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TIME-DEPENDENT RESPONSE OF A RUBBER-TOUGHENED CARBON/EPOXY COMPOSITE WITH DAMAGE ACCUMULATION

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Summary. Polymeric composites are frequently modeled as linear elastic materials. However, matrix-dominated properties, such as transverse and shear modulus, can display significant nonlinear time-dependence, especially under conditions of high stress and aggressive environment. This behavior is primarily due to the viscoelastic nature of the polymeric matrix. In addition, polymeric composites also present time-dependent damage growth. In this work, nonlinear viscoelastic constitutive equations were used to represent the time-dependent behavior of a rubber-toughened carbon/epoxy composite during damage growth. These equations were originally devised to characterize material response in a stable damage state. In this approach, however, nonlinearities due to damage and viscoelasticity were incorporated by the model stress-dependent functions, allowing its use in the presence of damage accumulation. A procedure was proposed and applied to separate viscoelastic and damage effects. An experimental program consisting of multiple cycle creep and recovery tests was performed to determine the time-dependence of the shear compliance and to verify the theory as well. The results obtained indicated an excellent agreement between theory and experiment. Constant stress rate tests were also used to validate the application of the theory.

Keywords: Composites, Viscoelasticity, Damage, Carbon/Epoxy

1. INTRODUCTION

Polymeric composites are frequently modeled as linear elastic materials. However, matrix-dominated properties, such as transverse and shear modulus, can display significant nonlinear time-dependence, especially under conditions of high stress and aggressive environment. For this reason, linear and nonlinear viscoelastic constitutive equations need to be considered in order to assess the durability and long-term performance of composite structures. Viscoelastic strains are reversible and decrease gradually upon load removal, reaching complete recovery if sufficient time is allowed. In addition to viscoelastic strains, time-dependent irreversible strains also develop in polymeric composite materials as a result of mechanical degradation such as matrix cracking, delaminations, fiber-matrix debonding, etc.

The occurrence of irreversible strains in polymeric composites is evidenced during multiple-cycle creep/recovery tests by observing the changes in the strain response from cycle to cycle. When a specimen is first loaded in a creep test the number of cracks or, in more general terms the micro-structural damage increases with time. As the cycling proceeds, damage saturation is often reached and none or negligible damage growth is observed thereafter. Subsequent loading at that stress level should result in identical strain response with all the time-dependence credited to the intrinsic material viscoelastic behavior. The material in this state is said to be mechanically conditioned.

The nonlinear viscoelastic constitutive equations proposed by Schapery (1969) have been widely used in modeling the time-dependent response of both neat resin and composites in the absence of significant damage growth. Attempts were also made in using these equations to represent material response with damage growth. See, for instance, Tuttle & Brinson (1986), Yen & Williamson (1990) and Walrath (1991). However, the recovery response obtained from the constitutive equations can only represent reversible strains. For this reason, most experimental data found in the literature using this theory or specialized cases for the characterization of material with damage growth are limited to represent creep behavior.

In this work, a procedure is developed for data reduction that extends the use of the constitutive equations, allowing the characterization of the recovery response in the presence of non-reversible strains. This procedure is based on the use of the mechanically conditioned material response to separate the viscoelastic and damage effects. Test results for the shear compliance of a carbon/epoxy composite are presented and the predicted results were found to correlate very well with the experimental data.

2. CONSTITUTIVE EQUATIONS

In the nonlinear constitutive equations proposed by Schapery, the time dependent strains for uniaxial loading are represented by:

$$\varepsilon(t) = g_o D(0)\sigma + g_1 \int_0^t \Delta D(\psi - \psi') \frac{\partial (g_2 \sigma)}{\partial \tau} d\tau \qquad (1)$$

where $D(0)$ and ΔD are the initial and transient components of the linear viscoelastic creep compliance, ψ and ψ' are defined by:

$$\psi = \int_0^t \frac{dt'}{\mathrm{a}_s} \quad ; \quad \psi' = \int_0^\tau \frac{dt'}{\mathrm{a}_s} \qquad (2)$$

and g_o, g_1, g_2 and a_s are stress-dependent material properties. For many polymeric materials, the transient component of the linear creep compliance can be approximated by a power law in time:

$$\Delta D(\psi) = D_1 \psi^n \qquad (3)$$

where D_1 is the creep coefficient of the linear viscoelastic creep compliance.

