Extension of Fractional Calculus Theory in Modeling the Memory Effects of Viscoelastic Polymeric Materials

Xiaojing Yu*

University of Sanya, Sanya, 572000, China
**Corresponding author: 1309048280@qq.com*

Abstract: *The complex history-dependent behavior exhibited by viscoelastic polymeric materials, known as the memory effect, has long posed a challenge for the accurate prediction of their mechanical properties. Classical linear viscoelasticity theory has inherent theoretical limitations in describing such non-local and cross-scale characteristics. Fractional calculus operators, due to their intrinsic non-locality and hereditary properties, provide a more physically consistent framework for the mathematical description of memory effects. This paper systematically elaborates on the extension of this theory in related modeling. First, it analyzes the physical mechanisms of memory effects and the shortcomings of classical models, establishing the physical significance of fractional operators. Subsequently, a generalized fractional constitutive model is constructed, revealing the cross-scale evolution characteristics of the Mittag-Leffler-type memory kernel, and extending the description to multi-field coupling and non-linear theories. Finally, the strategies for physical identification of model parameters, numerical implementation approaches, and current frontier challenges are discussed. Research indicates that fractional calculus theory can effectively and uniformly describe the memory behavior of polymeric materials from microscopic motion to macroscopic response through continuous relaxation spectrum characterization and flexible memory weight assignment, laying a significant foundation for the development of a new generation of viscoelastic constitutive theories.*

Keywords: *Fractional Calculus; Viscoelasticity; Polymeric Materials; Memory Effect; Constitutive Theory; Mittag-Leffler Function; Non-locality*

Introduction

The mechanical behavior of viscoelastic polymeric materials is strongly dependent on their loading and deformation history. This memory effect is rooted in the complex relaxation processes of molecular segments within the material. Although classical integral constitutive relations based on the Boltzmann superposition principle can initially describe linear memory behavior, their discrete relaxation spectrum representation and limited capacity to describe nonlinear and multi-field coupling effects constrain their predictive capability and physical depth under complex conditions. Therefore, developing a constitutive theory that closely links microscopic physical mechanisms while accurately characterizing macroscopic non-local and cross-scale memory behavior has become an important need in this field. Fractional calculus theory extends the order of differentiation and integration from integers to arbitrary real numbers. The non-locality of its operators naturally aligns with the history-dependent nature of memory effects, providing a powerful mathematical tool for constructing such constitutive relations. This study aims to systematically extend the application of fractional calculus theory in modeling the memory effects of viscoelastic polymeric materials. By constructing a generalized fractional model with clear physical meaning, analyzing its memory evolution laws, and exploring its descriptive framework under complex environments, it seeks to deepen the understanding of the time-dependent mechanical behavior of materials and promote the development of related mechanical modeling theories.

1. Theoretical Foundation and Mathematical Characterization of Memory Effects in Viscoelastic Polymeric Materials

1.1 Physical Mechanism of Viscoelasticity and Memory Effects in Polymeric Materials

Viscoelastic polymeric materials exhibit mechanical behaviors that combine the solid-like characteristics of elasticity with the fluid-like characteristics of viscosity. This dual nature originates from their unique molecular structure and motion patterns. The internal structure of these materials consists of long-chain molecules entangled with one another, and the motion of chain segments involves broad-spectrum relaxation processes ranging from local side-group rotation to the reptation of entire molecular chains. Under external forces, these chain segments must overcome energy barriers to rearrange and adapt to new conformations. This process is accompanied by energy storage and dissipation, which macroscopically manifest as nonlinear time-dependent responses such as stress relaxation, creep, and hysteresis. The current state of the material depends not only on the instantaneous load but is also profoundly influenced by the entire load history. This historical dependence forms the physical core of the memory effect.

The memory effect, in essence, reflects the non-local response capability of a polymer system to past stimuli. The evolution of a molecular chain from one conformational state to another requires time, and its evolutionary path depends on the initial state and the thermodynamic processes it has undergone. This path dependency means the material can "remember" its previous stress or strain history and subsequently release or superimpose its influence in its response. Therefore, the memory effect transcends simple instantaneous cause-and-effect relationships, embodying the complex, distributed energy storage and transfer mechanisms within polymeric materials. It serves as a crucial bridge connecting their microscopic molecular motion to macroscopic mechanical properties.

1.2 Description of Memory Effects by Classical Integral Constitutive Relations and Their Limitations

The linear viscoelasticity theory based on the Boltzmann superposition principle provides a classical framework for the mathematical description of memory effects. By introducing relaxation modulus or creep compliance functions, this theory expresses the stress or strain at the current moment as a convolution integral of the entire history of strain or stress changes. The integral kernel functions, namely the relaxation or creep functions, serve here as memory weights, quantifying the contribution of stimuli at each past moment to the current response. This type of integral constitutive relation possesses clear physical meaning and mathematical simplicity when describing linear memory effects under small deformations, successfully establishing a direct link between material functions and memory effects[1].

