Numerical modeling and simulation on deformation and failure behaviors of polymeric materials

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A thesis submitted in partial fulfillment of the requirements for the Doctor of Philosophy degree in Mechanical and Materials Engineering
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Abstract

Featured by biocompatibility, high compliance and capacity in sustaining large deformation, dielectric elastomers (DEs) and hydrogels have gained extensive research popularity for their potential applications in the fields of soft robots, biomimetics, tissue engineering, drug delivery, and energy harvesting. The design of such soft and smart material-based devices and structures requires deep understanding and accurate simulation of their constitutive behaviors, which is challenged by their nonlinear material properties due to unique microstructures and multi-physics coupling. Meanwhile, in different application contexts, those structures are also susceptible to different failure modes, imposing further challenges in simulating and predicting their performance. To fulfill the potential applications and maintain the structural integrity of these soft and smart material-based devices, it is essential to develop accurate and robust numerical models to simulate their complex deformation behaviors and investigate the potential failures.

As the first step, a general finite element (FE) framework is established to simulate the nonlinear viscoelastic behaviors of elastomers by developing a user-defined material (UMAT) subroutine in Abaqus, which is capable of adopting most constitutive models for hyperelasticity and thermodynamics evolution laws for viscoelastic materials. Combining the developed FE framework and the nonlinear field theory for electromechanical coupling, a highly customized user-element subroutine (UEL) in Abaqus is developed to numerically investigate various failure modes of DEs including electromechanical instability, buckling, wrinkling and crumpling. The effects of nonlinear material viscosity and loading conditions on different failure modes are identified to further unveil the failure mechanisms of DEs. The mechanical rupture of such polymeric materials are also simulated by incorporating the phase field modeling (PFM) into the proposed FE framework. For the first time, the driving force to the fracture of viscoelastic elastomers is identified, and the micro-mechanism of the material viscosity is further elucidated with the consideration of polymer chain breakage based on polymer dynamics. Furthermore, the dynamic breaking-healing kinetics of self-healing hydrogels is numerically investigated based on a generalized recursive integration algorithm, which is expected to act as a general avenue to numerically simulate the time/history-dependent constitutive behaviors of polymeric materials.
To conclude, this thesis aims at developing a FE framework to provide a general approach for deformation simulation and failure analysis of polymeric materials. This numerical framework can further function as a universal platform to accurately predict the performance of soft and smart material-based devices with the capability of implementing different multi-physics coupling mechanisms and time/rate-dependent constitutive models.

Keywords
Dielectric elastomers, hydrogels, finite element modeling (FEM), failure behaviors, electro-mechanical coupling, phase field modeling, recursive integration algorithm, self-healing kinetics.
Summary for Lay Audience

Different from hard materials like metals, polymeric materials such as elastomers and hydrogels are featured with softness and capabilities of sustaining large deformation. Some of those materials are also responsive to external stimuli, including electric field, pH, magnetic field, and etc. Those properties make them attractive for some trending applications in the fields of soft robots, biomimetics, tissue engineering, drug delivery, and energy harvesting. To fulfill the potential applications and maintain the structural integrity of these soft and smart material-based devices, it is essential to have a better understanding on their deformation behaviors. Meanwhile, polymeric materials are susceptible to various failure modes in their service, which will affect the originally designed functionality of the devices. With these considerations, there is an urgent need to establish a computational framework to accurately and efficiently evaluate the performance of polymeric materials under different loading conditions. In this thesis, the finite element method is adopted to provide a computational platform to predict the performance of polymeric materials. The complex deformation behaviors are simulated considering their unique microstructures. On this basis, some common failure behaviors are also investigated. This proposed computational framework will help to facilitate novel design and optimization of the soft and smart material-based devices and structures in engineering applications.
Co-Authorship Statement

The following thesis contains articles that are published in technical journals or submitted for publication as listed below. These articles are all based on the preliminary ideas from Heng Feng and Dr. L. Jiang. The derivation of the formulations and the simulation work presented in these articles were performed by Heng Feng. Shan Gao helped debug user defined subroutines in Chapter 3 and Chapter 4. The manuscripts of these articles were written by Heng Feng, modified and reviewed by Dr. L. Jiang.

**Chapter 3**: Heng Feng, Jianyou Zhou, Shan Gao, Liying Jiang (2021). Finite element simulation of the viscoelastic behavior of elastomers under finite deformation with consideration of nonlinear material viscosity. Acta Mechanica, 232, 4111-4132. [https://doi.org/10.1007/s00707-021-03042-0](https://doi.org/10.1007/s00707-021-03042-0)


**Chapter 5**: Heng Feng, Liying Jiang. Phase field modeling on fracture behaviors of elastomers considering deformation-dependent and damage-dependent material viscosity. *Submitted to Engineering Fracture Mechanics, under review.*

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Dedication

To my grandpa in loving memory
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**Nomenclature**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>φ</td>
<td>Phase field variable</td>
</tr>
<tr>
<td>φ^{ele}</td>
<td>Electrical potential</td>
</tr>
<tr>
<td>G</td>
<td>Material shear modulus</td>
</tr>
<tr>
<td>G_c</td>
<td>Material critical energy release rate</td>
</tr>
<tr>
<td>g(φ)</td>
<td>Degradation function</td>
</tr>
<tr>
<td>γ^H</td>
<td>Healing rate of secondary bonds</td>
</tr>
<tr>
<td>Π</td>
<td>Total energy of the material</td>
</tr>
<tr>
<td>ℋ</td>
<td>Maximum isochoric elastic energy density</td>
</tr>
<tr>
<td>J</td>
<td>Determinant of deformation gradient tensor</td>
</tr>
<tr>
<td>J_{lim}</td>
<td>Stretch limit of polymer chains</td>
</tr>
<tr>
<td>κ</td>
<td>Bulk modulus</td>
</tr>
<tr>
<td>λ</td>
<td>Principal stretch</td>
</tr>
<tr>
<td>n_b</td>
<td>Total number of broken bonds of secondary bonds</td>
</tr>
<tr>
<td>N_A</td>
<td>Shape function at node A</td>
</tr>
<tr>
<td>η_{B_n}</td>
<td>Instantaneous material viscosity in subnetwork B_n</td>
</tr>
<tr>
<td>η_n</td>
<td>Initial material viscosity in subnetwork B_n</td>
</tr>
</tbody>
</table>
$U^{vol}$  Volumetric deformation energy density

$W$  Deformation energy density function

$\bar{W}^{EQ}$  Isochoric equilibrium deformation energy density

$\bar{W}^{NEQ}_{B_n}$  Isochoric non-equilibrium deformation energy density

$W^{ele}$  Electric free energy

$\bar{b}$  Body force vector

$\bar{D}_R$  Nominal electric displacement

$\bar{E}_R$  Nominal electric field intensity

$\bar{t}$  Surface traction

$\bar{R}$  Residual vectors

$B$  Left Cauchy-Green deformation tensor

$C$  Right Cauchy-Green deformation tensor

$D$  Symmetric part of the velocity gradient tensor

$E$  Green-Lagrange strain tensor

$F$  Global deformation gradient tensor

$\bar{F}$  Effective deformation gradient tensor in F-bar method

$F^{iso}$  Isochoric part of global deformation gradient tensor

$F^{vol}$  Volumetric part of global deformation gradient tensor
\( F_{B_n} \) Total deformation gradient tensor in subnetwork \( B_n \)

\( F_{B_n}^e \) Elastic deformation gradient tensor in subnetwork \( B_n \)

\( F_{B_n}^i \) Inelastic deformation gradient tensor in subnetwork \( B_n \)

\( K \) Material stiffness matrix

\( L \) Velocity gradient tensor

\( P \) First Piola-Kirchhoff stress tensor

\( S \) Second Piola-Kirchhoff stress tensor

\( \sigma \) Cauchy stress tensor

\( \mathbb{P} \) Projection tensor form isochoric deformation to global deformation

\( \mathbb{P}_{B_n}^{NEQ} \) Projection tensor form isochoric elastic deformation to global deformation in subnetwork \( B_n \)

