APPLICATIONS AND LIMITATIONS OF NON-LINEAR VISCOELASTIC MODEL FOR SIMULATION OF BEHAVIOUR OF POLYMER COMPOSITES

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Keywords: Non-linearity, Viscoelasticity, Viscoplasticity, Material model

ABSTRACT

There are two alternative formulation of non-linear viscoelastic model to describe strain and stress controlled tests. Both models for non-linear viscoelastic materials are not compatible, and cannot be directly inverted if so required in certain cases. In order to do it numerical procedures has to be employed. Methodology for simulating nonlinear stress-strain response in iso-strain situations of fiber composites based on properties on constituents is presented.

1 INTRODUCTION

Numbers of studies have demonstrated that polymer composites (e.g. bio-based composites, conventional composites exposed to elevated temperatures) exhibit highly non-linear behaviour [1-8]. It has been shown that Schapery model for non-linear viscoelastic material [6, 9] complemented with Zapas model for viscoplasticity [10] and model accounting for damage [11] can be used to characterize such materials and simulate their behaviour in stress controlled tests. The model parameters in this case are obtained from creep tests.

However, it would be more practical and suitable to simulate displacement (strain) controlled tests. For instance, most of the codes for numerical structural analysis, analytical micromechanical models (e.g. rule of mixtures, concentric cylinder assembly model), classical laminate theory require constitutive model where stresses are expressed as a function of strain and time. Besides, most often experiments are performed in displacement (strain) controlled mode.

Schapery also has developed non-linear viscoelastic model where stress is expressed through strains [12, 13] but because both forms of model are not compatible (exact inversion is possible for linear viscoelastic materials only), non-linearity parameters obtained from creep tests cannot be used in strain controlled model. For example, relaxation modulus has to be obtained in relaxation tests, where viscoelastic part of the strain is kept constant. These tests are very straightforward for materials with no viscopalstic strain component. However, usually during the experiment material has also growing viscoplastic strain component. Keeping in test the applied strain constant the viscoelastic strain is reducing and the measured stress cannot be used to define the strain dependent relaxation modulus [14].

Thus, it is obvious from the reasoning presented above that it would be much more convenient and simpler to use creep tests at least for partial identification of the model.

For the cases when viscoplastic strain is present, material model where strains are expressed through stresses has been rewritten in inverted incremental form [15]. Previous results with simulations of simple stress-strain curves have showed good agreement with experimental data in cases, if it was possible to characterize material in creep at stresses approaching material failure [16].

This study examines capabilities and limitations of this method. Simulations using direct and inverted incremental model are performed with viscoelastic parameters obtained from relaxation and creep tests.

2 MATERIAL MODELS

In order to fully understand and use nonlinear viscoelastic materials, one has to be able to simulate stress and strain controlled tests. In this sections description of different nonlinear viscoelastic models will be presented.

2.1 Stress formulation

The bases for the theory of nonlinear viscoelastic and nonlinear viscoplastic materials employed in this work were developed by Schapery [6, 9]. The constitutive equations in this theory are obtained using expansion of the Gibb's free energy in viscoelasticity related internal state variables and using linear evolution laws with respect to thermodynamic forces. The resulting constitutive equations include many stress invariant dependent material parameters/functions that have to be experimentally identified if general loading case for anisotropic material is considered. In fixed environment (constant temperature and humidity) these functions/parameters are stress dependent only.

Assuming uniaxial tensile loading leads to one-dimensional model in which the nonlinearity is characterized by only three stress invariant dependent functions. The final form of the material model for one-dimensional case, including micro-damage [11], is as follows:

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

The integration is over "reduced time" represented by:

$$\psi = \int_0^t \frac{dt'}{a_\sigma} \qquad \text{and} \qquad \psi' = \int_0^\tau \frac{dt'}{a_\sigma} \tag{2}$$

In (1) ε_0 represents elastic strain in undamaged composite which, generally speaking, may be nonlinear function of stress. The time dependent part of the viscoelastic response (integral in (1)) is characterized by $\Delta S(\psi)$, which is the transient component of the linear viscoelastic creep compliance and according to the theory does not depend on the stress level. The parameters g_1, g_2 are material properties and a_σ is the shift factor, since environmental conditions are fixed, all of them are only stress dependent.

It was shown by Schapery [9] that the linear viscoelastic creep compliance can be expressed in the form of Prony series,

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

In (3) C_i are constants and τ_i are called retardation times.

