

# Predictive Creep Response of Linear Viscoelastic Graphite/Epoxy Composites Using the Laplace Transform Method

A. Rivera-Dominguez and W.M. Jordan

In this research project, creep and stress relaxation tests were run on angle-ply layups of a medium toughness graphite/epoxy system (Hexcel T3T145/F155). The creep and relaxation responses were represented with Maxwell/Kelvin-type models. Using a convolution integral and the Laplace transform method, a predictive creep response formulation was developed following a principle of virtual equilibrium state. Predictions of normalized creep compliance responses from short relaxation tests (based on a pseudo-elastic complementary approach) were compared to normal creep compliance values. As the fiber angles increased (moving farther away from the direction of the applied load), the difference between the compliances increased. This implies that, as toughness is increased, the accuracy of the pseudo-elastic method decreases. With the newer, tougher, resin systems, complementary approaches similar to the one used in this paper may be required.

## 1 Background

VISCOELASTIC materials display characteristics of elastic solids and viscous fluids.<sup>[1]</sup> They undergo a time-dependent and recoverable deformation process when they are under stress. The process is so slow that the viscoelastic response does not effect dynamic or inertial forces. Therefore, the material may be assumed to be under a quasi-static state.

This phenomenon varies according to the chemical composition and microstructure of the materials. Primary bonds such as ionic, covalent, and metallic bonds provide a stable material configuration. Secondary bonds, such as induced dipoles, polar molecules, molecular bridge, and coulombic forces, are weaker and more sensitive to the influence of physical forces.<sup>[2]</sup>

### 1.1 Superposition Integral

Because the viscoelastic response is a time-dependent process, viscoelastic formulae must obey hereditary integrity functions. For the particular case where the homogeneity and linear superposition conditions are satisfied, the function is shown as:<sup>[3]</sup>

$$F(t) = \int_{-\infty}^t F_Q(t-\tau) \frac{dI}{d\tau} d\tau \quad [1]$$

In the above equation,  $F_Q(t-\tau)$  is the response to an arbitrary input function  $I = Q(T-\tau)$ . Such integral is referred to as a hereditary integral because the linear functional  $F(t)$  depends on the history of the input  $I(t)$ . If the states of stress and strain are the engineering variables of interest, Eq 1 becomes Eq 2a and 2b shown below:<sup>[4]</sup>

$$\sigma(t) = \int_{-\infty}^t E(t-\tau) \frac{d\varepsilon}{d\tau} d\tau \quad [2a]$$

$$\varepsilon(t) = \int_{-\infty}^t D(t-\tau) \frac{d\sigma}{d\tau} d\tau \quad [2b]$$

In the above integrals, the engineering relaxation modulus  $E(t-\tau)$  and the engineering creep compliance  $D(t-\tau)$  replace  $F_Q(t-\tau)$ , respectively. These equations are based on the principle that the effects of sequential changes in stress or strain are additive. These equations are linear and are known as the superposition integrals.

### 1.2 Correspondence Principle

The correspondence principle of viscoelasticity states that at a fixed time, those displacement states which satisfy the force balance conditions give quasi-stationary functions similar to the constitutive relations of elasticity. The counterpart of this principle states that at a fixed time, those self-balancing force states which satisfy the strain-displacement conditions give quasi-stationary functions similar to the complementary constitutive relations of elasticity. This principle comes from the fact that the strain energy must always satisfy both the geometric compatibility and natural continuity conditions. The Laplace-transforms of the governing equations of viscoelasticity are analogous to the governing field equations of elasticity.

## 2 One-Dimensional Linear Viscoelastic Formulation of Dead Materials

The mutual correspondence between the stress and strain states in viscoelasticity depends on the material constitution. If the material behaves linearly, the hereditary function that correlates both states is a linear functional according to Eq 2. For a dead material, that is one unstressed at time zero, the lower limits of the integrals are replaced by zero.<sup>[3]</sup>

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## 2.1 Limiting Constant Values

