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Epoxy resin as a bonding agent in polymer matrix composites: material properties and numerical implementation

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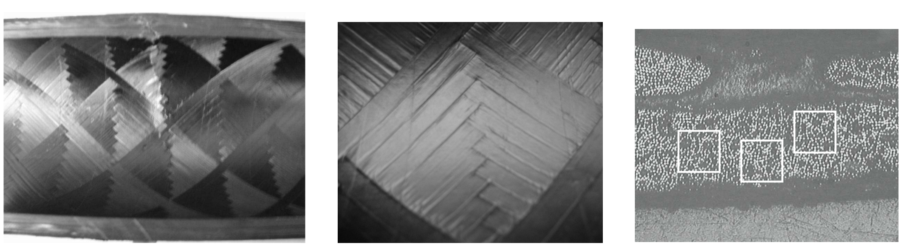
Summary

Time and stress dependent material properties used in the framework of the generalized Leonov model are derived for the PR100/2+EM100E epoxy matrix. A reliable procedure based on partial fractions is employed to extract elastic shear moduli and relaxation times for the Maxwell-chain model from creep experiments. Both fully-implicit and so called semi-implicit integration scheme is then proposed for the numerical solution of the resulting set of governing differential equations. For clarity reasons the presented applications are limited to a one-dimensional problem.

Introduction

Owing to their undoubtable benefits such as high strength, light weight, non-corrosive properties, production variability, etc., the polymer matrix composite systems reinforced either by aligned fibers, whiskers or fabrics are still in the continuous rise, particularly in civil infrastructure applications or sport industry. In a variety of applications the polymer matrix composites are often subjected to either cyclic or long-lasting loading that may trigger the nonlinear creep response of the matrix phase. Thus the long term behavior of polymer matrices at various stress levels should be carefully examined particularly in view of expected applications.

To introduced the subject we consider a large wound composite tube used, e.g., as a mast in sport sailing boats. In such an example the mechanical response of the polymer matrix should be explored within the framework of large multi-scale analysis that would address all possible deformation mechanisms including rate dependent behavior and damage evolution due to debonding at all scales, Fig. 1.



(a) (b) (c)

Figure 1: A scheme of three-scale modeling

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In the present contribution we substantially reduce the complexity of such a task and limit our attention to the description of rate dependent behavior of the epoxy resin both from the material and numerical point of view. As an example we consider the PR100/2+EM100E epoxy used as a bonding agent for the composite structure plotted in Fig. 1. An experimental program carried out on this type of material [2] demonstrated a relevant rate dependent response of the epoxy well described by the generalized Leonov model [1]. Although some experimental observations advocate a pressure dependent behavior of such materials [3] the present approach assumes negligible volume deformation during plastic flow, which leads to a standard Mises-like formulation. Due to expected applications in composites and confined extensibility of the reinforcements the formulation is limited to small strains.

Formulation of the Leonov model using small strain theory

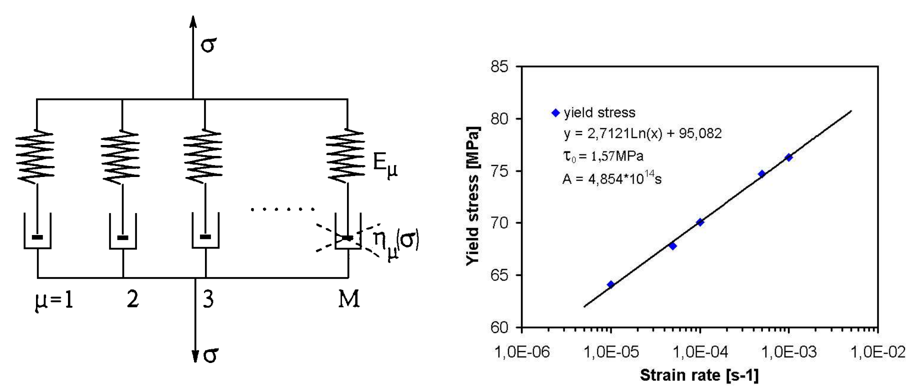
Combing the Eyring flow model for the plastic component of the shear strain rate

$$\frac{\mathrm{d}e_p}{\mathrm{d}t} = \frac{1}{2A}\sinh\frac{\tau}{\tau_0}, \tag{1}$$