Within the linear viscoelastic region viscoelastic strains are proportional to the stress level. This region may exist if stresses are sufficiently small and in this case $g_1 = g_2 = a_\sigma = 1$, thus (1) turns into the strain-stress relationship for linear viscoelastic, nonlinear viscoplastic material. The function $d(\sigma_{\max})$ in (1) is introduced as ratio between initial (E_0) and damaged state ($E(\sigma_{\max})$) stiffness:

$$d(\sigma_{\max}) = \frac{E_0}{E(\sigma_{\max})} \tag{4}$$

2.2 Strain formulation

The nonlinear viscoelastic model where viscoelastic strain is used as an independent variable was developed by Schapery [12, 13] using expansion of Helmholzs' free energy. In one-dimensional case, the constitutive law is in form

$$\sigma = E_r \varepsilon_{VE} + h_1 \int_0^t \Delta E(\xi - \xi') \frac{d(h_2 \varepsilon_{VE})}{d\tau} d\tau \tag{5}$$

In (5) E_r is relaxed elastic modulus and, generally speaking, may be nonlinear function of strain. The "reduced time" ξ is introduced in (5) as

$$\xi = \int_0^t \frac{dt'}{a_\varepsilon} \text{ and consequently } \xi' = \int_0^\tau \frac{dt'}{a_\varepsilon} \tag{6}$$

Parameters h_1 and h_2 and the shift factor a_ε are strain invariant dependent. The transient part of the viscoelastic response is characterized by $\Delta E(\xi)$ which does not depend on stress and have a form of Prony series

$$\Delta E(\xi) = \sum_i E_i \exp\left(\frac{\xi}{\lambda_i}\right) \tag{7}$$

In (7) E_i are constants and λ_i are relaxation times. If a region can be found where E_r is a linear function of the applied strain and $h_1 = h_2 = a_\varepsilon = 1$, equation (5) is reduced to stress-strain relationship for linear viscoelastic material.

All parameters for strain formulations can be obtained from relaxation tests where viscoelastic strain is kept constant. However, most of materials also exhibit viscoplastic behaviour, thus such experiments where viscoelastic strain is kept constant become very complicated [14].

Since relaxation tests are difficult to perform, it would be more convenient to use creep test for viscoelastic characterization. But both forms of material model cannot be directly inverted as it was demonstrated in [12, 13]. The exact inversion would lead to a qualitatively different form of the material model than the model derived from Helmholtz's free energy, with an unknown qualitative and quantitative discrepancy. This issue can be overcome by using incremental form of models as described in the next section.

2.3 Inverted incremental model

In order to obtain inverted form of the model the stress formulation model was written in incremental form and then numerically inverted [15].

In general incremental model (not inverted form) has a form of:

$$f\left(\sigma^{k+1}, \varepsilon^{k+1}, \sigma^1, \ldots, \sigma^k\right) = 0 \tag{6}$$

In this model the viscoelastic strain in instant $t = t_{k+1}$ is calculated using a known solution at t_k. The material model presented in an incremental form has stress as the independent variable whereas the strain is calculated, which is suitable for simulation of stress controlled tests. To simulate strain controlled tests, increment of the applied strain has to be the input and stresses have to be calculated. In order to do this the incremental model was inverted to manage cases when the applied strain dependence on time, $\varepsilon^k = \varepsilon(t_k)$ $k = 0,1,2...$ is given and the corresponding stress has to be determined. Once the stress σ^k for the instant t_k is known, the mathematical problem is to find stress σ^{k+1} corresponding to the time instant t_{k+1}. If in typical incremental model, the solution is simple, in the inverted form all viscoelastic functions g_1, g_2, a_σ and ε_0 are dependent on σ^{k+1}. Therefore solution, σ^{k+1} of (8) for each k has to be found numerically, for example, by iterations using "bisection method".

5 RESULTS AND DISCUSSIONS

When inverted incremental model was used to simulate simple stress-strain curves obtained results showed two different tendencies. In the case when it was possible to perform creep tests (to obtain model parameters) at stresses very close to material failure, simulation showed good agreement with experimental results (Fig. 1a). However, if creep rapture occurs and characterization is possible only at relatively low stresses, nonlinear viscoelastic and viscoplastic parameters have to be extrapolated from existing data and this can lead to inaccuracy. It can very well be seen in Fig. 1b: until 18 MPa, where creep test was still possible to perform, the fit between experimental results and simulations are good but after that stresses are overestimated. In [16] it was shown, that it is possible to obtain better agreement between experimental data and simulations by tuning viscoelastic and viscoplastic parameters using stress-strain curves. This was suggested as a way to further adjust extrapolations, if it is not possible to perform creep tests at high stresses.