When a material is loaded at time  $t = t_0$ , the creep response attains an extremum condition at that time. By using the Leibnitz's integral formula,<sup>[5]</sup> it can be shown that the derivative term of Eq 2a becomes Eq 3:

$$E(t - \tau) \frac{dD}{d\tau} = 0 \text{ for } t = t_0 = 0 \quad [3]$$

This implies the rate of change of the engineering creep compliance is zero at  $t = 0$ . Similarly, Eq 2b can be used to conclude the rate of change of the engineering relaxation modulus is also zero at that time. These extremum values are defined as  $D_0$  and  $E_0$ . For a material behaving like a viscoelastic solid, when time approaches infinite, the engineering creep compliance and the engineering relaxation modulus attain extremum conditions defined as  $D_\infty$  and  $E_\infty$ . With the help of these limiting values, the relaxation modulus and creep compliance may be represented in a normalized form as Eq 4a and 4b:

$$\theta(t) = \frac{E(t) - E_\infty}{E_0 - E_\infty} \quad [4a]$$

$$\eta(t) = \frac{D(t) - D_0}{D_\infty - D_0} \quad [4b]$$

## 2.2 Laplace Operation

The one-dimensional linear viscoelastic constitutive equation of a dead material can be transformed by using the Laplace operator<sup>[5]</sup> to get Eq 5:

$$D(s) E(s) = \frac{1}{s^2} \quad [5]$$

In this equation,  $s$  is the Laplace operation variable and  $D(s)$  and  $E(s)$  are the Laplace transform of the engineering creep compliance and the engineering relaxation modulus, respectively. Similarly, Eq 4 can be transformed with the Laplace operator, solved in terms of  $E(s)$  and  $D(s)$ , and combined with the transformed integral of Eq 5 to obtain Eq 6:

$$1 = \left[ \frac{D_0}{s} + (D_\infty - D_0) \eta(s) \right] \left[ \frac{E_\infty}{s} + (E_0 - E_\infty) \theta(s) \right] \quad [6]$$

The terms  $E(t)$  and  $D(t)$  are not independent properties, and unique relations among the limiting constant values  $D_0$  and  $E_0$ , and  $D_\infty$  and  $E_\infty$  must exist. These relations<sup>[6]</sup> are shown below in Eq 7 and 8:

$$D_0 E_0 = 1 \quad [7]$$

$$D_\infty E_\infty = 1 \quad [8]$$

After considering the conditions of Eq 7 and 8 and solving for  $\eta(s)$ , Eq 6 turns into the following transformed equation:

$$\eta(s) = \frac{1 - s \theta(s)}{s + \left[ \frac{(1 - r_e)}{r_e} \right] s^2 \theta(s)} \quad [9]$$

The term  $r_e$  is defined as the degree of pliability<sup>[6]</sup> in Eq 10:

$$r_e = \frac{E_\infty}{E_0} = \frac{D_0}{D_\infty} \quad [10]$$

Equation 9 can be evaluated under two special conditions, one for which  $r_e$  approaches unity and the other for which  $r_e$  is much smaller than one. If the pliability is close to one, that means that  $E_0$  and  $E_\infty$  are close to the same value. Considering the pliability to be close to one, the inverse of the Laplace operator transforms Eq 9 to form Eq 11:

$$\eta(t) = 1 - \theta(t) \quad [11]$$

This equation reveals that if the material behaves elastically rigid, both viscoelastic functions are always complementary. On the other hand, the case for which the material is highly pliable, for  $r_e \ll 1$ , Eq 9 is closely equivalent to Eq 12 shown below:

$$\eta(s) = \frac{r_e [1 - s \theta(s)]}{s [r_e + s \theta(s)]} \quad [12]$$

For the particular case where  $\theta(t) = \exp(-at)$ , that is, for a classical Maxwell/Kelvin model function,<sup>[6]</sup> the Laplace inverse<sup>[5]</sup> is shown in Eq 13:

$$\eta(t) = 1 - e^{-\mu t} \quad [13]$$

where

$$\mu \equiv a r_e \quad [14]$$

Because  $r_e$  is less than one, Eq 13 and 14 show that the time constant of the creep function is bigger than the time constant of the relaxation function. This means, in practical purposes, that creep tests last longer to attain compliant equilibrium than relaxation tests. The final conclusion applies for general values of  $r_e < 1$  and other model functions, and it gives the basis to conclude the following principle. Given two equivalent viscoelastic systems initially stressed to the same strain energy level, the material extension caused by fixed deformation will more quickly attain a virtual equilibrium state than will the one with the fixed force balance condition.