\( \mathbb{P}_{B_n} \) Projection tensor from elastic deformation in subnetwork \( B_n \) to global deformation
Chapter 1

1 Introduction

1.1 Background

Polymeric materials, including elastomers, hydrogels, shape memory polymers and etc., generally refer to materials composed of polymer chains that are commonly featured by high compliance, biocompatibility, and large deformation capacity. With the addition of different aggregates into the polymer networks, some of those polymeric materials exhibit stimuli-responsive properties, for example, the electromechanical coupling of dielectric elastomers and the sensitivity of hydrogels to light, temperature, and pH. With the merits of such unique properties, these soft and smart materials have attracted great research attention for the promising applications as soft robots [1-3], artificial muscles [4-6], sensors [7, 8], energy harvesters [9, 10], biomimetic devices [11,12], and to name a few. For the design and optimization of those soft and smart material-based devices, it becomes essential to model their complex deformation behaviors under different loading conditions with the help of numerical simulations. In addition, the functionality of such devices is commonly compromised by different failure modes in some application contexts. Thus, the corresponding numerical framework should also be extended to incorporate different failure mechanisms to facilitate the full design and optimization of polymeric material structures.

One of the main tasks for designing the polymeric material-based devices is to accurately characterize their constitutive behaviors and implement the appropriate constitutive models in the numerical modelling framework. Polymeric materials commonly exhibit hyperelasticity stemming from the cross-linked ground network for sustaining large deformation. In addition to the hyperelastic behavior, some polymeric materials also demonstrate time-dependent and rate-dependent responses, which are attributed to the material viscosity mainly originating from the reptation/diffusion of entangled polymer chains [13, 14]. It is also worth noting that the material viscosity of some elastomers is found as deformation-dependent especially under large deformation [15]. Meanwhile, for
some stimuli-responsive materials, the nonlinearity is further aggravated by polarization of polymer chains due to external stimuli. The constitutive behaviors become even more complicated if the polymeric materials consist of more than one type of polymer chain. One example is the self-healing hydrogels, which are designed to improve the fracture toughness by adding an extensible and loose crosslinked polymer network into a brittle but healable network with tightly connected sacrificial bonds [16, 17]. It is evident that the numerical simulation on such materials requires further mathematical treatment due to the complex constitutive modeling.

Different failure modes might occur during the service of the polymeric material-based devices. For example, actuators as artificial muscles commonly sustain large deformation, thus, they are susceptible to fractures especially when considering the inherent defects and stress concentration. It has been well recognized that the fracture of elastomers is dominated by the rupture of the crosslinked chains, and thus their fracture behaviors should be rate-insensitive which have been verified through experiments [18]. However, their rate-dependent constitutive behaviors due to the material viscosity contradicts such observations, which need further investigation to capture the role of material viscosity in the fracture mechanisms. In addition to the material rupture, the functionality of stimuli-responsive devices is also affected by some unique failure modes. Taking dielectric elastomers (DE) as an example, when subject to high electric field, DE actuators may encounter electromechanical instability which is also known as the snap through instability featured by the excessive thinning of the DE plate under the electric field [19]. In order to suppress such failure, certain boundary constraints are applied to avoid the potential excessive thinning of DE structure, which however might lead to buckling, wrinkling, or crumpling depending on the constraint configurations.

1.2 Motivation

Due to the complexity and nonlinearity of the constitutive models, it is difficult if not impossible, to get analytical solutions for general cases when polymeric materials have complex configurations or undergo non-homogeneous deformation. Under this circumstance, finite element method (FEM) becomes a feasible way in modeling the complex deformation behaviors of these polymeric materials. However, there are some
challenges when implementing the constitutive models into the FE framework. One of the challenges arises when the constitutive model is expressed in terms of the principal stretches, which needs special mathematics treatments to obtain the analytical expressions of the stress tensor and the spatial elasticity tensor due to their dependence on the explicit calculation of the principal stretches and their gradients [20]. In addition, when considering the self-healing mechanisms for some double-network polymeric materials, the numerical simulation on their constitutive behaviors is further challenged by the intractable computational cost due to the unavoidable convolution-like integrations [21]. As mentioned in the previous Section, polymeric materials are also susceptible to various failure modes. However, there is a knowledge gap that the evolution of microstructure and the effect of material viscosity on their failure behaviors are still not well understood. Therefore, there exists an urgent need for the development of an efficient numerical platform that is capable of characterizing various complex and nonlinear constitutive behaviors of polymeric materials and simulating their deformation and failures.

1.3 Objectives

The objective of this thesis is to establish an efficient FE framework to numerically investigate the deformation and failure behaviors of polymeric materials with various configurations, aiming to have a better understanding on their constitutive behaviors and failure mechanisms, and thus to provide guidelines for the optimal design of soft and smart material-based structures and devices in engineering applications. Dielectric elastomers and self-healing hydrogels are investigated as representative examples, while the full capacities of the proposed numerical framework are still reserved for other polymeric materials with the extension to incorporate multi-physics coupling. Attention will be focused on:

(i) Establishing a generalized FE framework that is capable of adopting different constitutive models with the consideration of hyperelasticity, nonlinear material viscosity, damage evolution, and dynamic breaking-healing kinetics.
(ii) Investigating the fracture behaviors of viscoelastic elastomers with the consideration of the evolution of microstructures due to the breakage of polymer chains and identifying the driving force of fracture.

(iii) Simulating various failure modes of dielectric elastomers, including electromechanical instability, buckling, wrinkling, and crumpling and identifying their influencing factors to unveil the underlying mechanisms.

(iv) Developing a novel algorithm in FE modeling to efficiently predict the deformation behaviors of self-healing hydrogels, which is expected to function as a general platform for numerical simulation on the time/history dependent constitutive behaviors of polymeric materials.

1.4 Thesis structure

Following a general introduction in Chapter 1, a detailed literature review is provided in Chapter 2. Then, the FE framework with the capability of adopting most of the constitutive relations in terms of either strain invariants or principal stretches is introduced in Chapter 3. In Chapter 4, the numerical simulation work on some common failure modes of dielectric elastomers is presented combining the nonlinear field theory with the micro–macro constitutive model incorporating nonlinear material viscosity. In Chapter 5, the fracture behaviors of viscoelastic elastomers are simulated by integrating the micro-macro viscoelasticity constitutive model and the phase field model. With further consideration of the polymer chain breakage, the phase field variable is connected to the topology constraints on the diffusive polymer chains. Chapter 6 performs a representative simulation on the transient and stabilized deformation behaviors of self-healing hydrogels by successfully alleviating the computational cost when dealing with the time-dependent breaking-healing kinetics. Chapter 7 summarizes the thesis and makes suggestions for the future work.
Reference


Chapter 2

2 Literature review

This chapter presents a literature review on modeling the constitutive behaviors and some common failure modes of polymeric materials. To be specific, the development of constitutive models considering different polymeric microstructures as well as external stimuli-responsive properties will be reviewed first. On this basis, computational modeling of potential bifurcation and instabilities of polymeric materials will be then discussed. Different analytical and numerical approaches in characterizing the fracture behaviors of polymers will also be presented. Lastly, the knowledge gap in literature will be highlighted.