Creep and recovery tests provide a simple stress history that can be used to determine the parameters needed in equation (1). A constant stress σ_o is applied instantly at time t = 0 and also instantly removed at time t = t_1. This stress history can be represented as:

$$\sigma = \sigma_o\left[H(t) - H(t - t_1)\right] \tag{4}$$

where H(t) is the Heaviside step function.

The strain response during the creep period is obtained by substitution of equation (4) into equation (1) for $t < t_1$:

$$\varepsilon(t) = g_o D(0)\sigma_o + g_1 g_2 D_1\left(\frac{t}{a_s}\right)^n \sigma_o \tag{5}$$

For $t > t_1$, the recovery response is given by

$$\varepsilon_{rec.}(t) = \frac{\Delta\varepsilon_1}{g_1}\left[(1 + a_s\lambda)^n - (a_s\lambda)^n\right] \tag{6}$$

where $\Delta\varepsilon_1$ and λ are given by:

$$\Delta\varepsilon_1 = g_1 g_2 D_1\left(\frac{t_1}{a_s}\right)^n \sigma_o \tag{7}$$

$$\lambda = \frac{t - t_1}{t_1} \tag{8}$$

3. MATERIAL AND EXPERIMENTS

The material used in this test program consisted of ASC4 carbon fibers with a matrix of E719LT rubber-toughened epoxy resin produced by B-P Chemicals. The panels were manufactured by filament winding on a flat mandrel and cured following an autoclave/vacuum bag procedure at a temperature of 121°C during 2 hours. The specimens were cut with a diamond grinding wheel from 30.5 cm x 30.5 cm panels and stored in plastic bags with desiccant before being tested. $[\pm 45°]_s$ and $[\pm 45°]_{3s}$ specimens were used in uniaxial tensile creep/recovery tests to determine the time-dependence of the shear compliance $S_{66}(t)$. The specimens were 152 cm long with a gage section of 102 mm, providing a gage length-to-width ratio of 8:1. Nominal specimen width was 12.7 mm and average layer thickness of 0.268 mm.

Every specimen was instrumented with two Micromeasurements CEA-06-062UT-350 strain-gages. This is a 350Ω two-element 90° tee rosette capable of measuring both longitudinal and transverse strains. The strain-gages were bonded to the specimens using M-Bond AE-10 adhesive, cured at 65°C for 2 hours followed by a post-cure period of 1 hour at 50°C to provide an essentially creep-free response.

Strain-gages were placed on both sides of each specimen to correct for eventual bending. In addition, the tests were performed under the condition that the difference between the readings from the two axial strains could not exceed 5%. Strain-gage readings were performed individually and averaged during the data reduction analysis. In order to compensate for temperature variations, a dummy specimen with identical fiber orientation and gage configuration was connected to an arm adjacent to the active arm in the Wheatstone bridge during each test.

A typical test consisted of a series of creep/recovery cycles applied successively, with each cycle consisting of 3 hours under creep and 21 hours under recovery. The response with damage growth was obtained from the first cycle. In order to characterize viscoelastic response with no damage growth, each specimen was subjected to a minimum of 6 and up to 12 creep/recovery cycles until no appreciable changes in the strain-time behavior were detected that could be credited to the development of damage.

4. TIME-DEPENDENCE WITH DAMAGE GROWTH

The constitutive equations described in section 2 have been mainly and most successfully used in the characterization of nonlinear viscoelastic behavior in the absence of significant damage growth. Experimental results (Tuttle & Brinson, 1986) indicate, however, that equation (1) is capable of predicting the creep response in the presence of damage growth. The parameters appearing in the instantaneous and transient strain components can be determined so as to account for the effects of growing damage. Indeed, it has also been shown that a power law in time can be used to represent the transient component of creep compliance with damage growth (Tuttle & Brinson, 1986). On the other hand, the recovery equation as presented in equation (6) can only represent viscoelastic, or completely reversible strains, not accounting for nonreversible strains resulting from damage. Note that $\varepsilon_{rec.} \rightarrow 0$ as $a_s\lambda >> 1$.

