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Stress Relaxation and Viscoelastic Creep of Polymer Composites: A Critical Review of Models, Methods, and Mechanisms

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Abstract

This review provides a comprehensive and critical assessment of the literature on stress relaxation and viscoelastic creep in polymer composites, covering constitutive modeling, relaxation-creep interconversion, energy dissipation, time-dependent damage, and temperature effects. Three major constitutive approaches are compared and evaluated: the Prony series, the Standard Linear Solid model with finite loading rate correction, and fractional-order viscoelastic models. For interconversion, the Boltzmann convolution integral, direct SLS prediction formulas, and the engineering $J \approx 1/E$ approximation are critically assessed. A time-dependent damage framework D(t) extended from Weibull fatigue damage is discussed. Key gaps in the literature are identified, and future research directions are proposed.

Keywords

Stress Relaxation; Viscoelastic Creep; Constitutive Modeling; Interconversion; Energy Dissipation; Damage Mechanics; Time-Temperature Superposition.

1. Introduction

Fiber-reinforced polymer composites are increasingly used in load-bearing structural applications across the automotive, aerospace, and energy sectors. Materials such as short glass fiber-reinforced polyamide (PA66+30%GF) are valued for their high specific strength, design flexibility, and cost-effectiveness. However, the polymeric matrix inherently exhibits time-dependent mechanical behavior, manifested as stress relaxation under constant strain and creep under constant stress[1],[2]. Understanding and quantifying these viscoelastic phenomena is essential for reliable long-term performance prediction and durability assessment.

Stress relaxation experiments offer significant practical advantages over creep tests. They require shorter testing durations, simpler load control, and are less susceptible to specimen rupture during testing. These advantages have motivated substantial research into methods for predicting creep behavior from relaxation data[3],[4]. This challenge spans multiple disciplines: continuum mechanics provides the constitutive framework, polymer physics explains the underlying molecular mechanisms, and experimental mechanics provides the data necessary for model validation.

This review critically examines the literature across five interconnected themes: constitutive models for stress relaxation, methods for converting relaxation data to creep predictions, energy dissipation analysis from a thermodynamic perspective, time-dependent damage characterization, and temperature effects on long-term behavior. The emphasis throughout is on comparative evaluation of different approaches and identification of remaining knowledge gaps.

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2. Constitutive Models for Stress Relaxation

2.1. Prony Series (Generalized Maxwell Model)

The Prony series is the most widely used mathematical representation of the relaxation modulus in both research and commercial finite element software [5]. It represents the relaxation modulus E(t) as a sum of exponential decay functions, each corresponding to a Maxwell element (a spring and dashpot in series) connected in parallel:

$$E(t) = E_{\infty} + \sum_{i=1}^{n} E_i e^{-t/\tau_i} \tag{1}$$

where E_inf is the equilibrium modulus, and E_i and tau_i are the relaxation modulus and relaxation time of the i-th Maxwell element. For polymer composites, n=3 to 4 terms are typically required to achieve $R^2 \geq 0.99$, resulting in 2n+1=7 to 9 parameters. The Prony series offers excellent mathematical flexibility and is directly compatible with commercial finite element codes such as ABAQUS and ANSYS. However, the individual parameters lack clear physical interpretation; they are essentially fitting coefficients rather than physically meaningful material constants. Furthermore, the formulation implicitly assumes instantaneous step loading, which does not reflect real experimental conditions where loading takes finite time [4]. The large number of parameters can also lead to over-fitting and poor extrapolation behavior.

2.2. Standard Linear Solid Model with Finite Loading Rate

The Standard Linear Solid (SLS) model provides a more physically transparent representation of viscoelastic behavior using two springs (E_1, E_2) and one dashpot (eta). The relaxation time constant is $\tau_R = \eta / E_2$,and the creep time constant is $\tau_C = \eta(E_1 + E_2)/(E_1 E_2)$. Each parameter has clear physical meaning: E_1 is the long-term equilibrium stiffness, E_2 is the viscoelastic contribution, and eta characterizes the viscous flow resistance.