2.1 Constitutive behaviors of polymeric materials

2.1.1 Material hyperelasticity and continuum mechanics description

Polymeric materials commonly exhibit nonlinear hyperelasticity originating from their ground network of cross-linked polymer chains. The development of the classic theories for such nonlinear elasticity is outlined in the works of Ogden [1] and Holzapfel [2]. In the framework of classic continuum mechanics for charactering hyperelasticity, we first denote a material point of the polymer at position $\mathbf{X}$ in the reference configuration $\Omega_0$, which moves to position $\mathbf{x}$ in the deformed configuration $\Omega$. The deformation gradient is defined as $\mathbf{F} = \frac{\partial \mathbf{x}}{\partial \mathbf{X}}$. When subjected to external loads, the virtual work principle states that,

$$\int_{\Omega_0} \frac{\partial W(\mathbf{F})}{\partial \mathbf{F}} : \mathbf{\delta F} dV = \int_{\Omega_0} \mathbf{\delta F} \cdot \mathbf{\delta \tilde{x}} dV + \int_{\partial \Omega_0} \mathbf{\delta \tilde{t}} \cdot \mathbf{\delta \tilde{x}} dA \quad (2.1)$$

where $\mathbf{\delta F}$ is the body force, $\mathbf{\delta F}$ is the surface traction, and $W(\mathbf{F})$ is the free energy density function. The first Piola-Kirchhoff stress is then defined as $\mathbf{P} = \frac{\partial W}{\partial \mathbf{F}}$. Within this framework, the stress-strain relation is derived from the specification of the free energy
density function. In literature, the hyperelastic constitutive models are generally classified into two categories, i.e., phenomenological models and mechanistic models.

The phenomenological models define the strain energy density functions in terms of strain invariants or principal stretches. The principal stretch-based free energy density function commonly takes the form of,

$$W(\lambda_1, \lambda_2, \lambda_3) = w(\lambda_1) + w(\lambda_2) + w(\lambda_3)$$  \hfill (2.2)

An example case for such models is the Ogden model [3] with the strain density function specified as,

$$W_{Ogden} = \sum_{k=1}^{K} \frac{\mu_k}{\alpha_k} \left( \lambda_1^{\alpha_1} + \lambda_2^{\alpha_2} + \lambda_3^{\alpha_3} \right)$$  \hfill (2.3)

On the other hand, a typical format of free energy density function in terms of strain invariants was proposed by Rivlin and Saunders [4] following,

$$W_R(I_1, I_2) = \sum_{i=0, j=0}^{M} C_{ij}^R (I_1 - 3)^i (I_2 - 3)^j$$  \hfill (2.4)

With certain specifications on the material constants $C_{ij}^R$, the free energy density function can be reduced into different forms. With $M=1$ and $C_{11} = 0$, the Mooney-Rivlin model is obtained from Eq. (2.4) [5]; with further specification that $C_{01} = 0$, the Mooney-Rivlin model is reduced to the neo-Hookean model; when $j = 0$ while $i$ takes 1, 2 and 3 respectively, the Rivlin model is reduced to the Yeoh model [6] which includes the effect of the higher order of the first invariant $I_1$. In order to incorporate the extensibility limit of polymeric materials into the constitutive modeling, Gent [7] introduced the extension limit that polymer chains can sustain into the free energy density function that can also be reduced to the neo-Hookean model for the limiting case.

Alternatively, the constitutive models stemming from the statistical mechanics treatment have also been developed to predict the hyperelastic behaviors of elastomers. From the
molecular-statistical perspective, a rubber-like material is idealized as consisting of a number of polymer chains cross-linked at certain junction points [8]. According to deformation affinity of the polymer chains with the macroscopic deformation, the statistical mechanics-based models can be further divided into affine models and non-affine models. The affine models were formulated based on the deformation affinity with different polymer chain distributions, and can be further classified into Gaussian and non-Gaussian models [9,10,11]. For the non-affine models, in addition to the deformation of crosslinked polymer chains, the effects of polymer chain entanglement can also be addressed. In the Edwards’s tube model [12], the topological entanglement is applied to each monomer of polymer chains, implying that the fluctuations of polymer chains are restricted in a confined tube. Rubinstein and Panyukov [13] combined the features of the confined tube model and the slip-link model to establish a new slip-tube model, which provides prediction in good agreement with the uniaxial experimental data of polybutadiene and natural rubber. In the slip-link model [14], the polymer chain entanglement is modeled as slip-links connecting neighboring polymer chains, which are allowed to slide along the contour of the two chains up to a fixed distance. Davidson and Goulbourne [10] developed the non-affine model by combining the concepts of the slip-tube model and the 8-chain model [15], which is capable of characterizing the effects of both the crosslinked polymer network and the entanglement of polymer chains for elastomers under finite deformation. The detailed expressions of strain energy density functions for the exemplary models are outlined in Table 2.1.
### Table 2.1 Hyperelasticity constitutive models

<table>
<thead>
<tr>
<th>Phenomenological models</th>
<th>Mechanistic models</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Mooney-Rivlin</strong></td>
<td><strong>Neo-Hookean</strong></td>
</tr>
<tr>
<td>[ W = C_1 (I_1 - 3) + C_2 (I_2 - 3) ]</td>
<td>[ W = \frac{\mu}{2} (I_1 - 3) ]</td>
</tr>
<tr>
<td><strong>Ogden</strong></td>
<td><strong>Arruda-Boyce</strong></td>
</tr>
<tr>
<td>[ W = \sum_{k=1}^{K} \frac{\mu_k}{\alpha_k} \left( \lambda_1^{\alpha_k} + \lambda_2^{\alpha_k} + \lambda_3^{\alpha_k} \right) ]</td>
<td>[ W = \mu \ln \left[ \sqrt[3]{\frac{I_1}{n}} + \ln \left( \frac{\beta_{ch}}{\sinh \beta_{ch}} \right) \right] ]</td>
</tr>
<tr>
<td><strong>Yeoh</strong></td>
<td><strong>Robinstein-Panyukov</strong></td>
</tr>
<tr>
<td>[ W = \sum_{i=1}^{3} C_i (I_i - 3) i ]</td>
<td>[ W = \frac{c k T}{2 N} \left( 1 - \frac{2}{\phi} \right) I_1 + \frac{c k T}{2 N} \sum_{\alpha=1}^{3} \left( \lambda_\alpha + \frac{1}{\lambda_\alpha} \right) ]</td>
</tr>
<tr>
<td><strong>Gent</strong></td>
<td><strong>Davidson-Goulbourne</strong></td>
</tr>
<tr>
<td>[ W = -\frac{G}{2} J_{\text{lim}} \ln \left( 1 - \frac{I_1 - 3}{J_{\text{lim}}} \right) ]</td>
<td>[ W = \frac{1}{6} G_e I_1 G_e \lambda_{\text{max}}^2 \ln \left( 3 \lambda_{\text{max}}^2 - I_1 \right) ]</td>
</tr>
</tbody>
</table>

#### 2.1.2 Viscoelasticity of polymeric materials

In addition to hyperelasticity, some of the polymeric materials also exhibit viscoelasticity due to the diffusion of polymer chains. Extensive research has been conducted to develop constitutive models for describing the viscoelastic nature of polymeric materials. Borrowing the concept from finite deformation plasticity theory [16], the parallel rheological framework (PRF) lays a foundation for modeling the viscoelastic effects of polymer networks by multiplicatively decomposing the deformation gradient [17]. This rheological framework stems from the idealization of the polymer networks of elastomeric materials, which comprise a strong and flexible cross-linked ground network and some subnetworks formed by different types of entangled diffusive polymer chains, as shown in Fig. 2.1.
From the continuum mechanics perspective, the mechanical properties of the hyperelastic ground network are commonly characterized by means of strain energy density functions as introduced in previous section. Similar formulations can also be applied for the viscous subnetworks to model the non-equilibrium deformation. In such models, the rate-dependent properties of materials are depicted by either introducing time-dependent coefficients into the free energy function [18] (known as integration form) or following the thermodynamics evolution laws in terms of internal stress-related or strain variables [19] (known as differential form). With the explicit expression of the free energy density function, the macroscopic viscoelastic behaviors of elastomers can therefore be quantified.