Tuttle and Brinson (1986) used Schapery's equations to characterize the nonlinear time-dependent behavior of T300/5208 graphite/epoxy composites in the presence of damage growth including recovery behavior. In their study they could only obtain stable values for the model parameters by first subtracting from the recovery data any residual strain existing at the end of the cycle, translating the entire recovery curve towards zero strain. This allowed the use of the recovery equation (6) to compute the nonlinearizing parameters appearing in the constitutive equation (1).

Due to the occurrence of damage, the recovery strains do not return to zero, but rather approach asymptotically a permanent strain level. One must recognize, however, that the strain remaining at the end of the recovery cycle contains not only a nonreversible component but also a reversible, viscoelastic component that did not have enough time to recover completely. This is particularly true if the time allowed for recovery is not sufficiently long.

Thus, in order to properly account for the amount of nonreversible strain to be subtracted from the total recovery strain, one must separate the reversible and non-reversible strain components. One option is to employ large recovery-to-creep time ratios, allowing the asymptotic behavior of the recovery strains to become apparent. Another option, introduced in this work, is to use the recovery response obtained from the mechanically conditioned material to compute the amount of viscoelastic strain remaining at the end of the first recovery cycle.

The microstructural damage that grows during the creep portion of the first cycle is assumed to remain constant during the recovery portion, when the specimen is unloaded. As a consequence, the nonreversible strains developed during creep are also assumed to stay constant during recovery. The resulting time-dependent response observed is due only to the recovery of the reversible, viscoelastic strain component. Figure 1 displays typical experimental recovery data, including the first cycle recovery, along with the averaged mechanically conditioned response for the same testing conditions. The mechanically conditioned response contains only reversible strains, while the first cycle response contains both reversible and non-reversible components. One can observe that for values of $\lambda < 3$ the difference between the first cycle and the mechanically conditioned recovery responses is not constant, indicating that they differ not only due to the presence of a nonreversible constant component but their viscoelastic responses are also different. It is postulated that the newly generated damage is more likely to affect the recovery response during short recovery times.

At long recovery times, however, the difference between these two responses becomes approximately constant. An approximation for the amount of nonreversible strain present in the first cycle can be obtained by subtracting the mechanically conditioned component from the first cycle total strains. A typical result for this strain difference is shown in the insert in Fig. 1. Notice that for values of $\lambda > 3$ it corresponds to approximately 31$\mu\varepsilon$ for this specific case. A more detailed description of the mechanically conditioned response of the ASC4/E719LT carbon/epoxy composite can be found in Soriano & Almeida (1998).

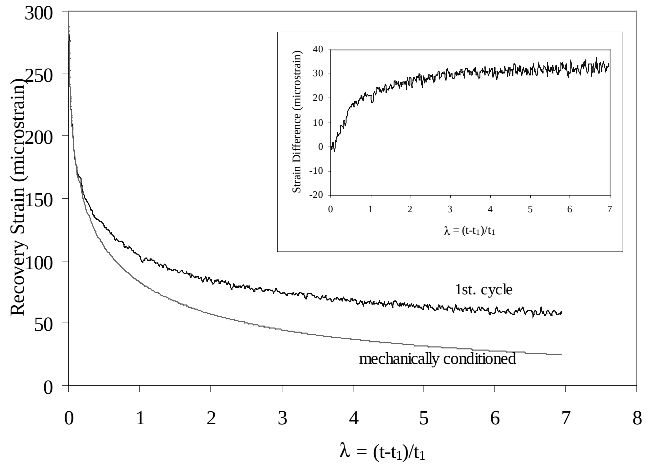


Figure 1 - Comparison between 1st cycle and mechanically conditioned recovery response.