A major advance was made by Lin et al.[4], who derived a modified SLS formulation that explicitly accounts for the finite loading rate during experiments. Their formulation introduces the loading strain rate r as a parameter:

$$\sigma(t) = [E_1 r(\tau_C - \tau_R)(1 - e^{-\varepsilon_0/\tau_r r})]e^{-(t/\tau_R)^k} + E_1\varepsilon_0 \tag{2}$$

The stretched exponential parameter k ($0 < k \leq 1$) provides additional flexibility for capturing non-exponential relaxation. Through an extensive finite element study involving 63 combinations of 9 materials and 7 loading rates, Lin et al. [4] showed that the conventional step-loading assumption leads to parameter identification errors exceeding 15%. Their finite loading rate formulation reduced these errors to below 5% across all tested conditions. This finding has significant practical implications, as most laboratory experiments are conducted at moderate loading rates where the step-loading assumption is not valid.

2.3. Fractional-Order Viscoelastic Models

Fractional calculus has emerged as a powerful mathematical framework for viscoelasticity over the past two decades [7],[8]. Unlike classical integer-order models, fractional-order models introduce a parameter beta that continuously spans the range from purely elastic (β=0) to purely viscous (β=1) behavior. This provides a physically intuitive measure of the material viscoelastic character, with beta correlating with microstructural features such as crosslink density.

Liu et al. [8]developed the Parallel Fractional-Order Viscoelastic (PFOV) model, which addresses the singularity at t=0 inherent in basic fractional formulations by introducing a finite

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loading time t_0. The PFOV model requires only 4 parameters while achieving $R^2 \geq 0.99$ for unidirectional prepreg relaxation data, outperforming the 7-parameter Prony series in terms of both accuracy and parameter parsimony. Despite these advantages, fractional-order models have seen limited adoption in commercial software due to the need for specialized numerical methods.

2.4. Comparative Evaluation

Table 1. Comparative assessment of constitutive models for stress relaxation.

Criterion	Prony Series	SLS + Finite Rate	Fractional PFOV
Parameter count	2n+1 (typically 7)	4	4
Loading rate accounted for?	No	Yes, explicitly	Partially via t_0
Physical interpretability	Low	High (E1, E2, eta)	High (β)
Typical fit quality	$R^2 \geq 0.99$	$R^2 \geq 0.98$	$R^2 \geq 0.99$
Software support	Excellent	Moderate	Limited
Best suited for	FEA implementation	Parameter identification	Microstructural insight

The SLS model with finite loading rate correction represents the best compromise for engineering applications, combining good accuracy, clear physical interpretation, and explicit treatment of realistic loading conditions. Fractional models are preferred where parameter parsimony is critical and microstructural insight is desired. The Prony series remains the most practical choice for commercial software implementation.

3. Relaxation-Creep Interconversion

The ability to predict creep behavior from stress relaxation data is of great practical importance. The most rigorous method is based on the Boltzmann superposition principle [9], which establishes a fundamental relationship between the relaxation modulus E(t) and the creep compliance J(t) through a Volterra integral equation. The Boltzmann approach is mathematically exact within linear viscoelasticity and provides the highest accuracy, but requires high-quality relaxation data over the full time range of interest and careful numerical implementation.

When material parameters have been identified using the SLS model, creep strain can be predicted directly using a closed-form expression without numerical integration[4]. This approach offers a balance of accuracy (typically within 5% of the exact solution) and computational efficiency.

A simpler but less reliable approach is the engineering approximation $J \approx 1/E$. While exact at t=0 and $t \to \infty$, it systematically overestimates creep compliance at intermediate times. For materials with less than 20% stress drop, the error is typically below 5%, but for materials with stress drops exceeding 50% (common near Tg), the error can reach 15-30%.

Table 2. Comparison of relaxation-creep interconversion methods.

Method	Accuracy	Computational Cost	Data Required
Boltzmann convolution	Highest	Moderate (numerical)	Full E(t) curve
Direct SLS formula	High ($\pm$ 5%)	Low (closed-form)	SLS parameters
$J \approx 1/E$	Low (error to 30%)	None	E(t) at same time

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4. Energy Dissipation Analysis

Energy dissipation provides a physically insightful perspective on viscoelastic behavior that complements the stress-strain approach. Amabili et al. [10]developed a Rational Extended Thermodynamics (RET) framework in which the internal energy is decomposed into a reversible elastic part and an irreversible viscous part. A key result is that the rate of energy dissipation is proportional to the square of the viscous stress, providing a direct link between the measurable stress relaxation response and the underlying thermodynamic processes.

A particularly significant finding from RET analysis is that cross-coupling terms between different relaxation modes account for a substantial portion (up to 44%) of the total dissipated energy. This indicates that relaxation mechanisms in polymer composites cannot be treated independently; their interactions are essential for accurate energy dissipation characterization. The three-stage dissipation process observed in practice (rapid initial, moderate intermediate, slow long-term) corresponds to molecular relaxation mechanisms at different length scales, from short-segment motion to long-chain relaxation and fiber-matrix interface phenomena.