with the elastic shear strain rate $\mathrm{d}e_e/\mathrm{d}t$ yields the one-dimensional Leonov model [1]

$$\frac{\mathrm{d}e}{\mathrm{d}t} = \frac{\mathrm{d}e_e}{\mathrm{d}t} + \frac{\mathrm{d}e_p}{\mathrm{d}t} = \frac{\mathrm{d}e_e}{\mathrm{d}t} + \frac{\tau}{\eta(\mathrm{d}e_p/\mathrm{d}t)}, \qquad \eta(\mathrm{d}e_p/\mathrm{d}t) = \frac{\eta_0\tau}{\tau_0\sinh(\tau/\tau_0)} = \eta_0 a_\sigma(\tau). \tag{2}$$

where η is the shear-dependent viscosity. In Eq. (1), A and τ_0 are material parameters; a_σ that appears Eq. $(2)_b$ is the stress shift function with respect to the zero shear viscosity η_0 (viscosity corresponding to an elastic response). Clearly, the phenomenological representation of Eq. $(2)_a$ is the Maxwell model with the variable viscosity η.



(a) (b)

Figure 2: (a) Generalized Maxwell chain model, (b) Eyring plot

To describe multi-dimensional behavior of the material, the generalized compressible Leonov model, equivalent to the generalized Maxwell chain model, Fig. 2(a), can be used.

The viscosity term corresponding to the μ-th unit receives the form

$$\eta_\mu = \eta_{0,\mu} a_\sigma(\tau_{eq}), \qquad \tau_{eq} = \sqrt{\frac{1}{2} s_{ij} s_{ij}}, \tag{3}$$

where τ_{eq} is the equivalent shear stress and s_{ij} is the stress deviator tensor. Admitting only small strains and isotropic material, a set of constitutive equations defining the generalized compressible Leonov model can be written as

$$\sigma_m = K\varepsilon_v, \tag{4}$$

$$\frac{\mathrm{d}s}{\mathrm{d}t} = \sum_{\mu=1}^{M} 2G_\mu \left(\frac{\mathrm{d}e}{\mathrm{d}t} - \frac{\mathrm{d}e_{p,\mu}}{\mathrm{d}t} \right), \tag{5}$$

$$s_\mu = 2\eta_\mu \frac{\mathrm{d}e_{p,\mu}}{\mathrm{d}t} = 2\eta_{0,\mu} a_\sigma(\tau_{eq}) \frac{\mathrm{d}e_{p,\mu}}{\mathrm{d}t}, \qquad s = \sum_{\mu=1}^{M} s_\mu, \tag{6}$$

$$\sigma = \sigma_m [\mathrm{I}] + s, \tag{7}$$

where σ_m is the mean stress, ε_v is the volumetric strain, K is the bulk modulus, G_μ is the shear modulus of the μ-th unit and $[\mathrm{I}]$ is the identity matrix.

Stress dependent material parameters of the Leonov model

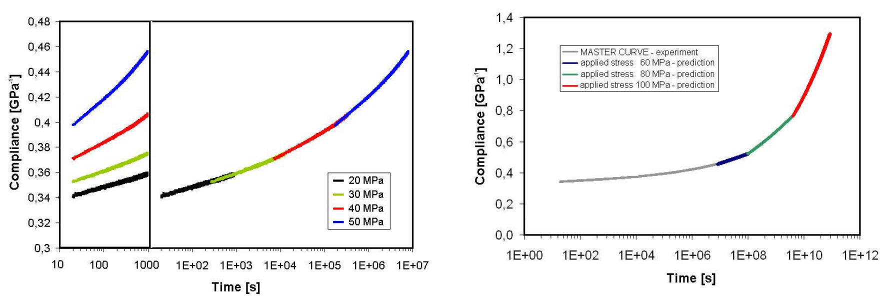
The first step in successful implementation of the Leonov material model requires the determination of stress dependent material parameters A and τ_0 that appear in Eqs. (1) and $(2)_b$. These parameters can be derived from the Eyring plot, Fig. 2(b), assuming that at yielding the overall deformation equals the plastic deformation. The yield stress is then associated with a stress state that does not change during continuous yielding under constant strain rate. Providing this assumption applies and assuming uniaxial tensile stress experiment, Eq. (7) can be recast in the form