4.1 Data Reduction

Equation (5) was fitted to experimental creep data and the set of parameters $g_0D(0)$, $g_1g_2D_1/a_s^n$ and n that minimized the square error were computed for every stress level. $D(0)$ and D_1 were computed from the linear viscoelastic response, in which $g_0 = g_1 = g_2 = a_s = 1$. As in the case of the mechanically conditioned material, creep data only is not enough to individually determine the functions g_1, g_2 and a_s. Recovery data is also needed.

The total recovery strain $\varepsilon_{rec.}(t)$ following a creep cycle in which the damage state of the specimen has been altered, consists of a time-dependent, viscoelastic component $\varepsilon_{reversible}(t)$ plus a constant, non-reversible strain component $\varepsilon_{nonreversible}$:

$$\varepsilon_{rec.}(t) = \varepsilon_{reversible}(t) + \varepsilon_{nonreversible} \tag{9}$$

The procedure discussed in the previous section is incorporated into the data reduction analysis by subtracting the time-independent nonreversible component from the total recovery strain. The resulting time-dependent component is assumed to keep the same form as presented in equation (6), but with material properties and nonlinear parameters representing a material whose damage state is different from the mechanically conditioned material. The resulting equation becomes:

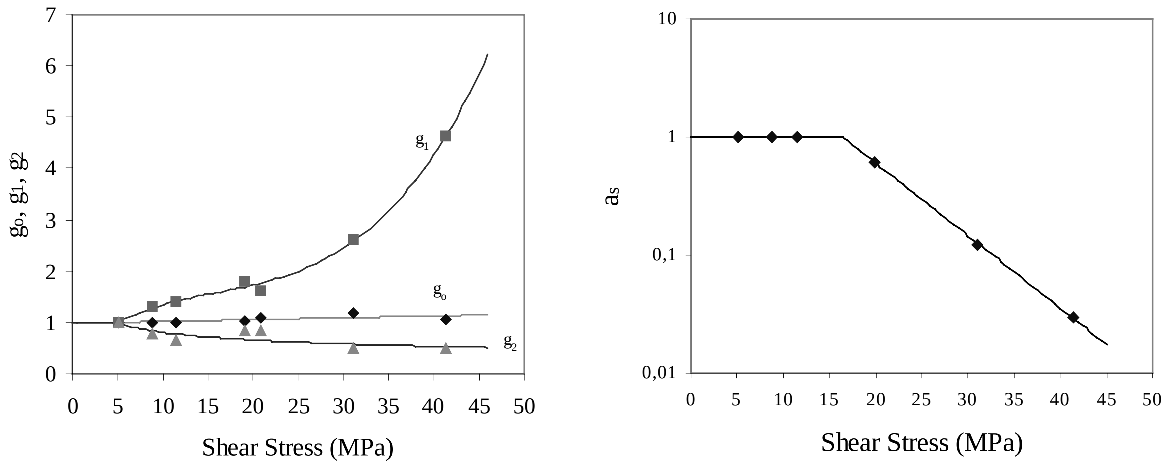
$$\varepsilon_{rec.}(t) - \varepsilon_{nonreversible} = \varepsilon_{reversible}(t) = \frac{\Delta\varepsilon_1}{g_1}\left[(1 + a_s\lambda)^n - (a_s\lambda)^n\right] \quad (10)$$

It should be clear that the exponent n may be different from the value obtained for the mechanically conditioned material. The amount of nonreversible strain $\varepsilon_{nonreversible}$ is computed as described in the previous section based on values of $\lambda > 3$. As a result, the right-hand-side of equation (10) can be used to determine the nonlinear functions for the 1st cycle through essentially the same procedures used for the mechanically conditioned material.

5. RESULTS

After subtracting the nonreversible component from the total recovery strains, the procedures described above were applied to separate the material functions g_1, g_2 and a_s describing the first cycle behavior. The creep exponent obtained for the first cycle was found to be higher than that obtained for the mechanically conditioned material. The first cycle exponent was found to be a function of the stress level, whereas the exponent for mechanically conditioned material is considered an intrinsic property of the matrix. One can argue that in the presence of damage growth the exponent encompasses effects such as the propensity of the laminate to nucleate, propagate and arrest cracks, which are certainly affected by the fiber orientation and stress level.