In the literature, the underlying physical mechanisms governing the viscoelasticity of polymeric materials is characterized by incorporating the statistical mechanics treatments [20] into the PRF, in which the strain energy density functions are modified to include the parameters of polymer physics and chemistry. Considering the topological constraints of the polymer chains due to the entanglement [21], the tube model [14] with the adoption of theories of polymer dynamics has been well recognized in the literature to capture the deformation-dependent viscosity. For this tube model, the macroscopic deformation of materials is linked to the material microstructure by assuming that polymer chains are confined in a tube-like region by other nearby chains, which is the basis for the
development of the micro-mechanism-inspired constitutive models for finite deformation viscoelasticity [22, 23]. When the elastomer is under large deformation, the more realistic scenario is that the tube diameter will change, which thus affects the reptation ability of the polymer chain confined in the tube and its relaxation process. With this consideration in the modified tube model [12], Tang et al. [24] and Li et al. [25] have proposed the micro-mechanically based viscoelasticity models to characterize the viscoelastic behavior of both vulcanized and un-vulcanized rubbers and successfully captured the nonlinear material viscosity. More recently, combining the polymer dynamics and the parallel rheological framework, Zhou et al. [26] established a micro–macro constitutive model that incorporates the nonlinear viscosity mechanisms into the continuum modeling framework, and hence is able to capture the typical viscoelastic behavior of elastomers more precisely, as well as quantitatively fit experimental data of different types of elastomeric materials.

2.1.3 Multi-physics coupling

With the addition of aggregates into polymer networks, some polymeric materials are stimuli-responsive [27], the deformation of which also depends on the external fields such as electric field [28], magnetic field [29], pH [30], and temperature [31]. The constitutive models for such materials need to be further modified to incorporate the multi-physics coupling effects. In addition to some other empirical and phenomenal models, a systematic approach is based on the introduction of the stimuli-induced free energy that follows the thermodynamics law for the minimization of the total potential energy. Accordingly, the free energy density function can be expanded as,

\[
W^{coupling}(F, X_1^{sti}, X_2^{sti}, ...)= W^{mech}(F, X_1^{sti}, X_2^{sti}, ...)
+ W_1^{sti}(F, X_1^{sti}, X_2^{sti}, ...)+ W_2^{sti}(F, X_1^{sti}, X_2^{sti}, ...)
+ ...
\]  

(2.5)

where \(X_i^{sti}\) is the responsive physical variable to the external stimuli, and \(W_i^{sti}\) is the corresponding free energy density caused by the stimuli. \(X_i^{sti}\) can be interpreted as electrical field \(\vec{E}_R\) for the electromechanical coupling dielectric elastomers [32],
magnetic induction $B$ for magnetic field-responsive elastomers, [33] or the number of solvent molecules $C$ for hydrogels [34] even though strictly speaking, neutral hydrogels are not responsive. Within this framework, the distribution and evolution of the stimuli variables need to be defined following certain physics laws, and the balance equations as well as the boundary conditions can be determined from the thermodynamics-consistent formulations. With the defined physics variable $X_{i}^{sti}$ and the stimuli induced free energy density, the corresponding constitutive relations with the multi-physics coupling can be derived.

The next step is to derive the field equations for solving the multi-physics coupling of polymer materials under external loads, which is illustrated with the example of dielectric elastomers following the standard thermodynamics formulation. For this electromechanical coupling, the physics variable is defined as the nominal electric field $\bar{E}_{R}$, which is defined as,

$$\bar{E}_{R} = -\frac{\partial \varphi^{ele} (\hat{X},t)}{\partial \hat{X}}$$

(2.6)

where $\varphi^{ele}$ is the applied stimuli, i.e., electric potential for DEs. For some other stimuli, such as chemical potential for hydrogels and temperature for the thermo-sensitive polymers, the directly applied stimuli might not keep constant but instead follow certain evolution laws or the distribution functions. For example, when the dynamic swelling behaviors are considered, an external kinetic law to quantify the water diffusion flux $J$ is adopted as,

$$J = -\frac{D}{vkT} F^{-T} F^{-1} \left[ \det (F) - 1 \right] \text{Grad} \mu$$

(2.7)

where $\mu$ is the chemical potential of the solvent and can be considered as the applied stimuli of the humidity-sensitive hydrogel. Therefore, the physical variable $C$ (the solvent concentration) that is directly linked to the free energy $W^{sti}(F,C)$ is determined following,
\[ \dot{C} = -\text{div} J \]  

(2.8)

It's worth noting that, in the case of temperature-responsive polymeric materials, it is necessary not only to consider the heat conduction laws that follow a similar diffusion law as Eq. (2.7), but also to explicitly specify the dependence of the material parameters on the change of temperatures. With the defined physics variable \( X_i^{st} \) and the stimuli induced free energy density, the corresponding constitutive relations with the multi-physics coupling can be derived. Accordingly, the total free energy of the coupled system in the variation form is extended as,

\[
\delta \Pi = \int_{\Omega_0} \frac{\partial W^{\text{coupling}}}{\partial F} : \delta FdV + \sum_{i=1}^{m} \int_{\Omega_0} \frac{\partial W^{\text{coupling}}}{\partial X_i^{s1}} : \delta X_i^{s1} dV \\
- \left( \sum_{i=1}^{m} \int_{\Omega_0} \delta \left( V_i^{s1} \phi_i^{s1} \right) dV + \sum_{i=1}^{m} \int_{\partial \Omega_0} \delta \left( U_i^{s1} \phi_i^{s1} \right) dA \right) \\
- \left( \int_{\Omega_0} \mathbf{b} \cdot \delta \mathbf{x} dV + \int_{\partial \Omega_0} \mathbf{t} \cdot \delta \mathbf{x} dA \right) 
\]  

(2.9)

where \( V_i^{s1} \) and \( U_i^{s1} \) are the volume and surface density of the external work by the stimuli with \( V_i^{s1} \) and \( U_i^{s1} \) being the volume and surface density of the non-conservative physical entities of the stimuli and \( \phi_i^{s1} \) being the effective loads on the them. Taking the electromechanical coupling behavior of the dielectric materials as an example, the variation of the total energy of the system is formulated as,

\[
\delta \Pi = \int_{\Omega_0} \frac{\partial W(F, \phi)}{\partial F} : \delta FdV - \int_{\Omega_0} \mathbf{b} \cdot \delta \mathbf{u} dV - \int_{\partial \Omega_0} \mathbf{t} \cdot \delta \mathbf{u} dA \\
+ \int_{\Omega_0} \frac{\partial W}{\partial E_R} \cdot \delta E_R dV - \int_{\Omega_0} \mathbf{q} \delta \phi^{\text{ele}} dV - \int_{\partial \Omega_0} \omega \delta \phi^{\text{ele}} dA 
\]  

(2.10)

where \( \mathbf{q} \) is the body charge density and \( \omega \) is the surface charge density applied on the surface \( \partial \Omega_0 \). Then the balance equations and the boundary conditions are determined from the energy conservation \( \delta \Pi = 0 \). For DEs, the explicit governing equations can be obtained by applying the divergence theorem as,
in domain $\Omega$, and

\[
\frac{\partial W}{\partial \mathbf{F}} \cdot \mathbf{n} = \mathbf{t} \tag{2.12a}
\]

\[
-\frac{\partial W}{\partial \mathbf{E}_r} \cdot \mathbf{n} = \mathbf{\omega} \tag{2.12b}
\]

as boundary conditions on surface $\partial \Omega$. With those formulations, the multi-physics coupling behaviors of stimuli-responsive polymeric materials can be solved analytically or numerically by certain numerical techniques such as finite element [35, 36] and finite difference method [37,38].

### 2.1.4 Self-healing behaviors of polymeric materials

The demands in the applications with long lasting usage and large load bearing capacity have motivated extensive research efforts for the improvement of fracture toughness of the polymeric materials. One of the pioneering examples is the development of double-network (DN) hydrogels, first synthesized by Gong et al. [39]. To obtain high mechanical strength, a loose and extensible polymer network with low ratio of cross-linking agents was incorporated into the tightly crosslinked polymer network to form an interpenetrating polymer structure. When subjected to large loads, some bonds of the tightly crosslinked chemical network also called as the sacrificial bonds will break to dissipate energy while the extensible network retains the material conformation, leading to greatly enhanced toughness and stiffness [40,41]. By replacing the crosslinks of the sacrificial network with weak but reformable bonds such as ionic [42], hydrogen [43, 44], dynamic covalent [45] and metal coordination [46] bonds, the toughness and fatigue resistance of the DN hydrogels were further improved owing to the healing capability of the sacrificial bonds [47]. Those hydrogels, also known as intrinsic self-healing hydrogels, exhibit highly
nonlinear viscoelasticity due to the time-dependent and history-dependent behaviors of the re-attached bonds.