However, classical integral constitutive models reveal theoretical limitations when dealing with the complex memory behavior of practical polymeric materials. Their linear superposition assumption struggles to accurately capture nonlinear memory effects under large deformations or complex loading paths. These models typically rely on specific functional forms (such as the sum of exponentials in the generalized Maxwell model) fitted from experimental data. Such functions often correspond to discrete relaxation time spectra, making it difficult to naturally represent the inherent continuous relaxation spectrum characteristics of polymers. More importantly, the description of memory effects in this class of models is "formalistic." Their memory kernel functions lack a direct physical mapping to the complex internal dynamical processes of the material (such as cooperative segmental motion and multi-scale coupling), which may lead to failure when predicting the long-term memory behavior of novel materials or under extreme conditions.

1.3 Basic Operators of Fractional Calculus and Their Physical Significance

Fractional calculus represents a natural extension of integer-order differential and integral operators to arbitrary orders. Its core operators primarily include the Riemann-Liouville definition and the Caputo definition. The Riemann-Liouville fractional derivative is defined by performing integer-order differentiation on a function followed by fractional-order integration, while the Caputo derivative reverses this sequence, giving it a clearer form for physical problems involving initial conditions. These two definitions are interrelated through specific transformations and both can reduce to the classical integer-order case. The non-locality of fractional differential and integral operators is their most fundamental mathematical characteristic, meaning the result of the operation depends on the historical

information of the function over the entire domain interval.

This non-locality provides an ideal mathematical tool for precisely characterizing the memory effects of viscoelastic materials. Integer-order derivatives only reflect the local rate of change of a function within an infinitesimal region, whereas fractional-order derivatives perform a weighted summation of the function's historical evolution through an integral kernel featuring power-law decay. This weighting method highly aligns in mathematical form with the physical phenomenon observed in polymeric materials where the relaxation modulus decays according to a power law over time (as revealed by principles such as time-temperature superposition). Consequently, the order of the fractional operator can be regarded as an intrinsic parameter characterizing the material's "memory strength" or "degree of non-locality." A lower order indicates a slower decay of historical influence and a more persistent "memory" of the material, thereby opening new pathways for unified modeling from the level of molecular motion to macroscopic mechanical behavior.

2. Extension of the Fractional-Order Theoretical Framework for Modeling Memory Effects

2.1 Principles for Constructing Generalized Fractional-Order Constitutive Models

The core principle for constructing generalized fractional-order constitutive models lies in systematically replacing the integer-order time derivative operators in classical constitutive relations with fractional-order differential operators, guided by physical mechanisms. This substitution is not merely a mathematical formal transformation; it is based on the inherent non-locality of fractional operators, which can more fundamentally capture the hereditary and memory characteristics of polymer chain segment motion. By introducing fractional-order stress and strain derivatives, models such as the fractional Maxwell model, the fractional Kelvin-Voigt model, and their generalized combinations can be constructed. Their constitutive equations manifest as fractional differential equations in the time domain and correspond to complex modulus expressions with fractional power-law characteristics in the complex frequency domain. The fractional order within the model establishes a direct correlation with the material's intrinsic relaxation time distribution spectrum. A non-integer differential order essentially corresponds to a continuous relaxation spectrum distribution with power-law features, thereby overcoming the limitation of classical models that rely on fitting discrete exponential series.

This construction principle is further reflected in the unified description of multi-level relaxation mechanisms. The memory effects in polymeric materials span multiple time scales, ranging from the local motion of chain segments to the disentanglement of entire molecular chains. Models employing a single fractional order may be insufficient to cover the entire relaxation spectrum. Consequently, fractional-element ladder models based on multiple fractional derivatives, or distributed-order fractional differential equations, have been developed. In these extended models, different fractional elements or operators of different orders respectively characterize relaxation processes at distinct time scales or governed by different physical mechanisms. The integrated modeling of the entire broadband memory behavior is achieved through parallel or series configurations. This modular construction method enhances the structural flexibility of the models and their characterization capability for complex material systems[2].

2.2 Evolution and Response Characteristics of Fractional-Order Memory Kernel Functions

The memory kernel function corresponding to the fractional-order constitutive model typically manifests as the Mittag-Leffler function or its generalized forms, a core special function in fractional differential equation theory. Compared to classical exponential decay kernel functions, the Mittag-Leffler function kernel exhibits a unique biphasic decay characteristic: it approximates a power-law decay in the short-time domain and asymptotically approaches an exponential decay in the long-time domain. This evolutionary characteristic precisely aligns with the measured relaxation behavior of many viscoelastic polymeric materials over a broad time range, which combines short-time rapid relaxation with long-time slow tailing. Consequently, it provides an accurate mathematical tool for the cross-scale description of memory effects. The fractional order within the kernel function, together with the parameters of the Mittag-Leffler function, collectively determines the rate and pattern of decay, offering definitive parameters for quantifying the "persistence" of memory.