The average creep exponent for the first cycle shear compliance was n = 0.18 for shear stresses below 21 MPa, increasing up to n = 0.28 at 41 MPa. The stress dependent functions g_0, g_1, g_2, and a_s obtained from the first cycle creep/recovery of ±45° specimens, representing the shear compliance, are shown in Fig. 2. In this case, one can observe that most of the nonlinearity is concentrated in g_1. Figures 3 and 4 display the first cycle creep and recovery responses. Despite the high nonlinearity displayed by the shear creep and recovery responses, excellent agreement was observed between the model predictions and experimental data. Again, this is a strong indication of the capability of the model and of the suggested data reduction procedure to represent highly nonlinear time-dependent behavior.



(a) (b)

Figure 2 - Stress functions for first cycle shear behavior: (a) g_o, g_1, and g_2; (b) a_s

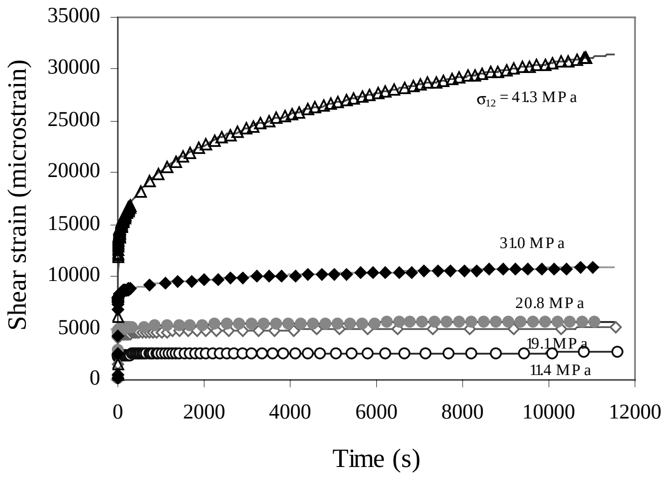


Figure 3 - First cycle creep response of ±45° specimens. Experiment and prediction.

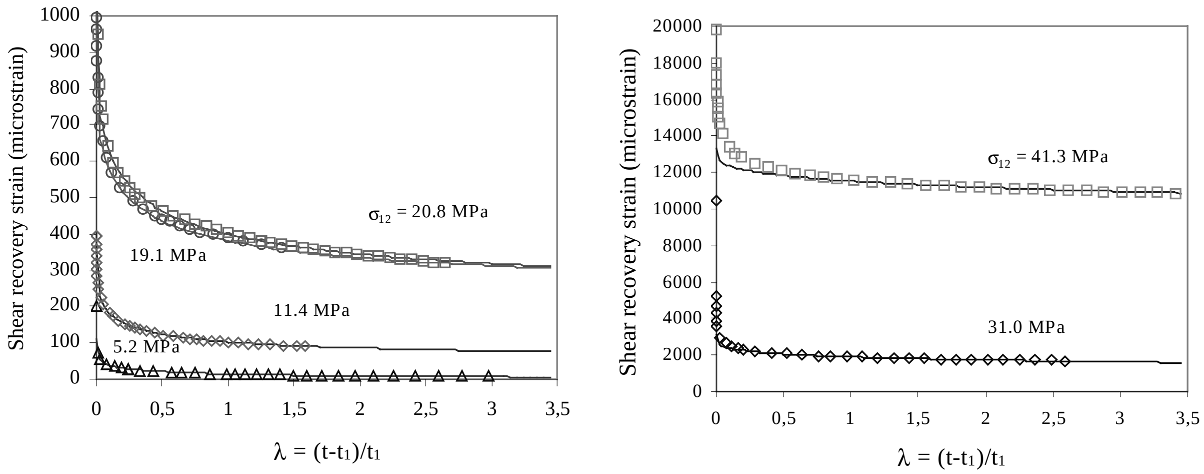


Figure 4 - First cycle recovery response of ±45° specimens. Experiment and prediction.

