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MATRIX DOMINATED STRESS/STRAIN BEHAVIOR IN POLYMERIC COMPOSITES: EFFECTS OF HOLD TIME, NONLINEARITY AND RATE DEPENDENCY

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Abstract

In order to understand matrix dominated behavior in laminated polymer matrix composites, an elastic/viscoplastic constitutive model was developed and used to predict stress/strain behavior of both off-axis and angle-ply symmetric laminates under in-plane, tensile axial loading.

The model was validated for short duration tests on graphite reinforced thermoplastic and bismaleimide composites at elevated temperatures. Short term stress relaxation and short term creep, strain rate sensitivity, and material nonlinearity were accurately accounted for. The testing times were extended to longer durations and periods of creep and stress relaxation were used to investigate the ability of the model to account for long term behavior. The model generally underestimated the total change in strain and stress for both long term creep and long term relaxation respectively.

Key Words

aging, material nonlinearity, constitutive model, elevated temperature, stress relaxation, creep, viscoplasticity

Nomenclature

a_{66} - potential function material parameter
E - elastic Young's modulus
G - elastic shear modulus
H - overstress
K - elastic/viscoplastic material parameter
m - elastic/viscoplastic material parameter
n - quasistatic elastic/plastic material parameter
S - compliance matrix
T_g - glass transition temperature
ϵ - strain
$\dot{\epsilon}$ - strain rate
γ - viscosity constant
ν - Poisson's ratio
σ - stress
$\dot{\sigma}$ - stress rate
$\bar{\sigma}$ - effective stress
$\dot{\bar{\sigma}}$ - effective stress rate
$\sigma^{\star}$ - quasistatic stress
$\bar{\sigma}^{\star}$ - effective quasistatic stress
Ψ - function defined in equation 11
Subscripts
1,2 = lamina material principal directions
Superscripts
e - elastic
qp - quasistatic plastic
vp - viscoplastic

Introduction

In the aerospace community, most laminated composite structures are designed for fiber dominated load paths. Despite this design criterion, a need exists for a fundamental understanding of the stress/strain relationships of laminates which are loaded along matrix dominated directions. This need arises from three possible sources. The first is the desire for analytical methods to perform local stress analyses of structural components (eg. discontinuities, stress concentrations, mechanically fastened joints, failure zones) which may lack fibers along the primary load path. Since most failure modes in composites are influenced by the state of stress in the matrix and fiber/matrix interface, accurate analytical methods are essential. The second need is for test methods to screen composites for matrix dominated behavior in order to assess the tendency towards material nonlinearity and rate dependent behavior. The third need arises from the requirement for designing composite structures with long operating lifetimes and developing associated accelerated test methods to verify the design. All of these needs may be critical for an aircraft such as a supersonic commercial transport due to the fact that the long term durability of composite materials, particularly at elevated temperatures, is directly dependent upon the stability and load carrying capability of the matrix.

As part of a research program at NASA to investigate long term durability issues in composites, test data on two model material systems, under conditions imposed by a Mach 2+ supersonic transport, were generated. Previous papers by the author [1], [2] have presented experimental methods for determining material constants and analytical models for advanced composites demonstrating elastic/viscoplastic behavior. The current research investigates the isothermal, matrix dominated, rate-dependent behavior of IM7/5260 [1] and IM7/8320[1] (graphite fiber reinforced thermoplastic and bismaleimide) composites under tension loading and verifies the predictive capabilities presented in [2] for short term behavior.

Long term exposure to load and temperature may need to be accounted for to accurately predict the effects on the time independent and time dependent mechanical behavior of matrix dominated polymer matrix composites. Recently, Gramoll et.al. [3] used time-temperature superposition principles to characterize the nonlinear thermoviscoelastic response on kevlar/epoxy laminates. In studies by Sullivan [4] and Hastie and Morris [5], physical aging of the polymer in the composite was found to change the laminate's viscoelastic properties. In these studies, Sullivan characterized physical aging in a glass reinforced thermoset composite while Hastie and Morris investigated the effect of physical aging on the

[1]The use of trade names in this paper does not constitute endorsement, either expressed or implied, by the National Aeronautics and Space Administration.

creep response of a carbon reinforced thermoplastic composite. Both of these investigations were limited to the linear viscoelastic range and developed shift factors to modify the elastic compliance matrix. They concluded that physical aging effects may occur in their study materials and that this aging may effect long term mechanical behavior.

Unlike these previous studies, the current research utilizes a elastic/viscoplastic analysis model capable of predicting nonlinear, short term time-dependent behavior at stress levels near tensile ultimate. The ability to extended the model to account for long term behavior will be discussed.

Constitutive Model Description

The elastic/viscoplastic model was developed [2] to approximate many of the experimentally observed phenomena in matrix dominated laminates over a range of typical operating temperatures. The model is considered to be macromechanical and phenomenological in form and was developed to account for variable strain rate loading and short term creep and stress relaxation. Other rate-dependent behavior such as strain recovery and aging were not accounted for explicitly in the model. Material constants were found from lamina level tests of unidirectional material. Temperature effects were handled through the variation of material properties with temperature [6]. An undamaged state was assumed for the laminate. Extensions to account for load and temperature induced damage would require incorporation of analysis schemes such as given by Martin [7].

For in-plane tension loading and assuming plane stress conditions, the total strain rate was assumed to be composed of elastic and viscoplastic components as follows

$$\{\dot{\epsilon}\} = \{\dot{\epsilon}^{e}\} + \{\dot{\epsilon}^{vp}\} \tag{1}$$

The individual constitutive relations were given as

$$\{\dot{\epsilon}^{e}\} = [S]^{e}\{\dot{\sigma}\} \qquad \text{elastic} \tag{2}$$

and

$$\{\dot{\epsilon}^{vp}\} = [S]^{vp}\{\dot{\sigma}\} \qquad \text{viscoplastic} \tag{3}$$

The elastic compliance term is linear and independent of stress level and was written as

$$[S]^{e} = \begin{bmatrix} \frac{1}{E_1} & \frac{-\nu_{12}}{E_1} & 0 \\ \frac{-\nu_{12}}{E_1} & \frac{1}{E_2} & 0 \\ 0 & 0 & \frac{1}{G_{12}} \end{bmatrix} \tag{4}$$

where the terms E_1, E_2, G_{12}, and ν_{12} are constants referenced to the material principal axis.