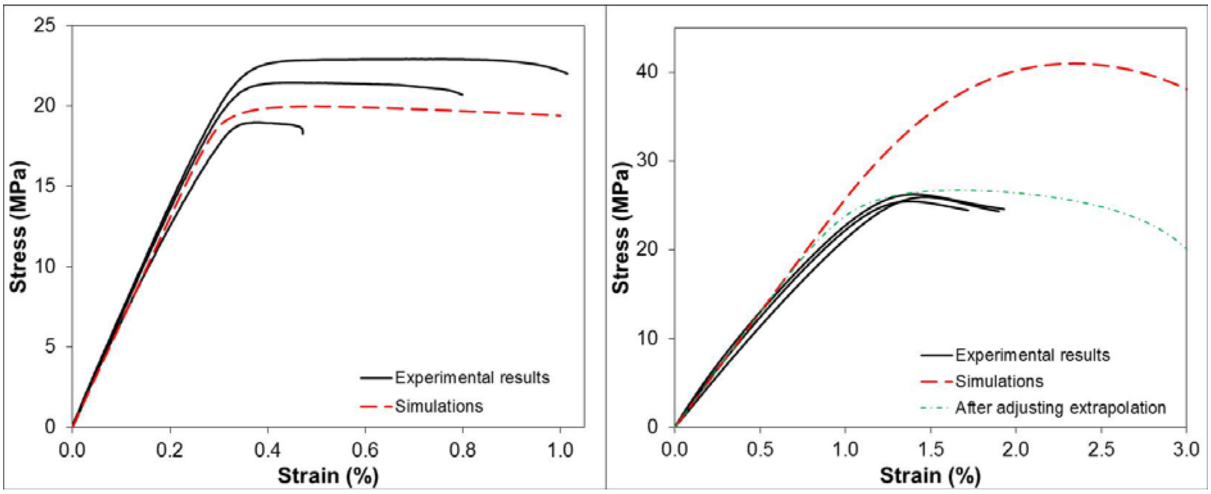

Figure 1: Experiments and simulations of stress-strain curves short randomly oriented flax fiber and a) Lignin, b) PLA composite. Tensile test were performed with 5mm/min crosshead displacement rate, more details about experiments can be found in [16].

These simulations (Fig. 1) show that inverted incremental model can be used to simulate strain controlled tests, but in cases when due to creep rapture viscoelastic and viscoplastic parameters have to be extrapolated, further analysis of quality of extrapolation has to be performed.

With all these models developed now it is possible to simulate stress or strain controlled tests. However in order to obtain all parameters needed for material models, large number of time consuming experiments have to be performed. If composite is characterized in this way, the parameters are obtained for specific combination of type of constituents (resin, fibers) and their volume fractions. Thus if one of these parameters is changed, characterization of composite has be performed all over again. Therefore it would be more convenient if instead of characterizing composite, each constituent would be characterized separately and composite performance is simulated based on these results.

This approach is demonstrated here by assuming iso-strain condition and Schapery's strain formulation. For simplicity, the rule of mixtures is used, but it is possible to use also more complicated models, such as concentric cylinder assembly, although in this case "nonlinear Poisson's ratio" has to be analyzed as well.

Validation of this model was performed on regenerated cellulose fiber (RCF) and epoxidized pine-oil based resin EpoBioX composite with fiber volume fraction between 0.55-0.70.

Experiments showed that EpoBioX has no viscoplastic strain component [17] and characterization of viscoelasticity was possible to obtain directly from relaxation experiments. However RCF exhibited high presence of viscoplastic strain [17]. Therefore, at first RCF were subjected to creep and then inverted incremental model was employed to simulate relaxation tests with pure viscoelastic strain. The results of these simulations were used to characterize viscoeaslticity. Full viscoleatic and viscoplastic characterization of these constituents, can be found in [17].

The four step strain controlled loading ramp was simulated for RCF/EpoBioX composite using strain formulation and ROM:

1) Loading with 0.03%/min up to 1% strain;
2) Unloading with 0.01%/min down to 0.7% strain;
3) Holding constant strain at 0.7% for 30 min;
4) Loading with 0.02%/min up to 4% strain

The experimental results and simulations can be seen in Fig. 2. Even though experimental curve shows some fluctuation, most likely due to experiment scatter, it fits well in between simulations for composites with two extreme cases of fiber volume fractions (0.55 and 0.70).