5.1 Constant Stress-Rate Tests

In order to verify the ability of the nonlinear viscoelastic constitutive equation displayed in equation (1) to represent loading histories other than creep and recovery, results from constant stress rate tensile tests performed in the ASC4/E719LT carbon/epoxy composite will be compared to the predictions of the theory. The specimens used for the constant stress rate tests were prepared and stored under the same conditions as the ones used for creep and recovery tests. All constant stress rate tests used here for comparison with the theoretical predictions were performed by Bocchieri (1996).

The stress history for the constant stress-rate test is expressed as

$$\sigma(\tau) = k\tau H(\tau) \tag{11}$$

where k is the constant stress rate and $H(\tau)$ is the Heaviside step function.
Substitution of equation (11) into equation (1) and the use of a power law to represent the transient component of the creep compliance results in

$$\varepsilon(t) = g_o D(0)kt + g_1 D_1 k\int_0^t (\psi - \psi')^n (g_2 + \sigma \frac{\partial g_2}{\partial \sigma}) d\tau \qquad (12)$$

The first step in using equation (12) to predict constant stress rate response was to determine analytic expressions for the functions g_o, g_1, g_2 and a_s. These expressions are shown below for both transverse and shear response and were obtained by interpolation of the experimental results shown in Fig. 2.

The nonlinear stress functions g_o, g_1, g_2 and a_s were determined entirely from experiments. As noted by Dillard (1991), there are no a priori temperature, moisture, or stress functional forms which can be considered appropriate for composite material systems. For this reason, no specific forms were defined a priori for the interpolation functions. The only requirement was to provide a good fitting within the range of stresses considered. It is likely that for some of the fittings other functions could have been used as well.

Functions for the shear compliance:

$S_{66}(0) = 0.199\ GPa^{-1}$; $S_{66}^* = 6.63x10^{-3}\ GPa^{-1}s^{-n}$

$g_0 = 0.98 + 3.50x10^{-3}\sigma_{12}$

$$g_1 = \begin{cases} 1 & \sigma_{12} \le 5 MPa \\ 0.47 + 0.14\sigma_{12} - 6.29x10^{-3}\sigma_{12}^2 + 1.31x10^{-4}\sigma_{12}^3 & \sigma_{12} > 5 MPa \end{cases}$$

$g_2 = 1.59\sigma_{12}^{-0.295}$

$$a_s = \begin{cases} 1 & \sigma_{12} \le 17\, MPa \\ 10.26\exp[-0.14\sigma_{12}] & \sigma_{22} > 17\, MPa \end{cases}$$

The functions defined above were then replaced in equation (12) and a numerical integration procedure was used to compute the strains. Note that $S_{66}(0)$ and S_{66}^* correspond to $D(0)$ and D_1 respectively in Eq. (5).

Such procedures would become much simpler if all nonlinearity could be concentrated in fewer parameters. It is observed from Fig. 2, for instance, that g_1 carries most nonlinearity of the transient component of shear compliance. This provides an argument for a first attempt in simplifying equation (12) by concentrating all nonlinearity in g_0 and g_1, with g_1 absorbing the nonlinearity from g_2 and a_s. In fact, concentrating all nonlinearity in g_o and g_1 provides the most straightforward and simple way of evaluating equation (12). Clearly, the most undesirable term to concentrate the nonlinearity is a_s. With all nonlinearity concentrated in g_o and g_1 equation (12) becomes:

$$\varepsilon(t) = g_o(\sigma)D(0)\sigma + g_1(\sigma)\frac{D_1}{k^n}\frac{\sigma^{n+1}}{n+1} \qquad (16)$$

Figure 5 shows comparisons between experimental data and predictions using equations (12) and (16) for two different constant stress rates. Again, good agreement was obtained between theory and experiment for both predictions. The effect of concentrating all nonlinearity in g_0 and g_1 is noticed only at high stresses. At low stresses the difference between the two theoretical curves is negligible.