The viscoplastic compliance matrix was found [2] to be a nonlinear function of stress and was partially derived using the overstress concept, which has been investigated by other authors [8], [9], [10] for a variety of material systems. It was assumed that the viscoplastic compliance matrix can be decomposed into two separate components given by

$$[S]^{vp} = [S]^{vp'} + [S]^{vp''} \tag{5}$$

The first part of the viscoplastic compliance matrix was found to be

$$[S]^{vp'} = \gamma \frac{3}{2\bar{\sigma}} \left(\frac{\langle H \rangle}{K} \right)^{1/m} \begin{bmatrix} 0 & 0 & 0 \\ 0 & \frac{\sigma_{22}}{\dot{\sigma}_{22}} & 0 \\ 0 & 0 & \frac{2a_{66}\sigma_{12}}{\dot{\sigma}_{12}} \end{bmatrix} \tag{6}$$

and the second term ($[S]^{vp''}$) in the viscoplastic compliance matrix ($[S]^{vp}$) was found to be

$$[S]^{vp''} = \frac{\dot{\bar{\sigma}}(n-3)}{3\bar{\sigma}} \Psi \begin{bmatrix} 0 & 0 & 0 \\ 0 & \frac{\sigma_{22}^3}{\dot{\sigma}_{22}} & 0 \\ 0 & 0 & \frac{4a_{66}^2\sigma_{12}^3}{\dot{\sigma}_{12}} \end{bmatrix} + [S]^{qp} \tag{7}$$

where H is the scalar overstress given by

$$H = (\bar{\sigma} - \bar{\sigma}^\star) \tag{8}$$

the term $\sigma^\star$ is the rate-independent quasistatic stress, K and m are material constants found from tests [1], γ is a constant with magnitude of unity and units of $\frac{1}{sec.}$ and the term Ψ was given as

$$\Psi = \frac{9}{4} n A \bar{\sigma}^{(n-3)} \tag{9}$$

The effective stress ($\bar{\sigma}$) is

$$\bar{\sigma} = \sqrt{\frac{3}{2}(\sigma_{22}^2 + 2a_{66}\sigma_{12}^2)} \tag{10}$$

and $\dot{\bar{\sigma}}$ is the effective stress rate. The terms σ_{22} and σ_{12} are the inplane transverse and shear stress components, respectively. The material parameter a_{66} was found from axial tests of off-axis specimens.

The theoretical lower bound of the rate dependent behavior was represented by a rate-independent (quasistatic) elastic/plastic constitutive relation developed by Sun and Chen [11] where

$$\{d\epsilon^{qp}\} = [S]^{qp}\{d\sigma^\star\} \qquad \text{quasistatic plastic} \tag{11}$$

The quasistatic plastic compliance matrix was written as

$$[S]^{qp} = \Psi \begin{bmatrix} 0 & 0 & 0 \\ 0 & \sigma_{22}^2 & 0 \\ 0 & 0 & 4a_{66}^2\sigma_{12}^2 \end{bmatrix} \tag{12}$$

and all of the stress terms in equation 12 are quasistatic ($\sigma^\star$) and A and n are material constants found from test [1].

For an orthotropic plate, these equations reduce to a first order nonlinear differential equation [2] which was solved using a Runge-Kutta fourth order numerical integration technique. The equations can be solved using either stress or strain rates as input. Analysis of a laminated plate required incorporating the constitutive relations into a form of lamination theory [6] which was numerically solved directly by updating the compliance matrices using the previous stress state.

Materials Testing

Two polymer matrix composite material systems were investigated in this study. The first, an amorphous graphite/thermoplastic, was composed of Hercules IM7 fiber and Amoco 8320 matrix. The second material under study was a graphite/bismaleimide composed of Hercules IM7 fibers and Narmco 5260 matrix. The glass transition temperatures (T_g) were measured to be $219°C$ and $257°C$ for the IM7/8320 and IM7/5260, respectively.

Material properties required by the model were found from isothermal, axial tension tests of off-axis specimens [1]. Specimens used for verification of the model were both off-axis and angle-ply laminates. A rectangular test specimen geometry similar to that described in ASTM specification D3039-76 was used. These specimens consisted of twelve plys and measured 2.54 cm. by 24.1 cm. The six temperatures selected for study were $23°, 70°, 125°, 150°, 175°$ and $200°C$.

Using procedures described in [1], the elastic constants (E_1, ν_{12}, E_2 and G_{12}), the three elastic/plastic (a_{66}, A, n) and two elastic/viscoplastic (K, m) material parameters, were measured and correlated against test temperature [6]. In [6] a strong temperature dependency was found for the parameters A and K. All of the material constants and parameters used in this study are given in table 1. The elastic constants were based upon an average value found from two to three replicates for each temperature. The remaining five parameters were found from master curves for each material at each test temperature. Two to three specimens were used to form each master curve.

All tests, with the exception of the long term creep, were performed on a servo-hydraulic test machine capable of running predetermined load or strain history profiles. For the tests on the hydraulic test stand, heat was applied to the test specimens via an aluminum fixture which utilized resistance heaters and provided contact of the heated section over the ungripped section of the specimen. The long term creep tests were performed on mechanical, cantilever arm creep frames with circulating air ovens.

For all tests, strain was measured by using extensometers or high temperature foil strain gages [1]. Load, as measured by load cells, was converted to stress using the average cross-sectional area of the specimen prior to testing.

Results and Discussion

In order to verify the analytical model, recent test data was compared to predicted behavior for off-axis and angle-ply laminates. Creep, stress relaxation, material nonlinearity and the effects of strain rate were explored experimentally and analytically. The observed differences between short term and long term exposure at temperature will be discussed.

The test data and the predictions, presented in figures 1-8, were all conducted under isothermal conditions using axial tensile loading. Off-axis specimens with fiber orientations of 15°, 25°, 30°, 40° and angle ply layups of $[\pm 45]_{2s}$ and $[\pm 30]_{2s}$ were tested at temperatures ranging form $23°C$ to $200°C$. The results shown in figures 1-8 represent typical data and predictions for these types of tests.

Effects of Extended Hold Times

The effects of short and extended hold times were explored using off-axis tests. Figures 1a and 1b show test and predicted values for off-axis specimens which have been tested by loading under strain control to a predetermined level, unloading under load control to zero load, holding under load control, and reloading under strain control. Two hold periods, 6 seconds and 600 seconds, were utilized. The objective of this type of test was to examine the stress/strain behavior after the hold at zero load. Both tests and predictions show little difference between the tests with different hold times, with the same stress/strain path being followed by both the long and short hold cases. For both cases, the model describes both loading and unloading segments with reasonable accuracy.

Test and predicted short and long term stress relaxation results are shown in figures 2a and 2b. The plot of normalized stress versus time (figure 2a) shows less than 1 percent difference between test and prediction for the short term test. Extending the time for the same test (figure 2b) shows a tendency for both test and prediction to approach a limiting value of stress. However, the long term test has a difference of approximately 12 percent between test and prediction after 20 minutes of test time.

Test data and predicted behavior of short and long term creep are shown in figures 3a and 3b. As in the stress relaxation case above, good correlation (approximately 4 percent difference) between test and prediction values are found for the short term test while a comparison to the extended time data shows a percent difference approaching 15 percent after 200 minutes of test time.

For these extended time tests, two major sources of error may be present. The first source is due to the fact that the material property tests [1] were conducted on a time scale on the order of minutes. The implication is that to accurately predict the long term response, some long term test data may be needed to develop the material parameters required by the model. One potential problem with conducting such long term tests would be that since stress relaxation was used to develop the parameters, accurate long term stress relaxation tests would be required. These tests may be difficult to perform due to the likelihood of electrical and/or mechanical drifting of the strain controller during stress relaxation. Electrical drift may be caused by noise or environmental fluctuations, while mechanical drift is caused by test machine vibration and compliance of the load train. Long term creep tests, which are easier to perform, may be required to generate the long term data.

The second major source of error for an extended time test is the aging which may occur in the polymer matrix. As shown by Struik [12] aging, which may be chemical (irreversible), physical (reversible), or a combination of both, will change the compliance of the polymer over time. This change will be directly affected by the temperature history. Changes in properties due to aging will be material system dependent. Additional environmental factors, such as moisture, and oxygen, may also influence the rate of aging. Accounting for aging in the viscoplastic model will require an understanding of how aging influences the compliance matrices given in equations 4, 6, and 7.