$$\sigma_y = \tau_0 \sqrt{3} \ln(2A\sqrt{3}) + \tau_0 \sqrt{3} \ln \dot{\varepsilon}, \tag{8}$$

where σ_y is the yield stress and $\dot{\varepsilon}$ is the prescribed strain rate. Derivation of parameters A and τ_0 thus calls for a set of experiments conducted at different strain rates until the corresponding yield stress is reached. The results from such an experimental program, in which the strain rates were taken from the interval $10^{-5}s^{-1} - 10^{-3}s^{-1}$, appear in Fig. 2(b). The solid line is the plot of Eq. (8) with parameters $A = 4.854 \times 10^{14}$ s and $\tau_0 = 1.57$ MPa found for the present PR100/2+EM100E epoxy matrix.

Time dependent material parameters of the Leonov model

To complete the nonlinear time dependent formulation of the Leonov model it remains to determine the coefficients G_μ and viscosities $\eta_{0\mu}$ related to relaxation times θ_μ These parameters are found by transforming the experimentally derived creep function into relaxation one using, e.g., the Laplace transform combined with the method of partial fractions [2]. The creep function is usually found by loading the specimen in a rather low stress level, which does not promote inelastic response, for a sufficiently long time. Neither requirement, however, was met for the presented experimental program where even the creep experiment carried out at lowest stress level 20 MPa violated the linear viscoelastic condition. The time elapse up to 900 s was also insufficient to allow prediction at much longer times. To avoid this obstacle we constructed the creep function as a master curve using the stress superposition principle and the creep data received at larger stresses for a short duration of time as shown in Fig. 3(a). Note that the largest stress level used in experimental measurements was 50 MPa. This value is still rather low, particularly when attempting to simulate yielding at larger stress levels (60 - 100 MPa). Note that in case of uniaxial stress the shift factor a_σ for 60 MPa equals approximately 10^{-8} which significantly reduces the applicability of the creep function in Fig. 3(a) at this stress level to app. 10 s. To solve this problem we interpolated the creep functions for available stress states in the form $J(t) = a(\sigma)t^{n(\sigma)}$. Analysis of this formula for individual curves provided analytical expressions for parameters $a(\sigma)$ and $n(\sigma)$ [2], which in turn allowed extension of the creep function up to 10^{11} s as shown in Fig. 3(b).

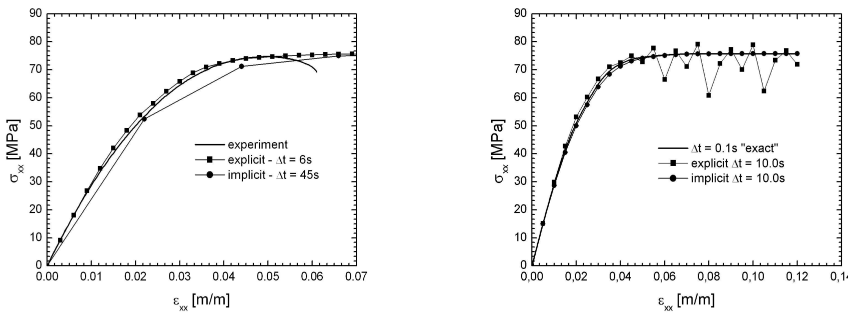


(a) (b)

Figure 3: (a) Master curve derived from available experimental data, (b) Analytical extension of experimentally derived master curve using interpolation

Numerical implementation of the Leonov model

As suggested by the title one of the objectives was to suggest a reliable and stable procedure for the integration of a set of governing equations (4) - (7). Here we present a brief comparison between the fully implicit integration scheme and the fully explicit procedure with forward integration step often used in linear viscoelastic analysis. A possible improvement by considering a linear variation of the stress shift variable a_σ in so called semi-implicit integration scheme is also considered. To keep the discussion simple we limit our attention to a one-dimensional problem. To introduce the subject consider Fig. 4(a) showing a typical uniaxial response of the PR100/2+EM100E epoxy subjected to a constant tensile strain rate. The plotted curves are found for $\dot{\varepsilon}_x = 5 \times 10^{-4}$ s^{-1}. The solid line is obtained experimentally while the others follow from the numerical analysis using the two integration schemes with the largest possible time increments, for which no stability problems occurred.