Analyzing from the perspective of response characteristics, the fractional-order memory kernel determines how the material allocates weights to different historical stimuli. Its non-exponential decay characteristic implies that, compared to classical models, the fractional kernel function does not assign

an exponentially dominant absolute weight to recent history. Instead, it employs a relatively gentle power-law weighting scheme while still retaining a non-negligible influence from the more distant past. This weight allocation mechanism enables fractional-order models to more reasonably simulate phenomena such as the material's hysteresis loops, incomplete recovery during creep recovery, and the slow decay in the later stages of stress relaxation. Under dynamic loading, the relationship between the storage modulus and loss modulus derived from the fractional kernel function and frequency exhibits an approximate power-law plateau across a broad frequency range. This provides a new theoretical perspective for understanding the material's energy storage and dissipation behavior over wide frequency domains.

2.3 Fractional-Order Description of Multi-Field Coupling and Nonlinear Memory Effects

In practical service environments, the memory effects of viscoelastic polymeric materials often exhibit strong coupling with multiple physical fields such as ambient temperature, humidity, and even electric or magnetic fields, while the mechanical response itself frequently shows significant nonlinearity. The extension of the fractional-order theoretical framework to such complex scenarios is reflected in associating fractional operators with field variables and state variables. For instance, under temperature field coupling, the fractional order or characteristic time constants within the model can be expressed as functions of temperature. Drawing on the concept of the time-temperature equivalence principle, the variation patterns of memory effects across different temperature ranges can be uniformly described through the temperature dependence of the fractional order. This approach inherently incorporates the influence of time-varying processes, such as physical aging and chemical aging, on the material's memory characteristics into the evolution equations of the model parameters.

For nonlinear memory effects, such as amplitude dependence under large strain amplitudes or modulus softening/hardening induced by loading history, linear fractional-order models require nonlinear extensions. One approach involves introducing fractional orders that depend on stress or strain amplitude, constructing variable-order fractional differential constitutive models. This allows the "strength" of memory or the "width" of the relaxation spectrum to dynamically adjust according to the current mechanical state. Another, more universal framework involves coupling fractional operators with nonlinear spring and damping elements in a nonlinear manner, or constructing rate-dependent hyperelastic-viscoelastic constitutive equations based on fractional calculus. These nonlinear fractional-order models can characterize the shape evolution of stress-strain hysteresis loops under cyclic loading, the nonlinear characteristics of loading rate effects, and the memory residue during recovery that depends on the maximum pre-strain. Thereby, they advance the application boundaries of fractional-order theory into the domains of strong nonlinearity and large deformation[3].

3. Application Prospects of the Extended Theory and Model Validation Methodology

3.1 Physical Identification and Inversion Strategies for Fractional-Order Model Parameters

The precise identification of parameters in fractional-order constitutive models is a crucial step linking theoretical extension to the characterization of real material behavior. Model parameters typically include the fractional differential order, generalized modulus coefficients, and characteristic times, among which the physical identification of the fractional order holds the most central significance. This order is not merely a mathematical fitting parameter; its numerical value is associated with the breadth and symmetry of the material's internal relaxation time distribution. A lower order often corresponds to a broader relaxation time distribution and more pronounced long-term memory effects. Data from dynamic mechanical thermal analysis, such as frequency-domain storage and loss modulus data, or time-domain data obtained from creep and stress relaxation experiments, provide the foundation for parameter inversion. Utilizing the analytical expressions of fractional-order models under Laplace or Fourier transforms allows for the establishment of theoretical prediction formulas for complex modulus or creep compliance.

The parameter inversion process is essentially an optimization problem based on experimental data. It requires the use of appropriate global optimization algorithms, such as genetic algorithms or particle swarm optimization, to minimize the error norm between model predictions and experimental data. This inversion strategy must consider the potential parameter redundancy of the model itself, as well as differences in the signal-to-noise ratio of experimental data across specific frequency bands or time periods. Successful parameter identification lies not only in obtaining the optimal fitting curve but,

more importantly, in analyzing the correlation between the inverted parameter set and the material's known physical characteristics (such as molecular weight distribution, crosslinking density, and plasticizer content). This analysis aims to endow the fractional-order parameters with a more profound microstructural interpretation and to verify their validity as intrinsic descriptors of the material.