Material Nonlinearity and Strain Rate Effects

Angle-ply laminates ($[\pm\theta]_n$) loaded under axial tension were used to explore material nonlinearity and strain rate effects. During the course of the test program, it was observed that some of the laminates exhibited matrix cracking. This cracking, which will alter the stress distribution in a laminate [13], can change the apparent stress/strain behavior. The laminates tested under this program were inspected for matrix cracks using microscopic examinations of the edges, and approximate threshold stress and temperature levels for crack initiation were established. All of the test results presented herein are for laminates which did not exhibit any matrix cracks.

The severity of the nonlinearity in the tested laminates was governed by the angle θ and test temperature. Figures 4a and 4b show test versus predicted stress/strain behavior for the highly matrix dominated angle-ply laminates $[\pm 45]_{2s}$ and $[\pm 30]_{2s}$ respectively. These laminates were tested using a constant strain rate of $200\mu\epsilon/sec.$ The model gave a good correlation for both type of laminates. The duration of these tests was short enough that aging effects during the test would not be significant. The expected effect of prior aging on such test results would be an increase in stiffness when compared to unaged specimens [12].

Using material properties from table 1 as input, figure 5 shows the capability of the numerical simulation to predict stress/strain behavior of angle-ply laminates for a variety of layups. Material nonlinearity is apparent for all layups except the laminate with 0° plys.

For exploring rate effects in strain controlled tests, strain rates were selected which were felt to be typical for aircraft structures and within the range of the test machine's capabilities. The effects of variable strain rates on the response of angle-ply laminates $[\pm 45]_{2s}$ and $[\pm 30]_{2s}$ are shown in figures 6a, 6b and 7a, 7b. Each figure depicts two tests, a constant rate ($200\mu\epsilon/sec.$) test and a jump rate ($10\mu\epsilon/sec.$ to $200\mu\epsilon/sec.$) test. Both test and predicted values are given for comparison. In general, the model appears to give a good prediction of the effect of jumps in strain rates. The jump from 10 to $200\mu\epsilon/sec.$ coincides with the constant $200\mu\epsilon/sec.$ rate test. This behavior is unlike some high temperature metallics which may exhibit dependence of stress level on strain rate history [14].

Using a numerical simulation, the predicted effect of varying applied strain rate is shown in figure 8. Each curve represents a prediction of laminate stress/strain behavior at a constant strain rate. Assuming that for a given strain level, predicting stresses higher than the test values gives a conservative estimate of peak stress, the use of the rate independent (quasistatic) expression may lead to unconservative estimates of the stress/strain response.

Summary

In order to address the need for an understanding of matrix dominated behavior in polymer matrix composites at elevated temperatures, an analytical model and associated test methods were previously developed. Using isothermal, short term stress relaxation testing, material parameters were developed for the model. The model, based upon elastic/viscoplastic constitutive relations was used to predict behavior of both off-axis and angle-ply laminates under in-plane tensile loading.

The model was validated for short duration tests at elevated temperatures. The model accounts for material nonlinearity, strain rate sensitivity, short term stress relaxation and short term creep. The testing times were extended for longer durations and periods of creep and stress relaxation were used to investigate the ability of the model to account for long term behavior. The effects of extended holds at zero load were negligible. The model generally underestimated by approximately 10 to 15 percent the total change in strain or stress for both long term creep and relaxation, respectively. In order for the present model to account for long term behavior, material parameter tests need to be extended to longer durations, and the analytical and test methods need to account for aging of the polymer matrix material.

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Material Type	°C	Elastic $E_1(GPa)$	$E_2(GPa)$	$G_{12}(GPa)$	ν_{12}	Elastic/Plastic a_{66}	$A(MPa)^{-n}$	n	Elastic/Viscoplastic $K(MPa)$	m
IM7/5260 Tension	23	152.8	8.7	5.2	0.30	0.60	2.91E-14	5.33	2.27E+05	0.95
	70	161.7	9.2	5.7	0.31	0.60	1.12E-13	5.33	1.51E+05	0.95
	125	156.5	8.8	5.3	0.36	0.60	8.29E-13	5.33	1.40E+05	0.95
	150	165.3	8.8	5.1	0.35	0.60	1.91E-12	5.33	1.19E+05	0.95
	175	136.4	7.7	5.1	0.30	0.60	1.15E-11	5.33	1.12E+05	0.95
	200	154.3	7.5	5.1	0.35	0.60	6.31E-11	5.33	1.03E+05	0.95
IM7/8320 Tension	23	157.9	7.1	4.3	0.32	0.30	1.94E-10	4.66	9.22E+03	0.81
	70	153.8	7.9	4.3	0.34	0.30	1.82E-10	4.66	2.10E+04	0.81
	125	142.0	7.5	4.7	0.35	0.30	6.25E-10	4.66	8.69E+03	0.81
	150	152.9	7.3	4.4	0.33	0.30	6.28E-10	4.66	7.43E+03	0.81
	175	153.9	7.2	3.4	0.32	0.30	3.92E-09	4.66	2.51E+04	0.81
	200	147.3	5.5	2.6	0.35	0.30	1.52E-06	4.66	6.78E+03	0.81

Table 1: Material properties and constants. [6]

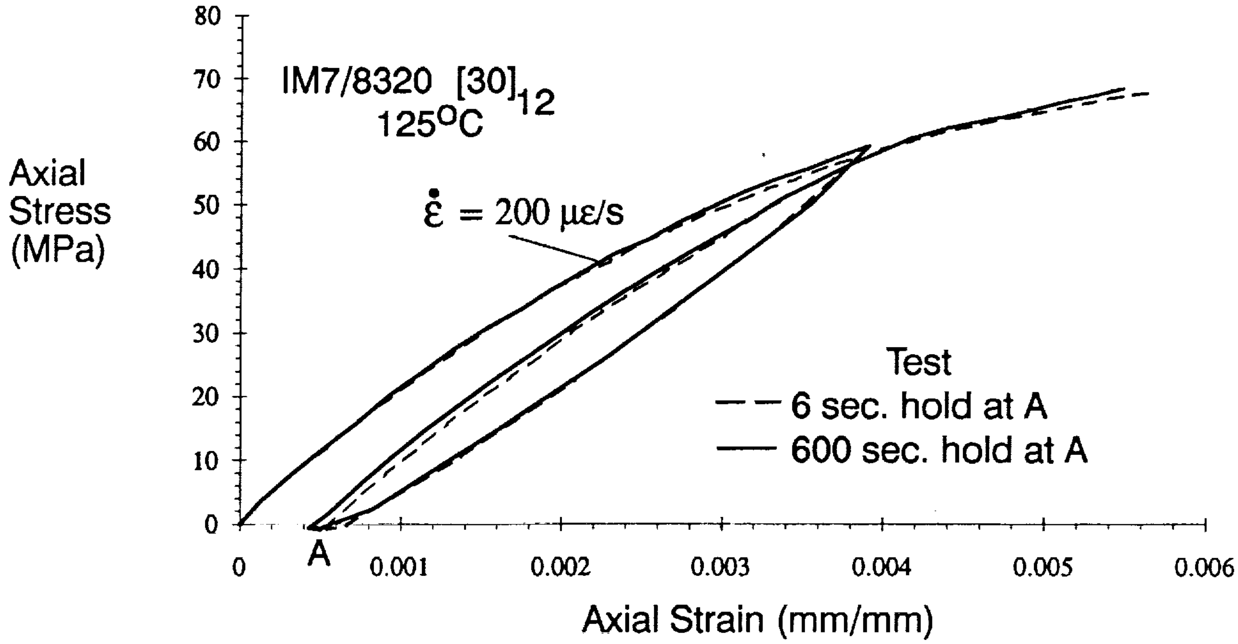

Figure1a. Test data for loading, unloading, recovery, loading sequence.