(a) (b)

Figure 4: Explicit vs. implicit integration scheme

Fig. 4(a) suggests that in order to avoid numerical instabilities with the explicit forward Euler method a relatively short time increment must be prescribed. This becomes clear once we recall the basic ingredient of the method, which is the assumption that all time dependent parameters are taken, for the forward integration step, from the beginning of a new time increment and kept constant. This assumption clearly breaks down for the stress shift parameter a_σ, which is a highly nonlinear function of stress and rapidly approaches zero with increasing stress level. This method, however, is extremely simple to implement and requires only a few calculations per time integration step. Nevertheless, the conditional stability may be the major obstacle in successful implementation within the framework of large multi-scale computation.

A rather different numerical response is evident for the fully implicit integration scheme. Although at a slight expense of accuracy, the stable behavior outlast even for a relatively large time step. On the other hand, a local Newton-Raphson iteration is required to arrive at correct values of the time dependent variables at the end of a given time step. While this may slow down the local integration, a significant reduction in number of required time steps may eventually prove beneficial. The fully implicit integration scheme will be now given in a one-dimensional setting. Equations driving the one-dimensional tensile viscoelastic response are

$$
\begin{aligned}
\sigma(t_i) &= \sigma(t_{i-1}) + \widehat{E}(t_i)\,(\Delta\varepsilon - \widehat{\varepsilon}(t_i))\,, \\
\widehat{E}(t_i) &= \sum_{\mu=1}^{M} E_\mu \frac{\theta_\mu a_\sigma(t_i)}{\Delta t}\left(1 - \exp\left(-\frac{\Delta t}{\theta_\mu a_\sigma(t_i)}\right)\right), \\
\widehat{\varepsilon}(t_i) &= \frac{1}{\widehat{E}(t_i)} \sum_{\mu=1}^{M}\left(1 - \exp\left(-\frac{\Delta t}{\theta_\mu a_\sigma(t_i)}\right)\right)\sigma_\mu(t_{i-1}), \\
\tau_{eq}(t_i) &= \frac{\sigma(t_i)}{\sqrt{3}}, \qquad a_\sigma(t_i) = \frac{\tau_{eq}(t_i)}{\tau_0} \Big/ \sinh\frac{\tau_{eq}(t_i)}{\tau_0}.
\end{aligned}
$$

The vector of residuals $\{\mathsf{r}\} = \{T, G, A\}^{\mathsf{T}}$ in Newton-Raphson iteration step becomes

$$\begin{aligned} T &= \tau_{eq}(t_i) - \frac{1}{\sqrt{3}}\left(\sigma(t_{i-1}) + \widehat{E}(t_i)\left(\Delta\varepsilon - \Delta\widehat{\varepsilon}(t_i)\right)\right), \\ G &= \widehat{E}(t_i) - \sum_{\mu=1}^{M} E_\mu \frac{\theta_\mu a_\sigma(t_i)}{\Delta t}\left(1 - \exp\left(-\frac{\Delta t}{\theta_\mu a_\sigma(t_i)}\right)\right), \\ A &= a_\sigma(t_i) - \frac{\tau_{eq}(t_i)}{\tau_0} / \sinh\frac{\tau_{eq}(t_i)}{\tau_0}, \end{aligned}$$

where the vector of unknowns receives the form

$$\{\mathsf{a}\} = \{\tau_{eq}(t_i), \widehat{E}(t_i), a_\sigma(t_i)\}^{\mathsf{T}}. \tag{9}$$

Under the condition that $\Delta\varepsilon$ is constant the Newton-Raphson iterative scheme reads

$$\{\mathsf{a}\}^{k+1}(t_i) = \{\mathsf{a}\}^{k}(t_i) - [\mathsf{H}]^{-1}\{\mathsf{r}\}^{k}, \tag{10}$$

where $[\mathsf{H}]$ is the Jacobian matrix. A simple extension to three dimensions is presented in [4].

The results displayed in Fig. 4(b) provide further notion about both methods when applied to the present problem. While for the forward (fully) explicit method the results show an oscillatory response attributed to the assumed constant stress shift parameter a_σ over a given time step, no such behavior was observed for the backward (fully) implicit method for all time increments marked with the success in convergence of the Newton-Raphson iteration. A slight improvement can be gained by considering a linear variation a_σ over the time increment in so called semi-implicit scheme. Details can be found in [2].

Acknowledgments

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