The patterns of the viscoelastic response depend on the material constitution and microstructure. They obey the superposition integral if the material has an admissible strain and complementary energy function. This implies a continuous strain function. Equation 9 may be used to express the transformed normalized creep function by the transformed normalized relaxation function. After applying the inverse of the Laplace transform operator, this results in Eq 15:

$$\eta(t) = \mathcal{L}^{-1} \left\{ \frac{1 - s \theta(s)}{s + \left[ \frac{(1 - r_e)}{r_e} \right] s^2 \theta(s)} \right\} \quad [15]$$

If the degree of pliability approaches the ideal value of one, the material behaves like a pure elastic solid and not like a viscoelastic material. Then the solution of Eq 15 is the same as the elastic complementary solution (Eq 11). In the case for which the viscoelastic solid is highly pliable, the relaxation modulus

exhibits a faster viscoelastic response than does the creep compliance. Intermediate generic solutions are expected to occur for any fractional value of  $r_e$ . From the above principle of virtual equilibrium state, Eq 15 serves to predict the creep response of a highly pliable viscoelastic solid from short relaxation tests.

### 2.3 Physical Models of Material Behavior

Although the above equations describe how the relaxation modulus and creep compliance are related to each other, they do not describe the actual shapes of the modulus and creep curves. Frequently, experimental data are curve fit to what are called Maxwell/Kelvin equations.<sup>[3-6]</sup> These models represent the material behavior using the responses of springs and dashpots. A Maxwell model represents the material as if it were a spring and dashpot arranged in series. With the spring, stress is proportional to strain. With the dashpot, stress is proportional to the strain rate. A Kelvin model is one in which the spring and dashpot are arranged in parallel. The Maxwell/Kelvin models used in this data reduction are those in which a Maxwell element and Kelvin element are arranged in series.

## 3 Experimental Procedure

### 3.1 Material Used

The graphite/epoxy prepreg used was the Hexcel T3T145/F155 system. It was fabricated into 10- by 6-in. panels and cured according to the manufacturer's suggested cure cycle.<sup>[7,8]</sup> The F155 resin is a medium crosslink density, medium toughness resin.<sup>[9,10]</sup> This is a commercial system that has been studied by several researchers.<sup>[7-10]</sup> Its toughness has been increased by the presence of small rubber particle additions.<sup>[9,10]</sup> The rubber particles occupy 5.6% (by volume) of the resin.<sup>[10]</sup> The panel layups were balanced and symmetric with the following stacking sequence:

$$[X_2 / -X_4 / X_2]_S$$

In this study,  $X$  could have values of 0, 30, 45, or 60°. The total number of plies in each laminate was 16.

### 3.2 Mechanical Test Procedure

Two different types of mechanical tests were performed during this project. They were creep tests and stress relaxation tests. These tests were conducted on a model 810 MTS tensile machine according to ASTM standard D638. All mechanical tests were done at room temperature, which in this laboratory was nominally 24 °C. The data were recorded on an IBM personal computer using a Keithley Series 500 system.

Viscoelastic data were obtained from the graphite-epoxy composite laminates tested in either constant load (for creep tests) or constant displacement (for relaxation tests) using the same MTS system. This allowed for a comparison of the creep and relaxation responses for the different angle-ply laminates. All tests ran for 15 min.

The samples were strain-gaged so that the strain could be determined accurately. Micro-Measurements CEA resistance-type strain gages were used. They are a general-purpose strain

gage capable of measuring up to 3% strain (in either tension or compression). The creep tests on angle-ply samples were loaded up to approximately 60% of their ultimate tensile strength. The relaxation tests were also loaded up to approximately 60% of their ultimate tensile strength. The displacement that corresponds to this load was maintained during the relaxation test.

### 3.3 Data Reduction

From the creep/relaxation data, samples were selected for the following times: 1, 10, 90, 300, and 900 seconds. This allowed for a stress-strain space, isochronous correlation. The data set was reduced to a characteristic value for each sampling time by using the least square linear regression with origin collocated (LSLROC) method. The data had an average correlation factor of approximately 0.98.<sup>[6]</sup>

These reduced data were curved-fitted to a Maxwell/Kelvin model function by using the LSLR method. This is documented by Ref 6. The function was determined for each laminate. Virtual limiting values were obtained by evaluating it at zero time and infinite time. The above approach was used for relaxation as well as for creep data, and they were compared according to Eq 11.

## 4 Results and Conclusions

Figures 1 through 4 show two different versions of the normalized creep compliance for the material system studied in this project. The pseudo-elastic results are those obtained by Eq 11, that is, the complement of the normalized relaxation modulus. This equation assumed that the pliability ( $r_e$ ) is close to one. The results labeled "normal creep" are those determined directly from the creep data.