Though the constitutive behaviors may vary among the self-healing hydrogels with different dynamic breaking-healing mechanisms, the preliminary task for material characterization is always to quantify the breaking-healing kinetics of the sacrificial network. Hui and Long [48] formulated the dynamic breaking-healing process by assuming that the debonding rate of the sacrificial ionic bonds depends on the maximum deformation that the current bonds have ever experienced, while the healing rate is related to the number of the broken bonds as well as the deformation history. Similar kinetics was also adopted for modeling the self-healing behaviors of hydrogels with metal-coordinated bonds [49] and other dynamic bonds [50, 51]. Later on, with further experimental investigation [52], the breaking-healing kinetics of the self-healing of hydrogels with ionic sacrificial bonds was modified by Long et al. [53] and Venkata et al. [54] to be only sensitive to the time history excluding the effects of the imposed deformation. In Long’s [53] theory, the instantaneous healing rate of the broken sacrificial bonds is dependent on the total number of the broken bonds, while the breaking rate depends on the number of the temporarily connected chains as illustrated in Fig. 2.1. It is worth noting that the breaking rates of the original and the reborn sacrificial bonds may differ considering that the reattached chains are less stable than the original ones.
Figure 2.2 Illustration of self-healing kinetics

A more physical interpretation of such kinetics was given by Lamont et al. [55] based on statistical damage mechanics to provide a deeper insight into the self-healing mechanisms. Similarly, the re-bonding of the broken chains was reported to be motivated by intermolecular forces and heating by Külcü [56] based on the thermodynamics of polymer microstructures. With those established time-dependent breaking-healing kinetics framework, the material constitutive behaviors can be accurately characterized by the convolution-like integral model in mathematics, as [53],

\[
W(t) = \frac{N_i + N_{20}(t)}{N_0} W_0(I_i) + \frac{1}{N_0} \int_0^t \phi(t-\tau) \gamma(\tau) W_0(I_i^{\rightarrow \tau}) \, d\tau \quad (2.13)
\]

where \( N_0 \) is the total number of polymer chains, \( N_i \) is the number of the permanent chains and \( N_{20}(t) \) is the number of the original sacrificial chains that exist at \( t=0 \) and remain undamaged at the current time \( t \). The contributions of the two networks of hydrogels to the sustained stresses are therefore distinguished with the proposed free energy density function.
2.2 Instability and bifurcation of polymeric materials

When subject to large loads and external stimuli, polymeric materials are vulnerable to different modes of instabilities and bifurcations. The causes for such failure modes are highly related to their unique stimuli-responsive properties. For polymeric materials responsive to stimuli with time dependence, the bifurcations might be caused by the inhomogeneous field-rebalance due to the rapid change in external stimuli such as the configurational bifurcations of swelling hydrogels considering the diffusion rate of solvents [57, 58]. For some spontaneously responsive polymeric materials, the failures might be caused by excessive deformation due to large external stimuli such as electromechanical instabilities of dielectric elastomers [59, 60]. Some other failure modes can be attributed to the external constraints under certain deformation configurations such as buckling of dielectric elastomers under electric field with fixed ends [61, 62].

Though instability and bifurcation modes might be distinctive for different stimuli-responsive polymeric materials originating from different mechanisms, the modeling approaches follow similar procedures from the mathematical perspective. The analytical approaches are based on either the analysis of the loss of material stiffness or on the sudden change of structural stiffness. Zhao and Suo [63] proposed the internal stress balance to predict the onset of electromechanical instability of DEs based on certain experimental observations on stress responses. Based on a more robust mathematical foundation, Hessian method [32] adopted the assumption that material stiffness tensor (or Hessian matrix) becomes singular at the onset of the instabilities and hence can also be utilized to predict the stimuli-induced instabilities. The limitation of the Hessian method is also obvious that it can only be used for the onset of instabilities of homogenous deformation. By comparison, the incremental method [64, 65] overcomes such limitation by imposing the incremental deformation and incremental external field on the known deformation configuration without considering the bifurcations. With different prescription of the perturbation deformation, different out-of-plane bifurcation modes of polymeric materials with certain configurations can be characterized.

Due to the complex formulation, it is very difficult, if not impossible, to get analytical solutions for the post-bifurcation of polymeric structures with nonuniform configurations.
As the alternative, finite element method provides a more feasible tool for the inhomogeneous instability analysis. In the FE models, the inhomogeneous instabilities are triggered by introducing imperfections to the original model geometries [66], while were simulated with the same formulation framework for homogenous instability analysis. With the implementation of multi-physics coupling constitutive models, the FE methods have been used to simulate different stimuli-induced bifurcations of hydrogels [67, 68], DEs [66, 69], and magnetic-responsive elastomers [70]. Due to the convenience of model setup, different bifurcation modes can be easily distinguished with the change of loading modes and constraints with the well-defined FE discretization and formulation. For example, the transition of the out-of-surface instabilities from buckling to wrinkling, crumpling and other modes can be easily captured with the simple modifications on the model configurations. It should be noted that the loss of material stiffness such as electromechanical instability of DEs will lead to the divergence of the Newton-Raphson solver, and in this case, the onset of the divergence can be assumed as the indication of the instability if FE models are correctly defined. The utilization of FE methods can not only be used to simulate the post-bifurcation behaviors of polymeric materials, but also be used as a feasible tool for the design and optimization of polymer structures based on the reversible bifurcations, such as the soft grippers controlled by inhomogeneous swelling [71], or some elastomeric devices controlled by buckling [70, 72].

In FE simulation of the failure behaviors of those polymeric materials, there exists a major challenge to apply the material incompressibility constraint as most polymeric materials are idealized as incompressible. A classical multiplicative decomposition scheme, which decomposes the strain energy density functions into volumetric (dilatational) and isochoric (deviatoric) parts, has been widely adopted in order to solve this issue. With such decomposition scheme, the formulation of the isochoric and volumetric stress tensors as well as the spatial elasticity tensors are then required in order for the Newton-Raphson iterative schemes to converge. For elements with low order shape functions, volumetric locking due to the excessive constraints on the elements by integration points occurs when the incompressibility is approached. Several additional mathematical approaches have been proposed in the literature to mitigate volumetric locking issues, including the methods modifying the element integration schemes such as
hybrid-element method [73] and reduced integration method [74], and some methods applying external constraints on the element deformation such as the J-bar method [75], the B-bar method [76], and the F-bar method [77, 78]. The superiority of the various methods remains inconclusive, with each method presenting its own set of pros and cons. A more detailed comparison among those methods is referred to [75]. In literatures, we noted that F-bar method gains its popularity in FE analysis of the nearly incompressible materials due to the ease of implementation. It can be simply implemented by substituting the deformation gradient at each integration point with a modified one, which has the same determinant of the deformation gradient at the centroid of the element.

### 2.3 Computational modelling in fracture analysis

In addition to the stimuli-induced failures, material rupture is another main cause for the loss of functionality of the polymeric material devices, especially for polymers with inherent defects or high stress concentration under large deformation. To fulfill the potential applications and maintain the structural integrity, it is essential to investigate the fracture behaviors of polymeric materials under various loading conditions, including the prediction of crack initiation, propagation, and fracturing path.