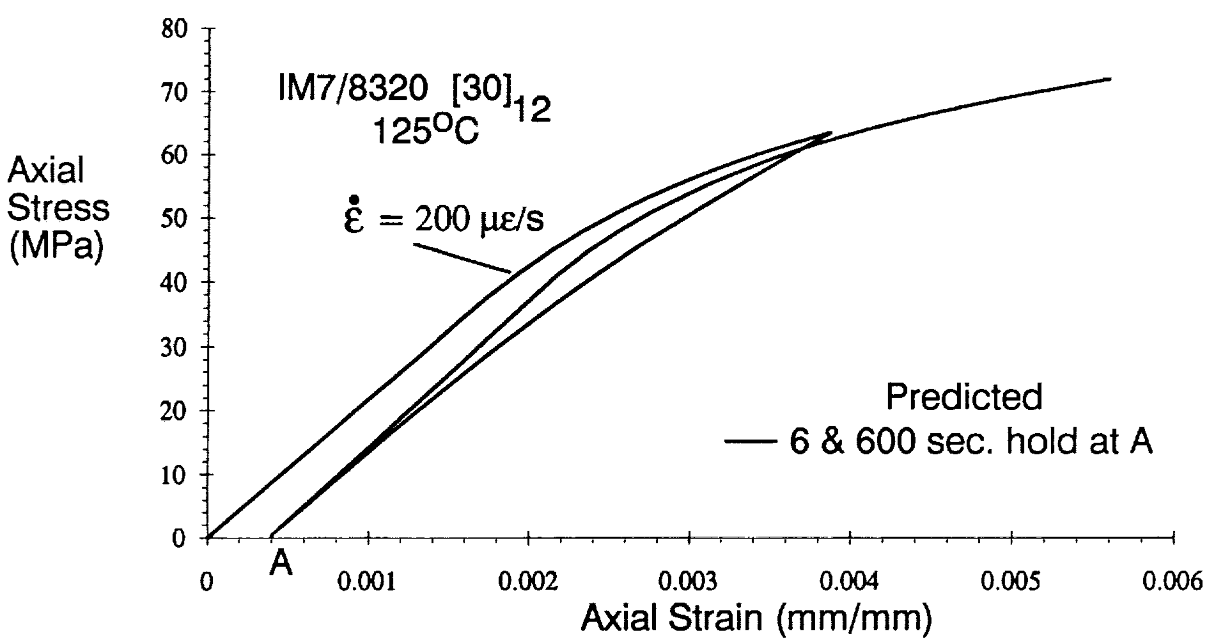

Figure1b. Prediction of loading, unloading, recovery, loading sequence.

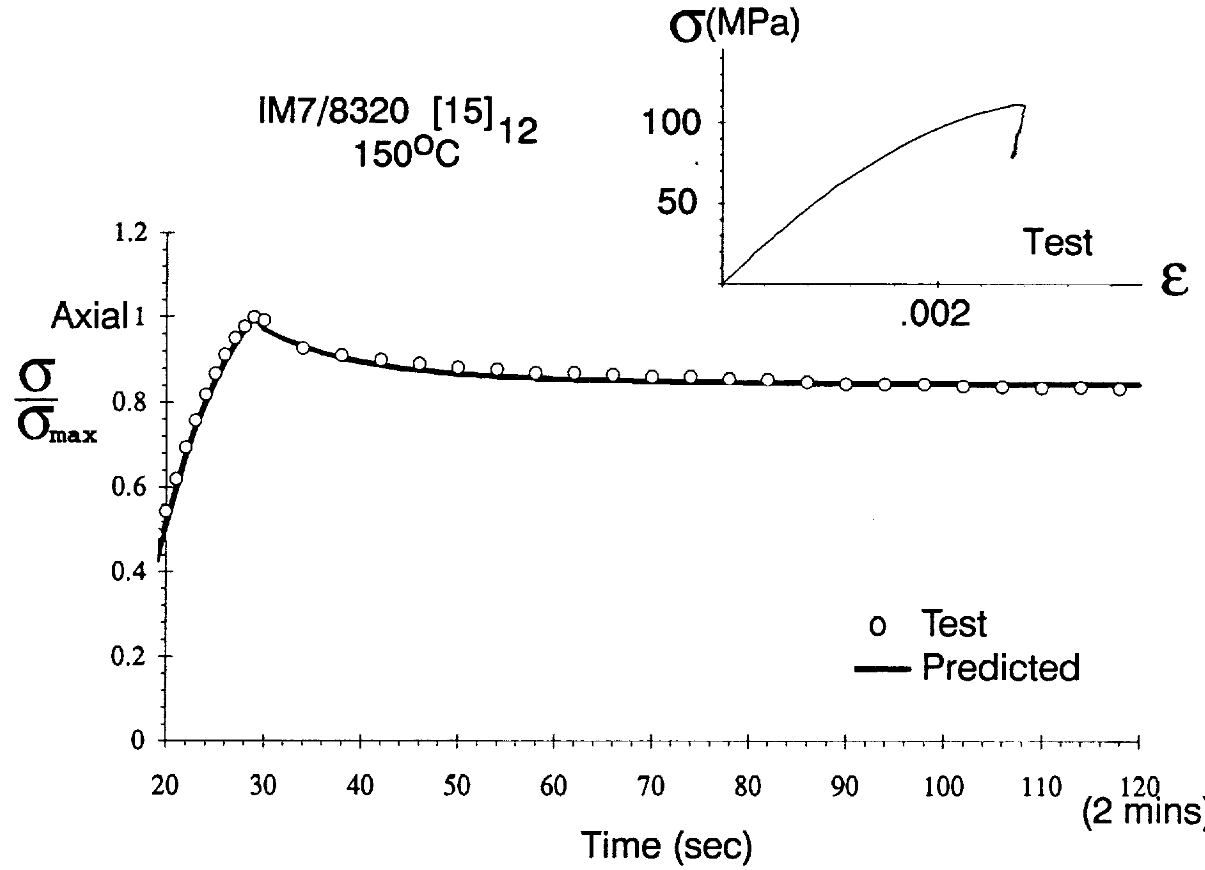

Figure 2a. Test data and prediction of short term stress relaxation.

