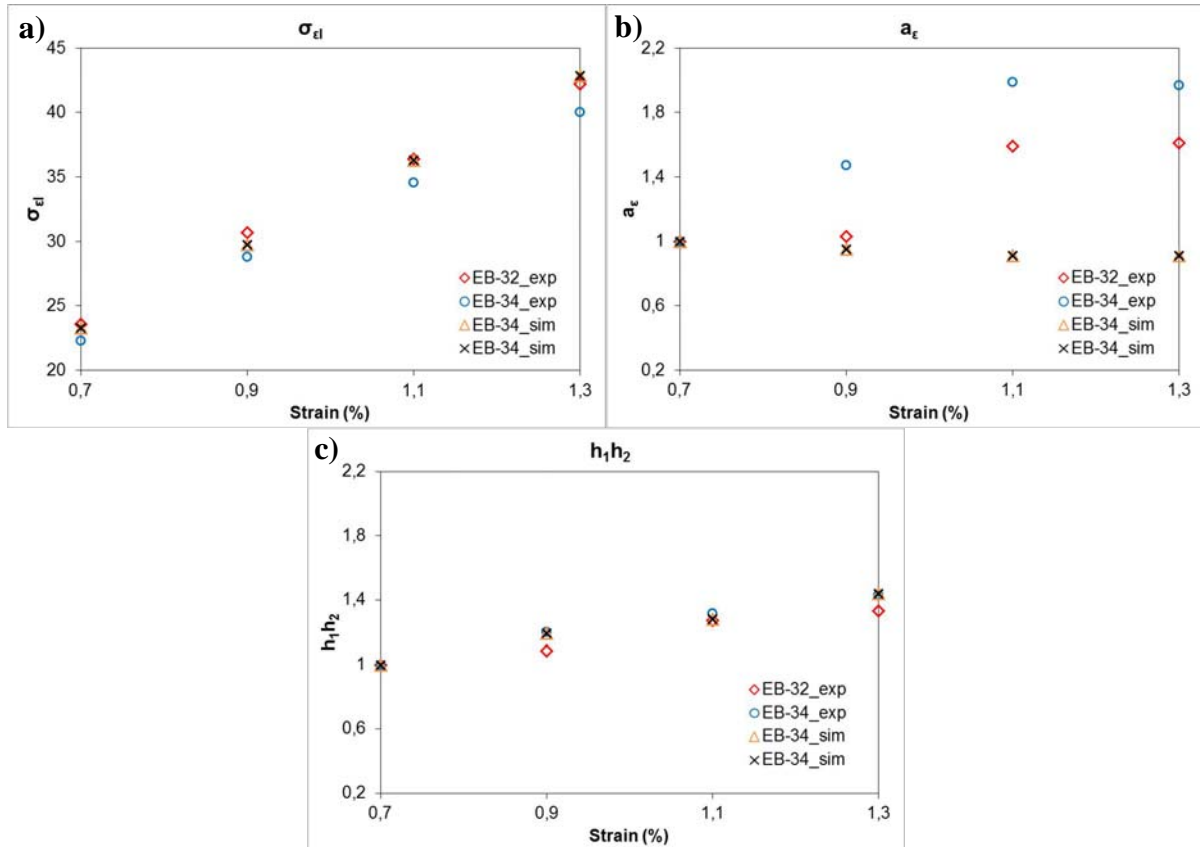


Figure 5. Viscoelastic parameters obtained from relaxation tests: a) elastic response  $\sigma_{el}$ , b) shift factor  $a_\epsilon$  and c) product  $h_1 h_2$ .

Test samples				Simulated samples			
EB-32		EB-34		EB-32		EB-34	
$\tau_m$ (s)	$E_m$ (MPa)	$\tau_m$ (s)	$E_m$ (MPa)	$\tau_m$ (s)	$E_m$ (MPa)	$\tau_m$ (s)	$E_m$ (MPa)
18	551.489	45	834.007	24	830.614	24	723.677
180	1138.818	450	1132.194	240	1277.642	240	1136.068
1200	506.248	3000	485.165	1600	310.835	1600	268.877
3600	2144.034	9000	2882.427	4800	2194.854	4800	1987.11

Table 2. Parameters in Prony series.

Although relaxation modulus showed, that 0.9 % strain belongs to linear viscoelastic region ( $h_1 = h_2 = a_\epsilon = 1$ ), the best fit of relaxation curves was obtained with parameters that are greater than 1. Analysis of results in fig. 5 confirms feasibility of using individual parameters for linear viscoelasticity for each sample when nonlinearity is analysed. It can be seen that nonlinear parameters obtained from test samples coincide or show similar trend. It is also worth noticing that results from simulated samples coincide, moreover, they even agree with

parameters obtained from experimental data with exception of  $a_e$  where results from simulated samples show slow decrease instead of increase.

## 5. Conclusions

Two alternative formulations of non-linear viscoelastic model in terms of stresses or strains were considered in this paper. Schapery showed that these two forms which are uniquely linked for linear viscoelastic materials are generally speaking incompatible for non-linear materials, because only the first terms in expansions are used. In this work the incompatibility and possible inaccuracy was weighted with the convenience of performing creep tests when stress controlled model is developed. Moreover, interpretation of relaxation test is problematic if viscoplastic strains are present. This study attempted to verify that the stress controlled model may be sufficiently accurate for simulating the simplest strain controlled test: stress relaxation test. If proved sufficiently accurate, the simulated relaxation data can be used as an input to identify the material model where strain is the independent variable.

The stress controlled material model was built. It was rewritten in incremental form, inverted and used to simulate the stress relaxation test. To verify the accuracy of the model EpoBioX bio-based polymer was used in creep and strain recovery tests. The simulated relaxation curves are in good agreement with experimental data for the same material with some distinct differences: the simulated relaxation curve is sensitive with respect to the value of the elastic modulus whereas the experimental curves are not. The simulated and the experimental stress relaxation curves were used to identify strain dependent functions in the strain controlled material model. The obtained results show that the results are rather similar: only the “reduced time” related function is slightly different. This conclusion is encouraging, because it suggests that the non-linear stress dependent model can be used to generate relaxation “test” data for identification of strain dependent parameters in the strain dependent model.

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