The pioneering work on fracture mechanics can be dated back to 1920s by Griffith to explain the failure of brittle materials [79], establishing the foundation of linear elastic fracture mechanics (LEFM). The LEFM was further complemented by Irwin [80] and Rice [81] to form the essential framework, which has been widely adopted to predict the fracture behaviors of elastic bodies with sharp cracks. Elastomers usually exhibit more complicated fracture behaviors due to large deformation, nonlinear hyperelasticity and viscoelasticity, which necessitates the further development of fracture mechanics modeling. In line with the Griffith’s theory, the Lake-Thomas model [82] provided feasible measures for fracture properties of polymeric materials by formulating the critical energy release rate accounting for the total energy required to rupture the chains lying in the crack plane. Within the framework of the Lake-Thomas model, different dissipation mechanisms have been introduced to characterize the fracture behaviors of viscoelastic polymers [83, 84]. Despite the successful applications of those classical models based on the LEFM, their inherent limitations are non-negligible, i.e., the crack
nucleation and propagation path need to be carefully prescribed and it becomes very challenging, if not impossible, to model the merging and branching of cracks. Meanwhile, the discontinuity of the displacement fields poses great challenges in the numerical context for the implementation of those classical fracture mechanics models.

Cohesive Zone Model (CZM), proposed by Dugdale [85] and Barenblatt [86], has also been adopted in fracture analysis but with certain limitations. As the CZM is naturally applicable to describe crack nucleation and pervasive cracking through various time and length scales. Meanwhile, the traction-separation laws also need to be defined based on either displacement or potential to define the material separation behaviors of the crack zone. If the separation (or displacement jump) exceeds a threshold length, the material is considered to be fully ruptured [87]. With the prescription of the crazing process zone, the CZM has also been adopted to model the crazing behavior of some polymers within the damaged zone but still sustained by fibrils [88]. The numerical implementation of the CZM is performed with several computational strategies. The cohesive zone FE models are established by embedding cohesive surface elements in the prescribed damage zone in FEM. Such surface elements can be inserted before the simulation (intrinsic model) [89] or whenever they are needed (extrinsic model) [90]. Alternatively, without the extra surface elements, the extended finite element method (XFEM) [91] provides another feasible tool for the simulation with discontinuous displacement field in the CZM. The nodal displacements are enhanced with the introduction of enrichment shape functions that are capable of approximating the discontinuity by Heaviside step functions.

In recent years, the research community has attempted to approach the fracture problems in an alternative way. Originally proposed by Francfort and Marigo [92], the phase field model has attracted great research attention in fracture simulation due to the fact of convenience that the pre-defined crack nucleation and path are not required. Instead, the crack path can be self-consistently determined based on the assumption that the total free energy must always remain stationary with the consideration of material damage. Within this framework, the material damage is characterized by an auxiliary variable that varies from 0 to 1 interpolating between an intact state and a cracked state [93, 94]. Adopting
the phase field model, different crack topologies can be modeled with various choices of geometric crack functions [95, 96] that depict how the phase field fracture variable distributes in the diffusive damage zone. The degradation effects of material damage on the material stiffness can also be included in the phase field model with the introduction of a degradation function [93, 97-99], which determines how much deformation energy is involved in crack evolution and how the material damage affects the mechanical properties of the material.

The implementation of numerical techniques in solving the coupled deformation-phase field problem requires further mathematical treatment due to the non-convexity of the free energy functional and non-symmetricity of the stiffness matrix considering the crack irreversibility constraint. To tackle this challenge, two types of numerical methods are proposed in literature, i.e., the modified monolithic and staggered approaches. The modified monolithic approaches aim to solve the fully coupled deformation-phase field equations with certain modifications on the traditional fully-monolithic solvers. Recent advances in the development of such models have been reported by adoption of different modifications on the Newton-Raphson iteration scheme, such as the primal-dual active set strategy [100], the line-search assisted monolithic iteration strategy [101], Jacobian matrix separation method [102], and Broyden-Fletcher-Goldfarb-Shanno (BFGS) algorithm [103-105]. On the other hand, staggered solver, also known as the alternate minimization (AM) solver, provides another feasible solution to the non-convex problems. Compared with the monolithic solvers, the advantage of the staggered algorithm is manifested by its inherent robustness: the coupled non-convex problem is decoupled into iterative linear problems by holding either the displacement or the phase field as constant while solving the other one [106]. The Jacobian matrix hence becomes always symmetric and positive definite in the staggered algorithm. Therefore, this numerical solver can be easily implemented in the existing platform and commercial software with only certain attention to the step size [107].

2.4 Knowledge gap

The existing research efforts have motivated the application of the FEM in modelling the non-homogeneous deformation of polymeric materials. Nevertheless, to the authors’ best
knowledge, most of the studies in FE modeling of the finite deformation of elastomers are limited to the phenomenological constitutive models, and few of them has considered the microscopic mechanisms underlying the material viscosity. Meanwhile, for the self-healing double-network hydrogels, due to the time/history-dependent breaking-healing kinetics, the corresponding numerical simulation is greatly challenged with the computational cost from the convolution-like integrations. Thus, there exists an urgent need to develop an efficient FE framework to numerically simulate the complex deformations of such polymeric materials with the consideration of different constitutive behaviors.

Despite the efforts devoted to the computational investigation on failure behaviors of polymeric materials in the literature, the effects of the material viscosity as well as the evolution of microstructures on their failure behaviors are still not well understood. To be specific, the delayed external stimuli-induced instabilities and bifurcations due to viscoelasticity need further elucidated from the numerical aspect. Some experimental research reported that the fracture behaviors for the rate-dependent viscoelastic polymeric materials are rate-insensitive, which also requires re-examination from a microscopic perspective. Therefore, such failure behaviors will also be numerically investigated with the incorporation of polymer dynamics to fill the knowledge gap.

Reference


Chapter 3

3 Finite element simulation on the viscoelastic behaviors of elastomers under finite deformation with the consideration of nonlinear material viscosity

Governed by the diffusion process of the highly mobile and flexible polymer chains, viscoelasticity is one crucial property in modeling the finite deformation of elastomers. While the development of new constitutive models has drawn remarkable attention to reveal the underlying mechanisms of material viscosity, it becomes very challenging to predict the viscoelastic behaviors of elastomers, particularly when a complex structure is undergoing non-uniform deformation. The current work attempts to fill this knowledge gap by establishing a finite element (FE) framework to numerically predict the viscoelastic behaviors of elastomeric materials. In this FE framework, the micro-macro constitutive model, which is capable of capturing the nonlinear viscosity and the microstructure features of the material, is implemented by developing the user-defined material (UMAT) subroutine in Abaqus. The developed UMAT is featured by the capability of adopting most of the constitutive relations in terms of either strain invariants or principal stretches, indicating the adaptiveness of the FE framework. The accuracy and the modeling capacity of the FE framework are validated with several numerical examples on three commonly used elastomeric materials, including VHB 4910, HNBR50, and carbon black (CB) filled elastomers, under various loading conditions. The comparison of the viscoelastic responses shows an excellent agreement between the FE modeling results and the theoretical analysis. The established FE model is expected to provide guidance for novel design and applications of elastomer-based structures. The framework can also be further extended to characterize the multiphysics coupling behaviors of elastomeric materials under coupled fields.

3.1 Introduction

Elastomers are typically characterized by high compliance and capacity in sustaining large deformation, which has enabled their potential applications in many engineering and industrial fields, including energy harvesting devices [1-3], artificial muscles [4, 5],
acoustic actuators [6, 7], different types of sensors [8-10], and soft robots [11-13]. In addition to the hyperelastic behavior, elastomeric materials also demonstrate time-dependent and rate-dependent responses, which are attributed to the material viscosity mainly originating from the reptation of polymer chains [14, 15]. According to the theory of polymer dynamics [16] and experimental observation [17-19], the material viscosity is also found as deformation-dependent which becomes more evident when the elastomers undergo large deformation. The incorporation of both finite deformation and nonlinear viscoelasticity in modeling the behavior of elastomer-based structures becomes very challenging, particularly when the structure is under non-uniform deformation.