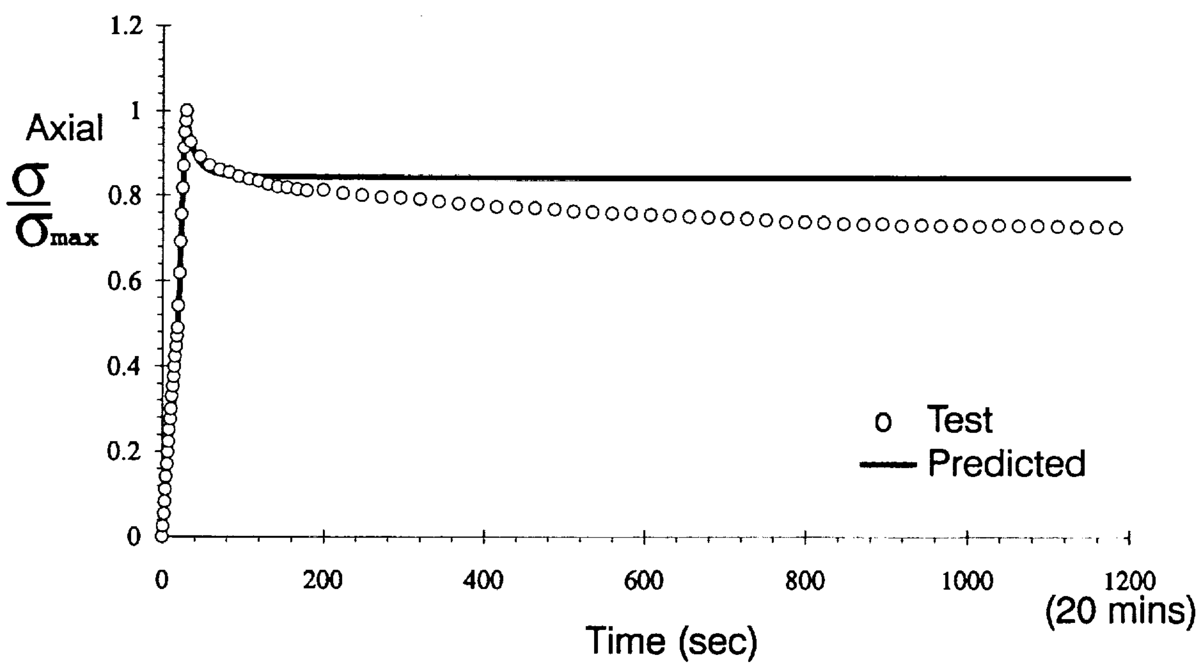

Figure 2b. Test data and prediction of long term stress relaxation.

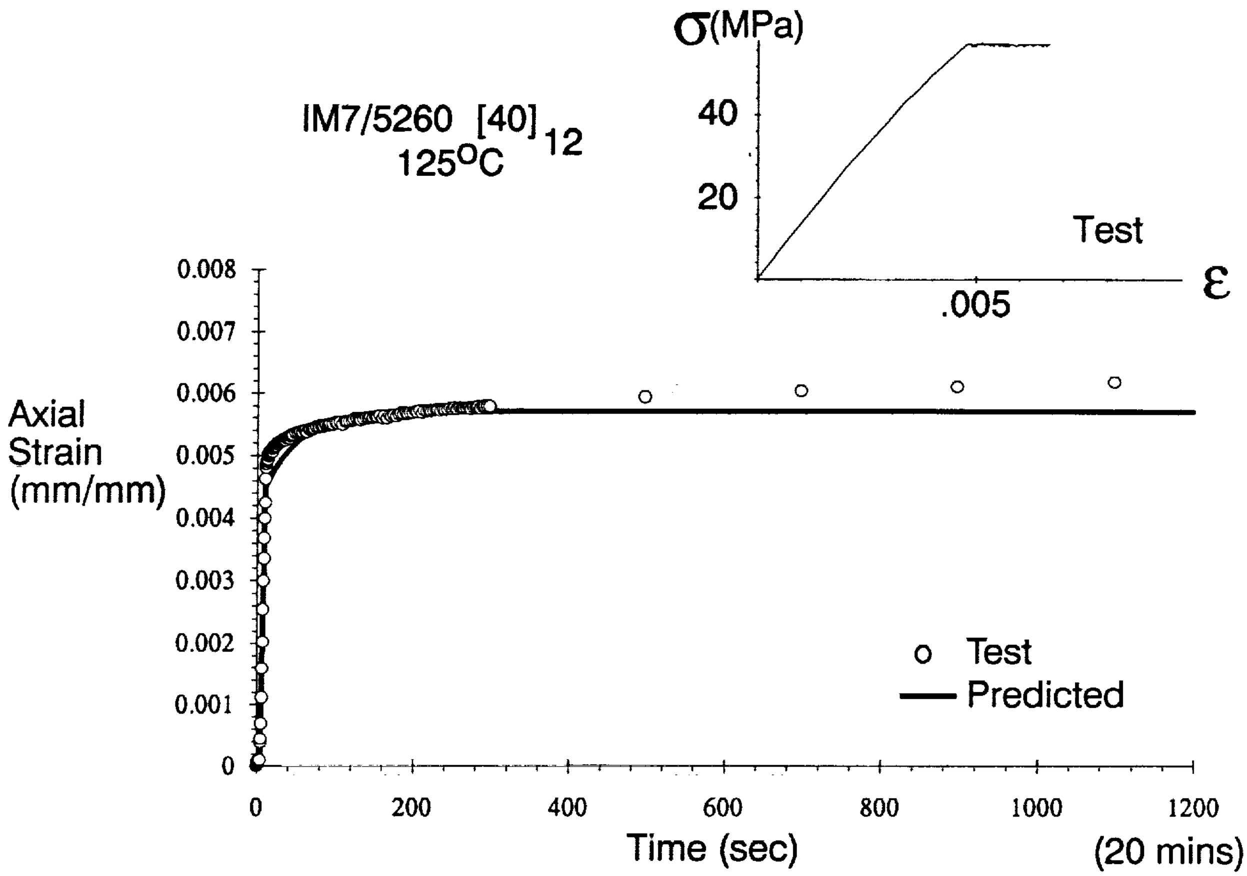

Figure 3a. Test data and prediction of short term creep.

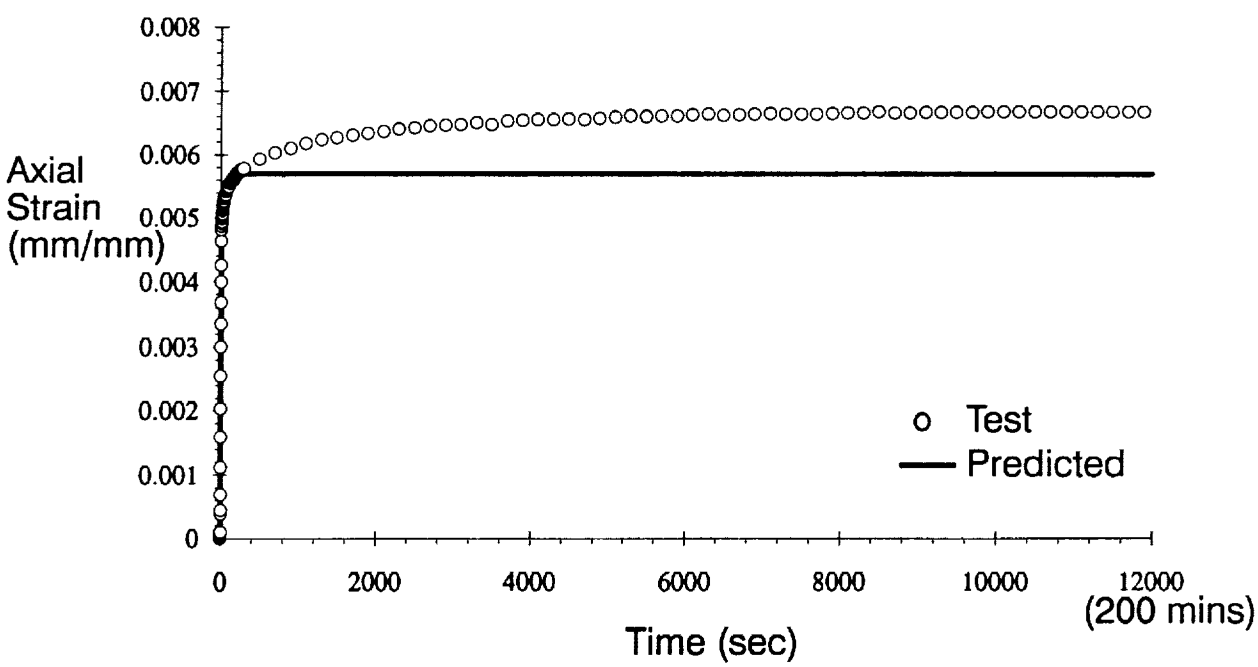

Figure 3b. Test data and prediction of long term creep.