The complementary solution, developed from the viscoelastic formulation, presents an alternate way to compare the vis-

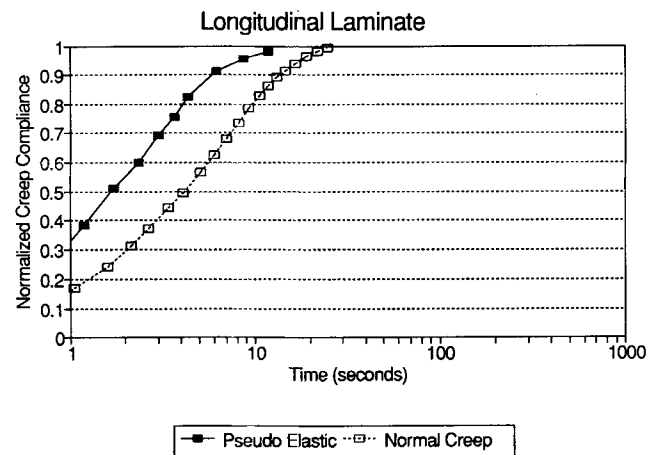
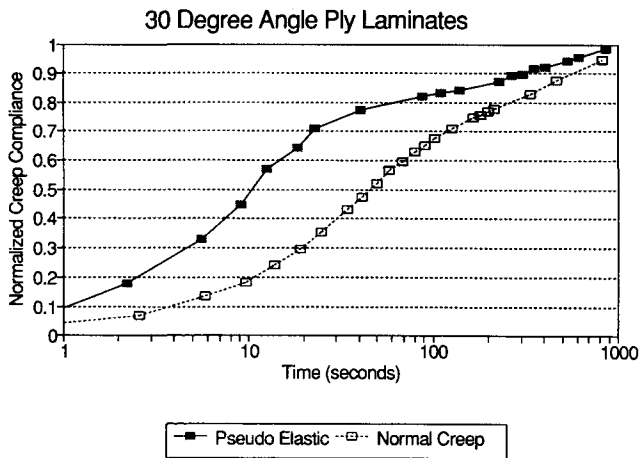
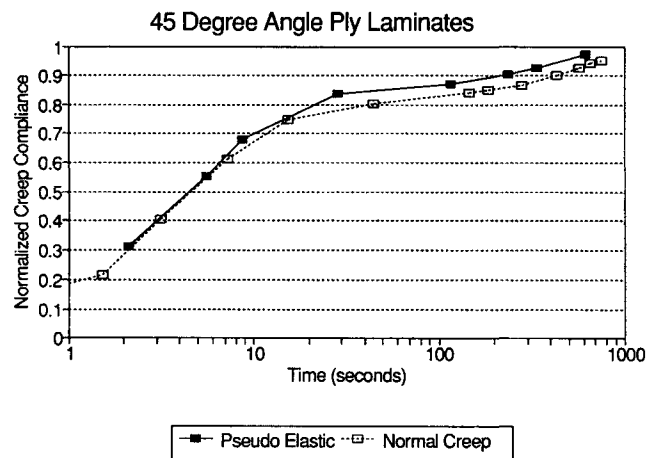


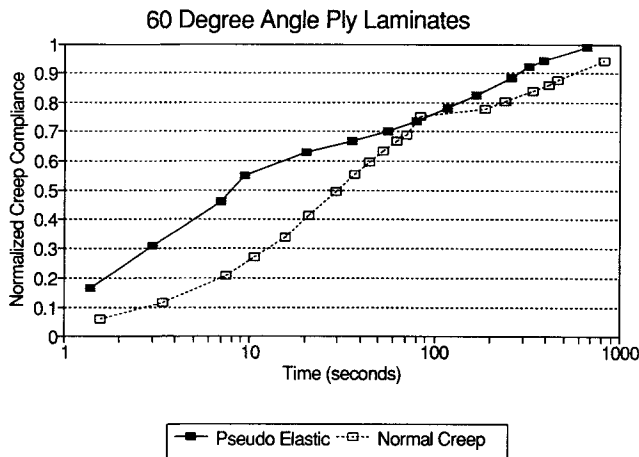
Fig. 1 Creep of T3T145/F155 longitudinal laminate. Normalized creep compliance is plotted versus the logarithm of time using two different calculation methods. The normalized pseudo-elastic compliance was calculated using the complement of the normalized relaxation modulus.