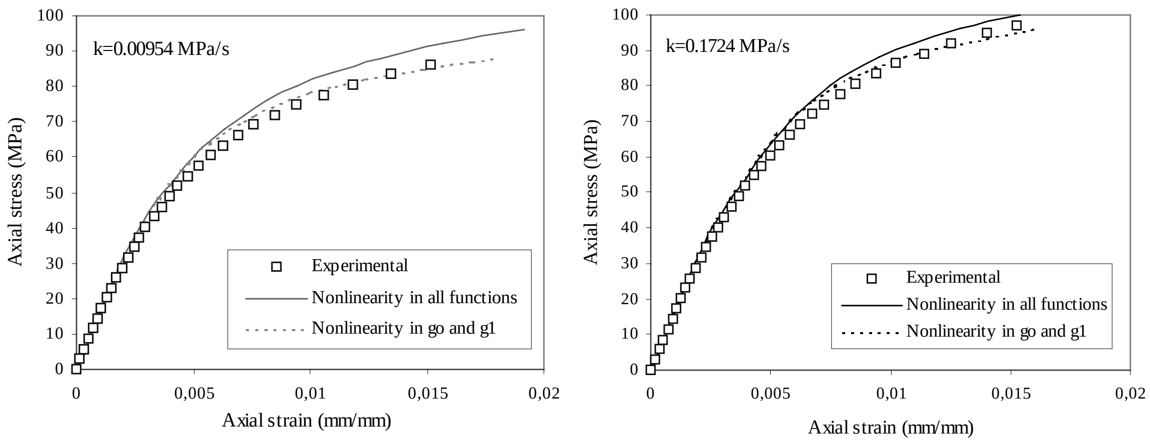


Figure 5 - Stress-strain behavior under constant stress rate. Comparison between prediction and experiment for a ±45° specimen.

6. CONCLUSIONS

The presence of a nonreversible strain component prevents the direct use of the recovery equation in Schapery's constitutive model. As a result, it is not possible to determine individually the model parameters g_1, g_2, and a_s. The nonreversible strain component must be removed from experimental recovery data before the data reduction procedure can be applied. Thus, in order to account for the amount of nonreversible strains to be removed from the first cycle recovery data, a procedure based on the mechanically conditioned recovery response was proposed in this work. This procedure was applied to the experimental recovery data and the model parameters g_1, g_2, and a_s were determined individually. This technique allows the use of Schapery's constitutive equations in the case where significant damage growth exists. It was proved to be very effective and an excellent representation for first cycle creep and recovery data was obtained.

In order to obtain further verification of the proposed method, the model parameters and material properties obtained from creep and recovery with damage growth were used to predict the material response to constant stress rate loading. Predictions were made under two conditions: with the nonlinearity applied to all model parameters, and with g_0 and g_1 concentrating all nonlinearity. Both conditions provided good agreement with experimental data. The advantages of concentrating all nonlinearity in g_0 and g_1 is that it greatly simplifies the use of the constitutive equation, while keeping the predictions in good agreement with experimental results.

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REFERENCES

Bochieri, R. T., 1996, A baseline nonlinear material characterization for predicting the long-term durability of composite structures, M.S. Thesis, The University of Texas at Austin, Austin, Texas, USA.

Dillard, D. A., 1991, Viscoelastic behavior of laminated composite materials, in Fatigue of Composite Materials, ed K. L. Reifsneider, Elsevier, Amsterdam.
Schapery, R. A., 1969, Further development of a thermodynamic constitutive theory: stress formulation, Purdue University Report 69-2.
Soriano, E. A. & Almeida, S. F. M., 1998, Nonlinear viscoelastic behavior of a rubber-toughened carbon/epoxy composite material, Proceedings of the XIII CBECIMAT, Curitiba, pp. 5121-5132.
Tuttle, M. E. & Brinson, H. F., 1986, Prediction of the long-term creep compliance of general composite laminates, Experimental Mechanics, vol. 26, n. 1,pp. 89-102.
Walrath, D. E., 1991, Viscoelastic response of a unidirectional composite containing two viscoelastic constituents, Experimental Mechanics, vol. 31, n. 2, pp. 111-117.
Yen, S. C. & Williamson, F. L., 1990, Accelerated characterization of creep response of an off-axis composite material, Composites Science and Technology, vol. 38, pp. 103-118.