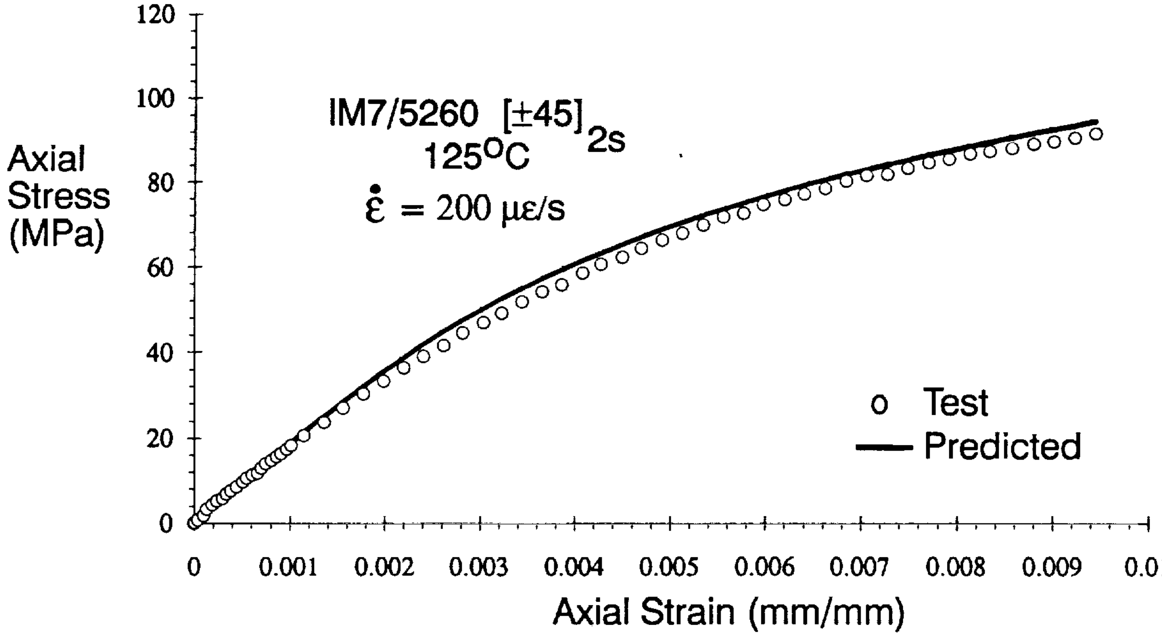

Figure 4a. Test versus prediction for an angle-ply laminate.

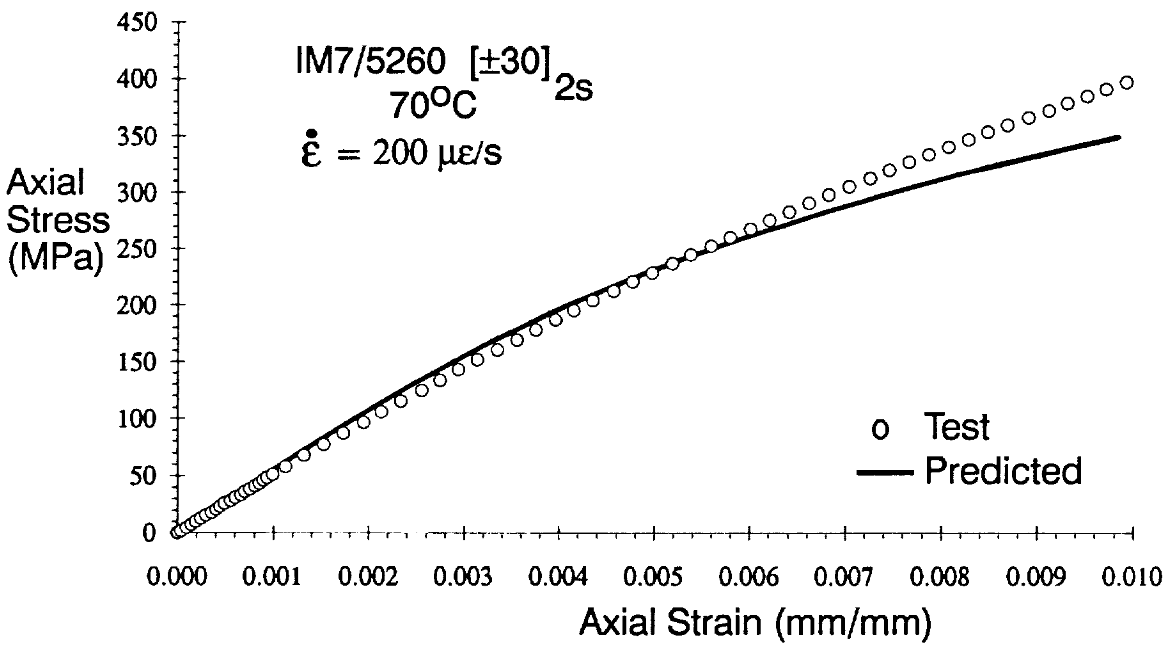

Figure 4b. Test versus prediction for an angle-ply laminate.