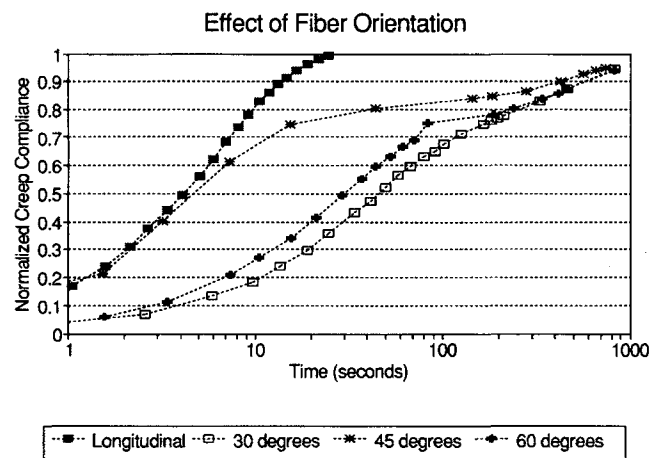
**Fig. 2** Creep of T3T145/F155 laminate using a 30° angle-ply layup. Normalized creep compliance is plotted versus the logarithm of time using two different calculation methods. The normalized pseudo-elastic compliance was calculated using the complement of the normalized relaxation modulus.



**Fig. 3** Creep of T3T145/F155 laminate using a 45° angle-ply layup. Normalized creep compliance is plotted versus the logarithm of time using two different calculation methods. The normalized pseudo-elastic compliance was calculated using the complement of the normalized relaxation modulus.



**Fig. 4** Creep of T3T145/F155 laminate using a 60° angle-ply layup. Normalized creep compliance is plotted versus the logarithm of time using two different calculation methods. The normalized pseudo-elastic compliance was calculated using the complement of the normalized relaxation modulus.



**Fig. 5** Normalized creep compliance of T3T145/F155 laminates using angle-ply layups with 0, 30, 45, and 60° angles.

coelastic responses of creep and relaxation tests. The results of the composite laminates vary considerably according to the fiber orientation. For longitudinal laminates, the pseudo-elastic solution was close to the normal creep solution, after a period of only 10 seconds duration. This is shown in Fig. 1. The deviation of the creep response from the pseudo-elastic complementary solution increases when the fibers are at an angle to the applied load. For the  $\pm 30$ ,  $\pm 45$ , and  $\pm 60^\circ$  laminates, the results using these two methods differed even more significantly. They only converged after a period of up to 1000 seconds had elapsed. These are depicted in Fig. 2, 3, and 4. The material behaves more elastically as the fiber orientation angle decreases, with an optimal case for which the fiber angle is zero.

This result of increasing viscoelasticity as the angle moves away from zero degrees is in accord with previous results found by Jordan *et al.*<sup>[7,9]</sup> They found that as the fibers move away from a unidirectional orientation, the resin is allowed to deform more, which results in a material that is less elastic.

Figures 1 through 4 illustrate that the normalized creep response curves are always below the pseudo-elastic complementary curves. The pseudo-elastic results are the complementary part of the normalized relaxation curves. This means that the relaxation response reaches a virtual equilibrium state faster than does the creep response. This reinforces the principle of the virtual equilibrium state of viscoelasticity. However, further analytical schemes must be developed to model predic-

tive creep responses to explore the natural effects accompanied between the creep compliance and the relaxation modulus for different degree of pliability ( $r_e$ ).

Figure 5 shows the normalized creep results for the four layups studied in this project. The orientation that was most off-axis was the one that had the lowest compliance. This is to be expected, because there is less fiber resistance to deformation at this angle. The off-axis layups not only have lower creep compliances, but they also respond more slowly than the layups that are aligned more closely with the load direction.

The approach used in this project shows the validity of using relatively short relaxation tests to predict the creep response of a medium toughness epoxy-based composite material. As tougher resins are developed, they will behave less and less in an elastic fashion. As was shown with the  $\pm 60^\circ$  laminates (the toughest orientation tested in this study), the usefulness of the pseudo-elastic method diminishes with increased toughness. With these tougher materials, methods such as the ones based on relaxation tests described above will be required. For these materials, the pliability ( $r_e$ ) may be much less than one, and Eq 15 must be used.

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