5. Time-Dependent Damage Mechanics

In classical fatigue and fracture mechanics, damage is typically expressed as a function of the number of loading cycles using the Weibull distribution[11]. This approach has been highly successful for metals, where damage accumulates with each loading cycle and accelerates as cracks propagate.

For viscoelastic materials subjected to sustained loading, the natural independent variable is time rather than cycle number. Extending the Weibull concept to the time domain gives the time-dependent damage model[12],[13]:

$$D(t) = 1 - \frac{E(t)}{E_0} = 1 - \exp\left[-(\frac{t}{\eta})^m \right] \tag{3}$$

The shape parameter m provides a quantitative distinction between different classes of materials. Metals typically exhibit m>1, indicating damage acceleration as cracks propagate. Viscoelastic polymers, by contrast, exhibit m<1, indicating damage deceleration as relaxation mechanisms progressively slow down. For PA66+30%GF near its glass transition temperature, experimental data yield m ≈ 0.4, confirming this decelerating damage characteristic. This fundamental difference has important implications for remaining life prediction: polymer components may experience most of their damage early in their service life, with degradation rates decreasing over time.

Coupled creep-damage models, which combine the viscoelastic creep compliance with the time-dependent damage variable using the effective stress concept, provide a unified framework for predicting the full creep curve from initial loading through tertiary acceleration to final failure. Zhang et al. [14]successfully applied such an approach to asphalt mixtures, demonstrating excellent agreement with experimental data across multiple temperatures and stress levels.

6. Temperature Effects and Long-Term Prediction

The time-temperature superposition (TTS) principle is one of the most powerful tools for predicting long-term viscoelastic behavior. Based on the WLF equation[15], TTS enables the construction of master curves covering many decades of time from short-term tests at multiple elevated temperatures. For semi-crystalline polymers operating below Tg, the Arrhenius equation is often more appropriate.

A remarkable demonstration was provided by Mahdavipour et al.[16], who showed that 150-minute creep tests on PA66 fibers at multiple temperatures could predict 19-year creep

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behavior with R2 > 0.99. The apparent activation energy was determined as 234 kJ/mol. The Larson-Miller parameter [17] provides a complementary approach for creep rupture life prediction. The combination of TTS master curves with time-dependent damage models represents a promising direction for long-term durability assessment, though experimental validation remains limited.

7. Critical Discussion and Future Directions

Several important gaps in the current literature merit attention. First, constitutive modeling, interconversion, and damage characterization have largely developed as separate research streams. A unified framework that couples advanced viscoelastic models with time-dependent damage would enable consistent prediction of the full creep curve from relaxation data alone, but such a framework remains to be developed.

Second, direct experimental validation of relaxation-creep interconversion for specific engineering materials is surprisingly sparse. Most studies validate against numerical solutions rather than experimental creep data. Comprehensive validation programs across multiple material systems are needed to establish reliability.

Third, the combined effects of temperature and damage on long-term viscoelastic behavior are poorly understood. Experimental data at multiple temperatures coupled with systematic damage characterization would enable temperature-dependent damage models.

Fourth, standardized testing and data analysis protocols for relaxation-creep interconversion are needed, analogous to existing standards for creep (ASTM D2990) and stress relaxation (ASTM E328).

8. Conclusion

This review has critically examined the literature on stress relaxation and viscoelastic creep of polymer composites. The principal findings are as follows.

(1) Among the three major constitutive approaches, the SLS model with finite loading rate correction offers the best combination of accuracy, physical interpretability, and practical applicability for engineering use. The fractional-order PFOV model achieves superior parameter parsimony and is recommended for research applications where microstructural insight is desired.

(2) The Boltzmann convolution integral remains the most accurate method for relaxation-creep interconversion, but the direct SLS formula provides a convenient closed-form alternative when model parameters are available. The $J \approx 1/E$ approximation should be used with caution and only for materials exhibiting weak relaxation.

(3) The time-dependent damage model $D(t) = 1 - \frac{E(t)}{E_0}$ extends classical fatigue damage concepts to viscoelastic materials. The shape parameter m < 1 provides a quantitative signature distinguishing polymer damage evolution from metal-like behavior.

(4) Time-temperature superposition enables long-term behavior prediction from short-term tests, representing a powerful tool for engineering practice. However, validation across diverse material systems and coupling with damage evolution require further investigation.

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