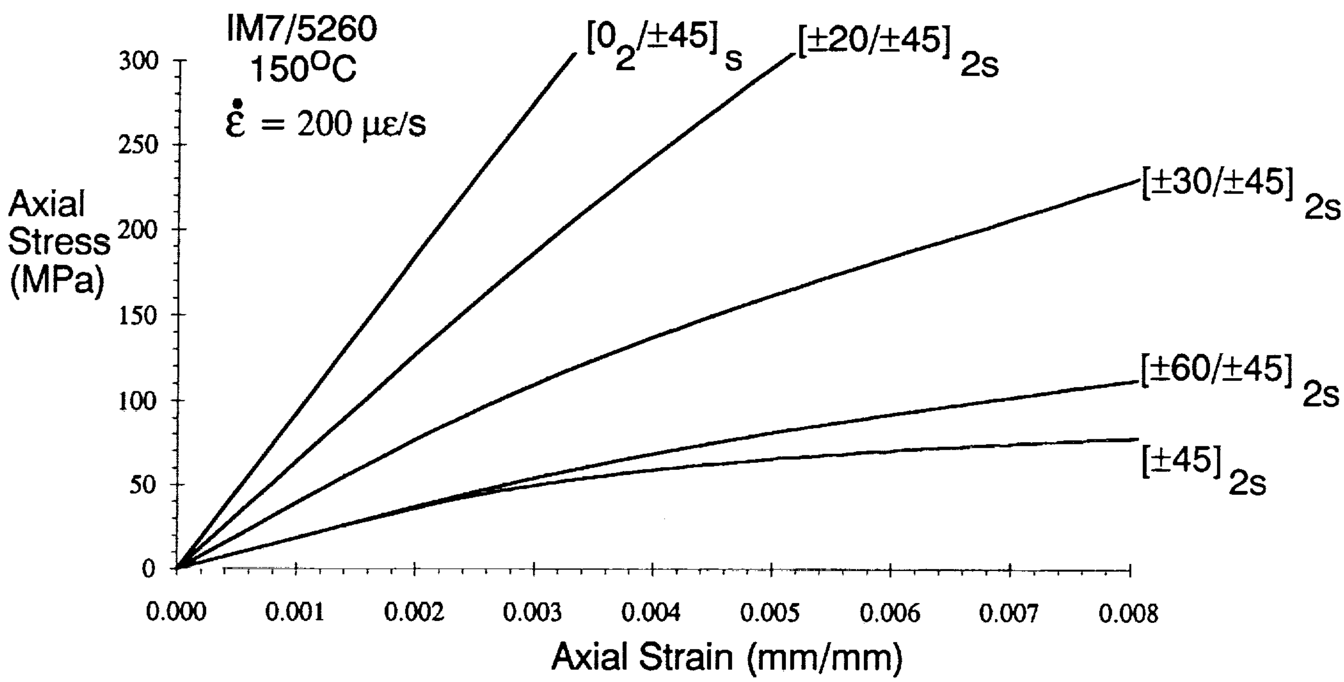

Figure 5. Predicted behavior of angle-ply laminates.

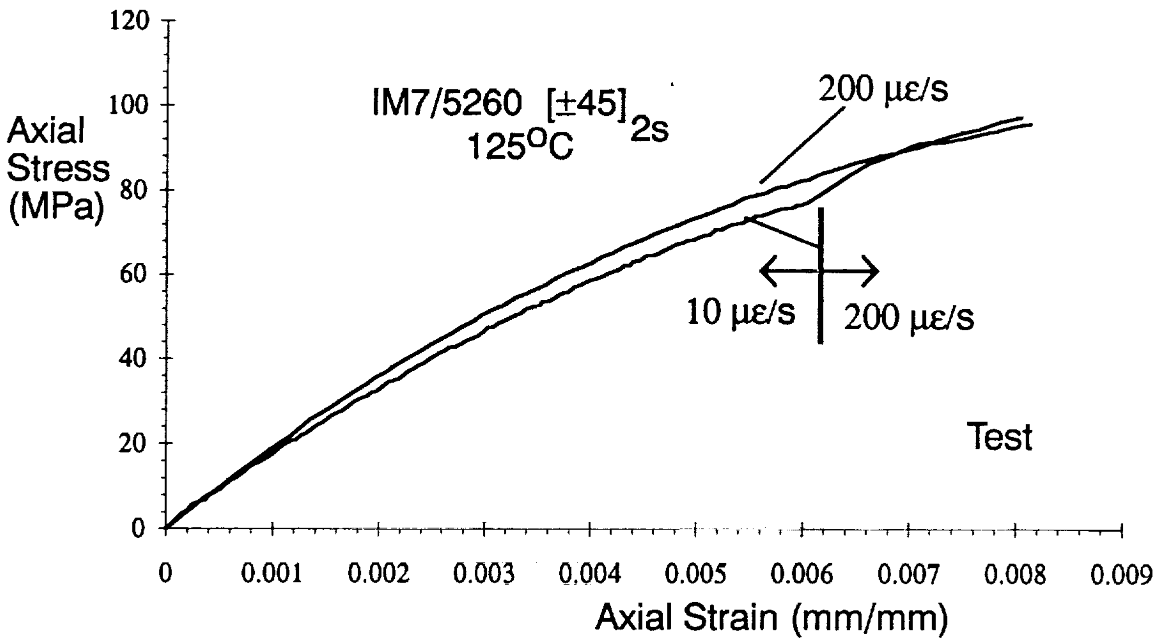

Figure 6a. Test data for constant rate and jump rate loading.

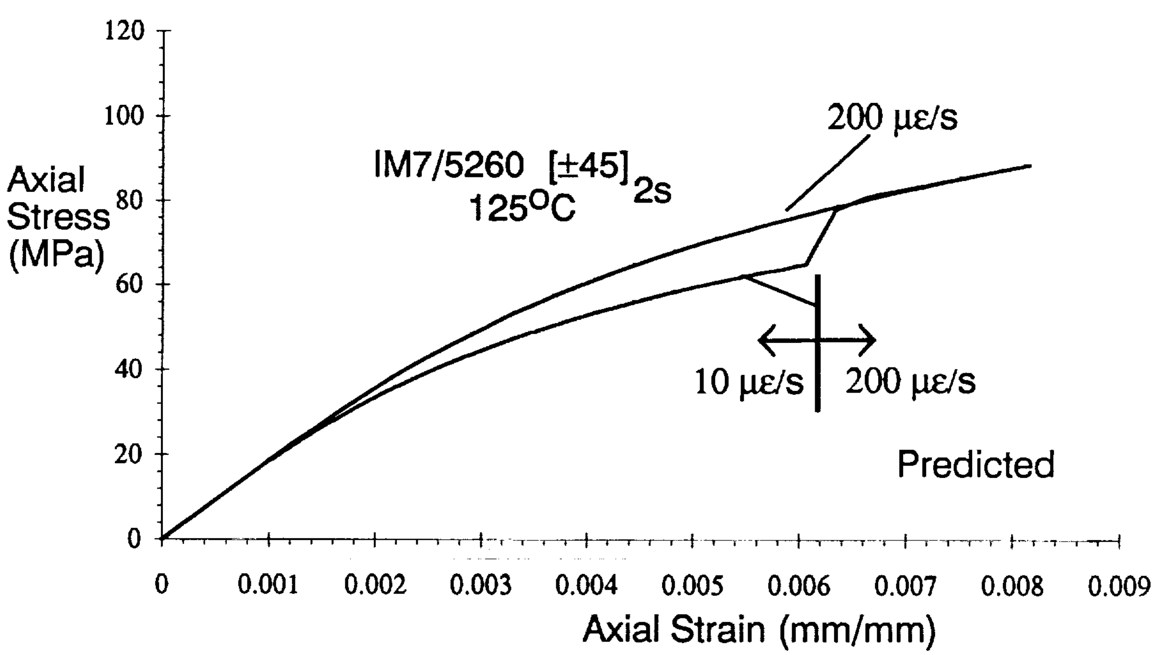

Figure 6b. Prediction for constant rate and jump rate loading.