In the literature, extensive research has been conducted to develop constitutive models for describing the viscoelastic nature of elastomers. Borrowing the concept from finite-deformation plasticity theory [20], the parallel rheological framework (PRF) lays the foundation for modeling the viscoelastic effects of elastomers by multiplicatively decomposing the deformation gradient [21, 22, 23, 24]. This rheological framework stems from the idealization of the polymer networks of elastomeric materials, which comprise a strong and flexible cross-linked ground network and some subnetworks formed by different types of diffusive polymer chains. From the continuum mechanics perspective, the mechanical properties of the hyperelastic ground network are commonly characterized by means of strain energy density functions in terms of either principal stretches [25] or invariants of right Cauchy-Green deformation tensor [26-30]. Similar formulations can also be applied for the viscous subnetworks to model the non-equilibrium deformation. In such models, the rate-dependent properties of materials are depicted by either introducing time-dependent coefficients into the free energy function [31, 32] (known as integration form) or following the thermodynamics evolution laws in terms of internal stress-related or strain variables [33-35] (known as differential form). With the explicit expression of the free energy density function, the macroscopic viscoelastic behaviors of elastomers can therefore be quantified.

The nonlinear viscosity of elastomers is commonly characterized by incorporating the statistical mechanics treatments [36-40] into the PRF in the literature. Meanwhile, the underlying physical mechanisms governing the viscoelastic deformation of elastomers
can also be revealed as the strain energy density functions are modified to include the parameters of polymer physics and chemistry. Considering the topological constraints of the polymer chains due to the entanglement [17, 41], the tube model [14, 16] with the adoption of theories of polymer dynamics has been well recognized in the literature to capture the deformation dependent viscosity. For this tube model, the macroscopic deformation of materials is linked to the material microstructures by assuming that polymer chains are confined in a tube-like region by other nearby chains, which is the basis for the development of the micro-mechanism inspired constitutive models for finite deformation viscoelasticity [23, 33, 42-44]. When the elastomer is under large deformation, the more realistic scenario is that the tube diameter will change, which thus affects the reptation ability of the polymer chain confined in the tube and its relaxation process. With this consideration in the modified tube model [16], Tang et al. [45] and Li et al. [46, 47] have proposed the micro-mechanically based viscoelasticity models to characterize the viscoelastic behavior of both vulcanized and un-vulcanized rubbers and successfully captured the nonlinear material viscosity. More recently, combining the polymer dynamics and the parallel rheological framework, Zhou et al., [48] established a micro-macro constitutive model that incorporates the nonlinear viscosity mechanisms into the continuum modeling framework, and hence is able to more precisely capture the typical viscoelastic behavior of elastomers, as well as quantitatively fit experimental data of different types of elastomeric materials.

It is worth noting that constitutive modeling lays foundation and provides useful tools to characterize how materials deform when subjected to applied loads. However, it is difficult if not impossible, to get analytical solutions for general cases when the material has complex configurations or undergo non-homogeneous deformation. Under this circumstance, finite element method (FEM) is a feasible way in modeling the complex deformation behaviors of these rubber-like materials. The first challenging task in simulating the finite-deformation of elastomers by using the FEM is the material incompressibility constraint as most elastomeric materials are idealized as incompressible. For elements with low order shape functions, volumetric locking due to the excessive constraints on the elements by integration points occurs as the incompressibility is approached. A classical multiplicative decomposition scheme [49],
which decomposes the strain energy density functions into volumetric (dilatational) and isochoric (deviatoric) parts, has been widely adopted in order to solve this issue. With such decomposition scheme, the formulation of the stress tensor as well as the spatial elasticity tensor is then required in order for the Newton-Raphson iterative schemes to converge. In the literature, the generalized forms of spatial elasticity tensor were first formulated for nearly-incompressible hyperelastic [50] and elastoplastic materials [51], and then were extended to elastomeric materials for FE simulation [35, 52-54]. Some other strategies have also been proposed in the literature to avoid the volumetric locking, including the B-bar method [55], the mixed variation method [56], and the F-bar method [57, 58].

Another challenge rises when one attempts to adopt the constitutive models in terms of principal stretches such as Ogden model [25]. Special mathematics treatments are required to obtain the analytical expressions of the stress tensor and the spatial elasticity tensor due to their dependence on the explicit calculation of the principal stretches and their gradients. Feng et al. [59] and Dui et al. [60] derived the material elasticity tensor for the Ogden model by taking the second-order derivatives of the eigenvalues of the right Cauchy-Green deformation tensor. Kiran and Khandelwal [61] also obtained the explicit expressions of the material elasticity tensors for various hyperelasticity models using a numerical approximation scheme to evaluate the derivatives of the principal stretches. More recently, Connolly et al. [62] established a FE discretization scheme for hyperelasticity models in terms of the principal stretches, and improved the accuracy of the model when two or three of the principal stretches are similar or equal by adopting the L’Hôpital’s rule. Those pioneering studies that focus on the hyperelasticity models have paved the way for the implementation of the viscoelastic constitutive models involving the principal stretches in the FE analysis.

The existing research efforts have broadened the application of the FEM in modelling the non-homogeneous deformation of elastomers. Nevertheless, to the authors’ best knowledge, most of the studies in FE modeling of the finite deformation of elastomers are limited to the phenomenological constitutive models, and few of them has considered the microscopic mechanisms underlying the material viscosity. Therefore, in the present
study, a new micro-macro constitutive model developed by Zhou et al. [48] is implemented into the FE framework by developing the user-defined material (UMAT) subroutine in Abaqus to describe the viscoelastic behavior of elastomeric materials under finite deformation. This model is able to capture the nonlinear material viscosity through both continuum mechanics and statistical mechanics treatments. Due to the capability of adopting most of the constitutive relations in terms of either strain invariants or principal stretches, the established FE framework can function as a universal platform for simulating the finite deformation behaviors of elastomers. The robustness of this computational model has been validated through case studies and can be further extended for simulating the multiphysics coupling behavior of elastomeric materials under coupled fields.

3.2 Continuum mechanics framework

In this section, the continuum mechanics framework [21, 23, 24, 48] for the constitutive models is briefly reviewed. The viscoelasticity of the elastomer is described by the parallel rheological model as shown in Fig. 3.1, in which network A is a ground network and parallel subnetworks B_1 to B_N represent different types of polymer chains with similar diffusion behaviors.

![Figure 3.1 Illustration of parallel rheological model for elastomers with ground network A representing the equilibrium deformation and subnetworks B_1 to B_N representing the non-equilibrium deformation.](image)
When subjected to deformation $F$, the equilibrium network $A$ sustains pure elastic deformation $F_A$, while the non-equilibrium subnetworks relax with time and dissipate energy. For each non-equilibrium subnetwork, the deformation gradient $F_{B_n}$ is further expressed as $F_{B_n} = F_{B_n}^e F_{B_n}^i$ according to the multiplicative decomposition [24], where $F_{B_n}^e$ stands for the elastic part for the spring deformation and $F_{B_n}^i$ is the inelastic part representing the deformation gradient of the dashpot. It should be mentioned that $F = F_A = F_{B_n} (n=1, 2, \ldots, N)$ as the deformation acts on all the parallel polymer networks. Accordingly, the elastic and inelastic right Cauchy-Green deformation tensors in each non-equilibrium subnetwork are expressed as $C_{B_n}^e = (F_{B_n}^e)^T F_{B_n}^e$, and $C_{B_n}^i = (F_{B_n}^i)^T F_{B_n}^i$, respectively. The Helmholtz free energy density of the whole network is expressed as,

$$W(C_A, C_{B_1}^e, C_{B_2}^e, \ldots, C_{B_N}^e) = W^{EQ} (C) + \sum_{n=1}^{N} W_{B_n}^{NEQ} (C_{B_n}^e)$$

The second Piola-Kirchhoff stress is then defined as,

$$S = \frac{\partial W}{\partial E} = 2 \frac{\partial W^{EQ} (C)}{\partial C} + 2 \sum_{n=1}^{N} \frac{\partial W_{B_n}^{NEQ} (C_{B_n}^e)}{\partial C}$$

where $E = \frac{1}{2} (F^T F - I)$ is the Green-Lagrange strain tensor. Also, the Cauchy stress $\sigma$ can be determined from the second Piola-Kirchhoff stress through the following relation,

$$\sigma = J^{-1} F S F^T$$

where $J = \text{det}(F)$.