3.2 Computational Implementation and Numerical Simulation Approaches for the Extended Model

Incorporating fractional constitutive relations into numerical simulation frameworks faces the core challenge of high computational cost and historical data storage requirements due to the non-local nature of the operators. A commonly used numerical implementation approach is based on the discrete definition of the Grünwald-Letnikov fractional derivative. This definition approximates the fractional derivative as a weighted sum of function values at the current and multiple past time steps. The weight coefficients are determined by the fractional order and can be efficiently calculated using recursive relations. While this direct discretization method is conceptually clear, it requires storing the history of variables throughout the entire solution process, which creates pressure for long-term simulations. To address this, improved algorithms based on exponential integration or the short memory principle have been developed. These algorithms truncate the influence of distant history while maintaining a certain level of accuracy, thereby effectively managing storage demands[4].

Analyzing material behavior under complex boundary conditions and geometric configurations requires integrating the fractional-order constitutive model into numerical computational frameworks such as the finite element method. This is typically achieved by introducing a fractional-order stress-strain update algorithm at the element integration points. Within each time increment, the algorithm must call upon the stored historical strain sequence to compute the current stress according to the discretized fractional-order constitutive relation. Developing an efficient and robust fractional-order finite element program necessitates proper handling of the stability of the time integration scheme, optimization of historical data access, and interface issues with existing commercial software. This numerical simulation approach makes it possible to predict the time-dependent mechanical performance of viscoelastic polymeric components with complex memory effects, providing tools for virtual design and performance evaluation.

3.3 Frontiers and Challenges in Theoretical Extension

A key frontier for the further extension of fractional-order theory in modeling viscoelastic memory effects lies in developing a multi-scale fractional framework with deeper physical information embedding. While current models have achieved success at the macroscopic phenomenological level, the quantitative mapping relationship between fractional parameters and molecular dynamics simulations or microstructural evolution has not yet been fully established. Future research needs to focus on constructing theoretical links to derive fractional operators from microscopic segmental motion or mesoscopic network models. This would enable parameters such as the fractional order to directly reflect specific molecular motion modes or entanglement dynamics, thereby realizing a closed loop for cross-scale prediction.

Another frontier direction involves advanced forms of models for complex systems. These include variable-order fractional models coupled with multiple physical fields, fractional stochastic differential equation constitutive relations describing random excitations and non-equilibrium processes, and data-driven fractional neural network modeling approaches. These extensions aim to capture the evolution of memory effects arising from environmental interactions, internal fluctuations, and high nonlinearity. The core challenge faced is that these complex models often involve increased parameter dimensionality, which may reduce their identifiability and physical interpretability. Simultaneously, the corresponding mathematical theoretical analysis and numerical solution methods also become more difficult. Developing a new generation of fractional-order theory that balances physical consistency, mathematical rigor, and computational feasibility constitutes a fundamental challenge that must be addressed for the continued advancement of this field.

Conclusion

This paper systematically discusses the research on extending the modeling of memory effects in viscoelastic polymeric materials using fractional calculus theory. By replacing classical integer-order derivatives with fractional-order operators, the constructed constitutive models can naturally relate to

the material's internal continuous relaxation time spectrum through the core parameter of the fractional order. They accurately characterize the memory evolution process from short-term power-law decay to long-term exponential tailing via the Mittag-Leffler-type kernel function. This framework has been further extended to the description of multi-physics coupling and nonlinear memory effects. By introducing strategies such as variable-order or nonlinear coupling, the model's adaptability to actual complex service environments has been enhanced. The discussion on parameter inversion identification and numerical implementation methods provides feasible pathways for the verification and application of the theory. The focus of future research lies in establishing a quantitative theoretical bridge between fractional-order parameters and microstructural characteristics, developing multi-scale models with deeper physical information embedding, and addressing challenges such as the identifiability of high-dimensional complex fractional models, computational efficiency, and mathematical theoretical foundations. The ultimate goal is to achieve a fundamental leap from phenomenological description to physical prediction.

References

[1] Zhao Zhengzheng, Fan Qiuhua, and Yuan Zhaokai. "State of Charge Estimation for Lithium-ion Batteries Based on Fractional Order and FOSVD-MIAUKF." Journal of Power Supply, 1-11.
[2] Zhao Yanfei. "Vibration of Fractional Viscoelastic Nanobeams and Nanorods Based on Nonlocal Strain Gradient Theory." MA thesis. Lanzhou University of Technology, 2023.
[3] Mei Shengqi, et al. "Study on Frequency Dependence of Concrete Damping Based on a Fractional Viscoelastic Model." Journal of Vibration and Shock, vol. 41, no. 15, 2022, pp. 201-208.
[4] Ma Hui. "Free Vibration of Viscoelastic Thin Films Based on Fractional Calculus Theory." MA thesis. Lanzhou University of Technology, 2022.