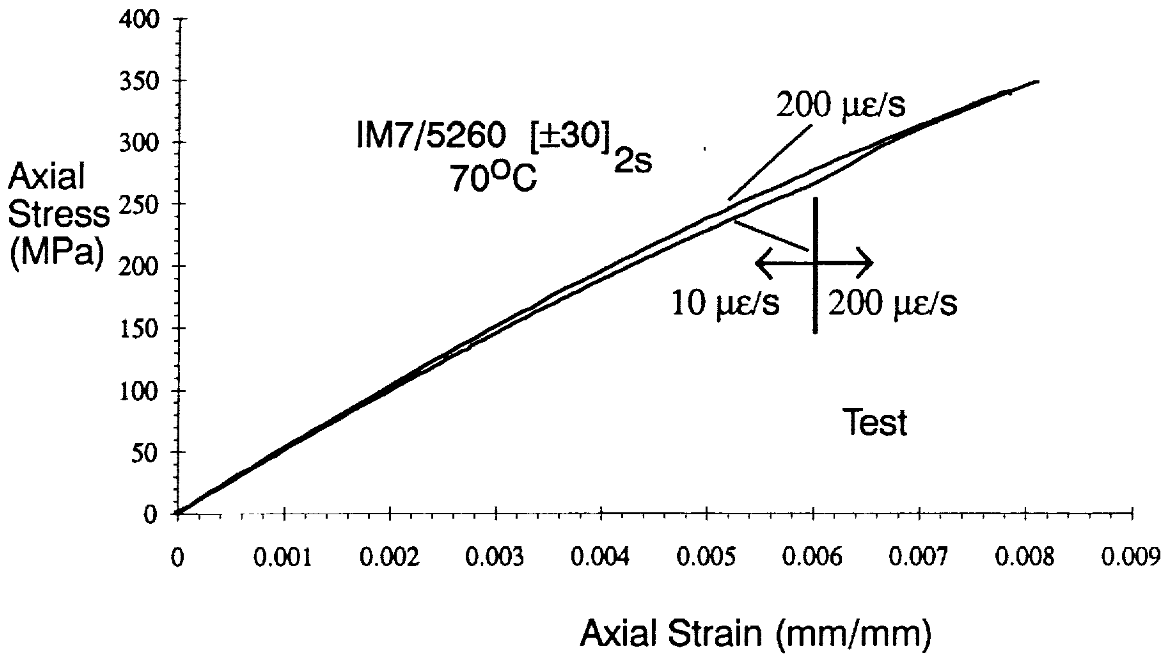

Figure 7a. Test data for constant rate and jump rate loading.

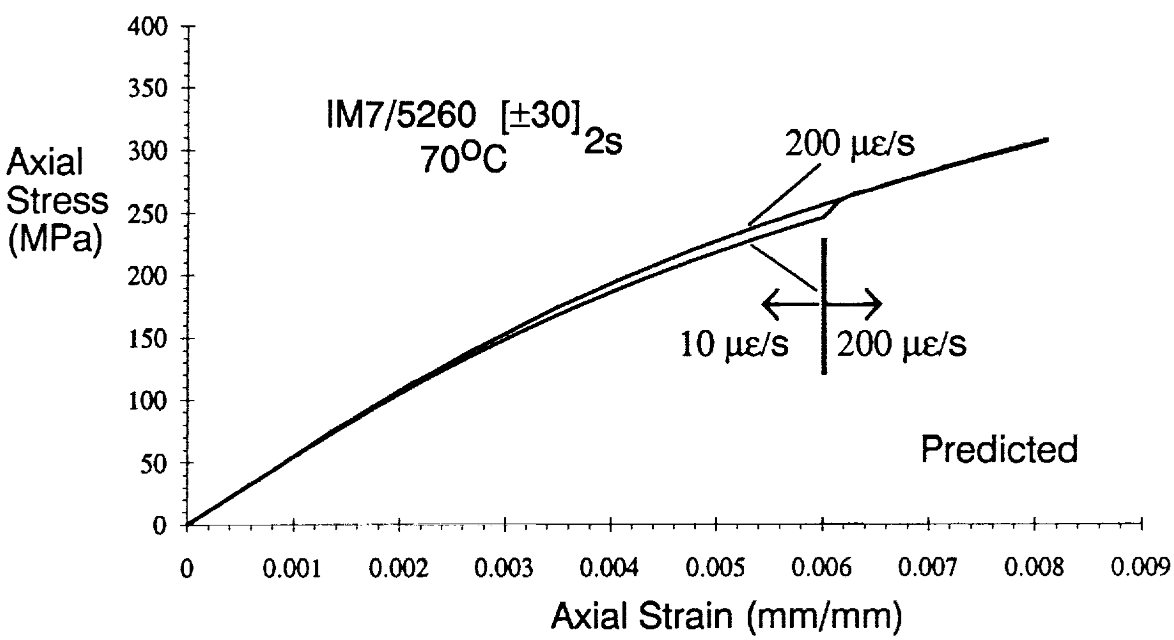

Figure 7b. Prediction for constant rate and jump rate loading.

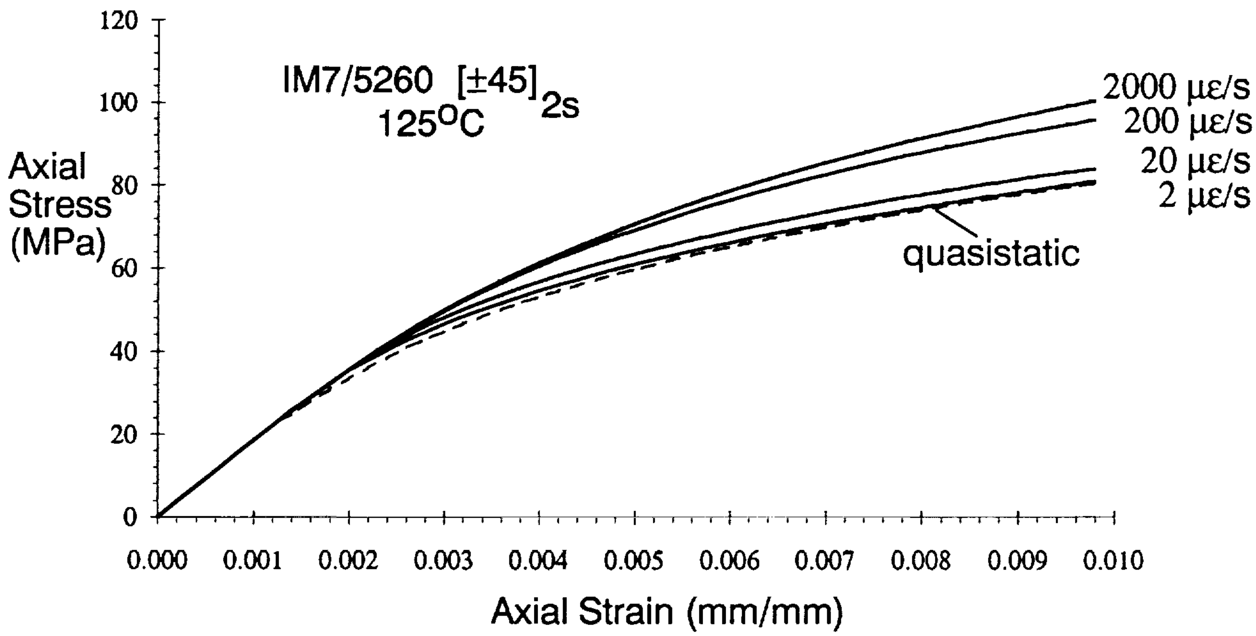

Figure 8. Predicted behavior of varying applied strain rate.

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In order to understand matrix dominated behavior in laminated polymer matrix composites, an elastic/viscoplastic constitutive model was developed and used to predict stress/strain behavior of both off-axis and angle-ply symmetric laminates under in-plane, tensile axial loading.

The model was validated for short duration tests at elevated temperatures. Short term stress relaxation and short term creep, strain rate sensitivity, and material nonlinearity were accounted for. The testing times were extended for longer durations and periods of creep and stress relaxation were used to investigate the ability of the model to account for long term behavior. The model generally underestimated the total change in strain and stress for both long term creep and long term relaxation respectively.

14. SUBJECT TERMS
Aging; Material nonlinearity; Constitutive model; Elevated temperature; Stress relaxation; Creep; Viscoplasticity

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