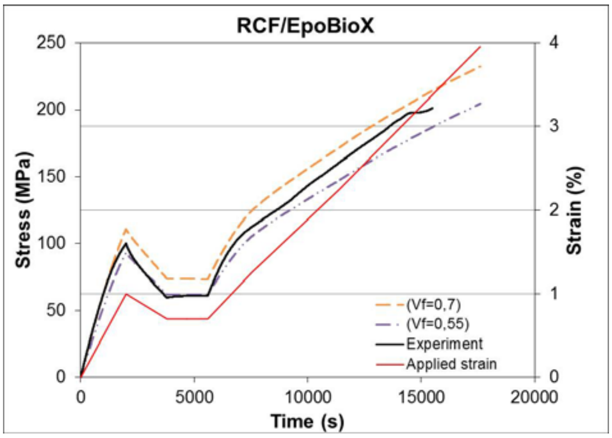

Figure 2: Comparison of simulation and experiment of RCF/EpoBioX composite with different fiber volume fractions.

With this approach now it is possible to simulate strain controlled tests of composites with different fiber volume fraction (Fig. 3a) and it is also possible to simulate relaxation tests with pure viscoelastic strain to obtain from these curves viscoelastic parameters for composite (Fig. 3b).

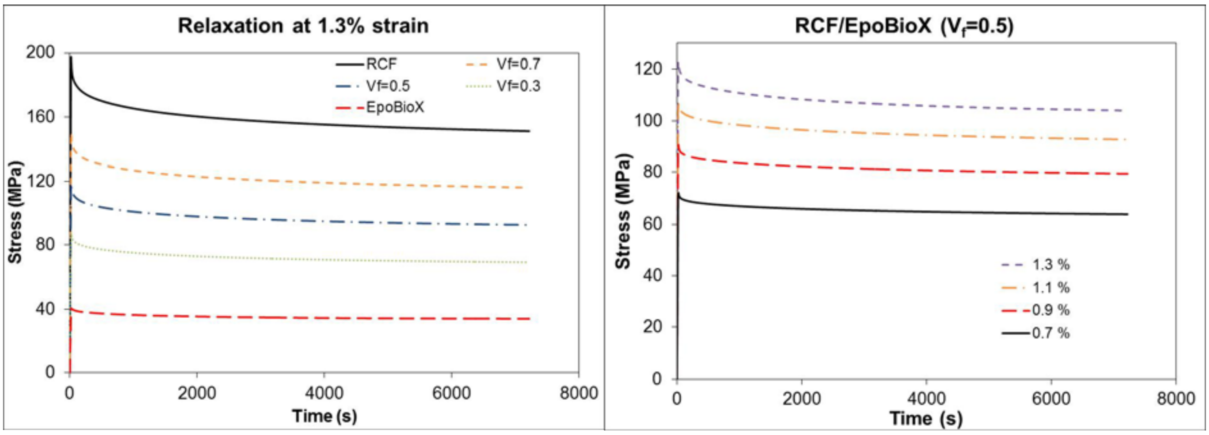

Figure 3: Simulation of RCF/EpoBioX composite relaxation test a) at 1.3 % strain and different fiber volume fractions and b) composite with 0.5 fiber volume fraction at different strain levels.

6 CONCLUSIONS

Two alternative formulations of non-linear viscoelastic model in terms of stresses or strains can be used to simulate experiments. In order to obtain parameters for strain formulations, relaxation test has to be performed. Due to viscoplastic strain component such tests are difficult to perform, therefore other methodology for characterization of viscoelasticity has to be found. Instead creep test (stress formulation) is used but since both models cannot be exactly mutually inverted, numerical procedures are employed.

Inverted incremental model has showed ability to simulate strain controlled tests, in region where creep test can be performed to characterize viscoelasticity. If creep rupture occurs at relatively low stresses, stress-strain curves can be used to further tune extrapolation of viscoelastic and viscoplastic parameters at high stress region. Inverted incremental model can be further employed to simulate relaxation tests and these results used to obtain viscoelastic parameters for Schapery's strain formulation.

Schapery's strain formulation combined with micro-mechanical models (for example in situations where iso-strain assumption is applicable it may be rule of mixtures or concentric cylinder assembly) can be used to simulate composite performance based on properties on constituents. Simulations with this approach showed good agreement with experiment. This methodology allows simulation of strain controlled tests for various compositions (e.g. different fiber volume fractions), without time-consuming analysis and experiments performed on each new composite. This method can be further used to simulate relaxation tests to obtain macroscopic viscoelastic properties for composite.

ACKNOWLEDGEMENTS

Part of this study was financially supported by EXCEL project, "Increased competitiveness and visibility of Composite Centre Sweden through excellence in bio-nano multi-scale composites and high-temperature composites" funded by local government Norrbotten, SWEDEN.

Swerea SICOMP is acknowledged for supplying of materials and help with manufacturing.

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