As indicated in Fig. 1, the stresses in non-equilibrium subnetworks are tightly related to the inelastic (viscous) deformation that is governed by the diffusion of the polymer chains. In order to calculate the second Piola-Kirchhoff stress in each non-equilibrium
subnetwork, the inelastic deformation gradient tensor $\mathbf{F}^{i}_{B_n}$ needs to be quantified which evolves following the dissipation inequality [24, 48], i.e.,

$$
\left( \mathbf{S} - 2 \frac{\partial W}{\partial \mathbf{C}} \right) : \frac{1}{2} \dot{\mathbf{C}} - \sum_{n=1}^{N} \left( \frac{\partial W_{\text{NEQ}}}{\partial \mathbf{C}_{B_n}^{e}} : \frac{\partial \mathbf{C}_{B_n}^{e}}{\partial \mathbf{F}^{i}_{B_n}} : \mathbf{F}^{i}_{B_n} \right) \geq 0 \tag{3.4}
$$

As Eq. (3.2) can be further expressed as,

$$
\mathbf{S} - 2 \frac{\partial W}{\partial \mathbf{C}} = 0 \tag{3.5}
$$

the dissipation inequality (3.4) is thus reduced as,

$$
\mathbf{\tau}^{i}_{B_n} \left( \mathbf{B}_{B_n}^{e} \right)^{-1} \left[ \mathbf{F}_{B_n}^{e} \mathbf{L}_{B_n}^{i} \left( \mathbf{F}_{B_n}^{e} \right)^T \right] \geq 0 \tag{3.6}
$$

where $\mathbf{L}_{B_n}^{i}$ is the velocity gradient tensor with the expression of $\mathbf{L}_{B_n}^{i} = \dot{\mathbf{F}}^{i}_{B_n} \left( \mathbf{F}_{B_n}^{e} \right)^{-1}$. $\mathbf{\tau}_{B_n} = 2 \mathbf{F}_{B_n}^{e} \frac{\partial W_{\text{NEQ}}}{\partial \mathbf{C}_{B_n}^{e}} \left( \mathbf{F}_{B_n}^{e} \right)^T$ and $\mathbf{B}_{B_n}^{e} = \mathbf{F}_{B_n}^{e} \left( \mathbf{F}_{B_n}^{e} \right)^T$ are the Kirchhoff stress tensor and the left Cauchy-Green deformation tensor, respectively. With the assumption of the material isotropy, one sufficient condition satisfying the inequality (3.6) is that the internal variables evolve following the thermodynamics evolution law [24], i.e.,

$$
\left\{ \mathbf{F}_{B_n}^{e} \frac{1}{2} \left[ \mathbf{L}_{B_n}^{i} + \left( \mathbf{L}_{B_n}^{i} \right)^T \right] \left( \mathbf{F}_{B_n}^{e} \right)^T \right\} \left( \mathbf{B}_{B_n}^{e} \right)^{-1} = \mathbf{\gamma}^{-1} : \mathbf{\tau}_{B_n} \tag{3.7}
$$

where the 4th rank tensor $\mathbf{\gamma}^{-1}$ takes the form of

$$
\mathbf{\gamma}^{-1} = \frac{1}{2\eta_{B_n}} \left( \mathbf{I}^4 - \frac{1}{3} \mathbf{I} \otimes \mathbf{I} \right) \tag{3.8}
$$

with $\eta_{B_n}$ being the viscosity for the individual subnetwork and $\mathbf{I}^4$ being the fourth order symmetric identity tensor.
In most of the existing literatures, for simplicity purpose, the material viscosity is assumed as a constant. However, the more realistic scenario is that the viscosity varies with the deformation as observed in experiments. In the current work, the viscosity of each subnetwork is linked to the macroscopic deformation according to the polymer dynamics theory [47,48], i.e.,

$$\eta_{\alpha} = \frac{\eta_n}{\alpha(F)^2}$$

(3.9)

where $\eta_n$ is the viscosity in each subnetwork in the reference state before the deformation; $\alpha(F)$ is the function of the deformation gradient tensor, which can be expressed through the modified tube model [16], as,

$$\alpha(F) = \frac{\langle \frac{1}{2} \hat{R}_{ee}^2 \rangle}{\langle \hat{R}_{ee}^2 \rangle_0 \int \frac{\hat{F} \hat{u}_0}{4\pi} d^3 \hat{\rho}_0}$$

(3.10)

The mean square end-to-end distance of the primitive chain $\langle \hat{R}_{ee}^2 \rangle$ is determined by

$$\langle \hat{R}_{ee}^2 \rangle = \int |\hat{F} \hat{R}|^2 f_0(\hat{R}) d^3 \hat{R},$$

where $f_0(\hat{R})$ is the Gaussian distribution function of the end-to-end vector $\hat{R}$ of the primitive chain in a unit sphere. $\hat{u}_0$ is the initial unit tangent vector of the primitive chain, and $\langle \hat{R}_{ee}^2 \rangle_0$ is the mean square end-to-end distance of the primitive chain before the deformation. The detailed derivation and explanation can be referred to Zhou et al. [48].

### 3.3 Material model implementation in FE framework

In this study, a user-defined material (UMAT) subroutine will be developed in Abaqus for the finite element analysis on the viscoelastic behaviors of elastomers. In developing a UMAT, two formulation processes are essential in each time increment step, including
stress tensor update and spatial elasticity tensor (Jacobian matrix) update. Meanwhile, in order to impose the incompressibility constraint of the material, the volumetric and isochoric decomposition scheme is utilized to decompose the deformation gradient tensor and hence the stress tensor and the Jacobian matrix into the volumetric and isochoric parts. In this FE framework, in order to capture the deformation dependent material viscosity, the evolution of the internal variables, i.e., the inelastic deformation, must be numerically determined. Therefore, the integration scheme of the evolution law of the internal variables with time discretisation is also presented here.

3.3.1 Stress tensor formulation with nearly incompressibility constraint

Considering the incompressibility of elastomers, the deformation gradient tensor is firstly decomposed into two parts according to the multiplicative decomposition scheme \([49,50]\), i.e., the volumetric deformation (pure volume change) and the isochoric deformation (pure shape change),

\[
F = F^{\text{iso}} F^{\text{vol}} = \left( J^{-1/3} F \right) \left( J^{1/3} I \right)
\]  

(3.11)

where the quantity with superscript ‘vol’ represents the volumetric part. It is obvious that there is no volume change in the first term \(F^{\text{iso}} = J^{-1/3} F\) as its determinant is always 1 regardless of the deformation. Correspondingly, the isochoric right Cauchy-Green deformation tensors are also formulated as \(C^{\text{iso}} = J^{-2/3} C\) and \(C^{\text{iso},e} = \left( J^{\text{iso}, e} \right)^{-2/3} C_{B_n}^{e}\). On this basis, the Helmholtz free energy density can also be decomposed into both volumetric and isochoric parts as,

\[
W = \bar{W}^{\text{EQ}}(\bar{F}) + \sum_{n=1}^{N} \bar{W}_{B_n}^{\text{NEQ}}(\bar{F}_{B_n}^{e}) + U^{\text{vol}}(J)
\]  

(3.12)

The framework of the rheological model allows the flexibility in choosing the specific strain energy density functions for the isochoric parts \(\bar{W}^{\text{EQ}}(\bar{F})\) and \(\bar{W}_{B_n}^{\text{NEQ}}(\bar{F}_{B_n}^{e})\).
depending on the material properties. While the volumetric strain energy function can be expressed in terms of the bulk modulus [63], as,

\[ U^{\text{vol}}(J) = \frac{1}{2} \kappa (J - 1)^2 \]  

(3.13)

With the relatively large bulk modulus \( \kappa \), it becomes difficult for the pure volume change deformation to occur, and hence the material incompressibility condition is enforced. The recommended value of the bulk modulus lies in the range of \( 10^4 \sim 10^6 \) times of the shear modulus as reported in the literature [64].

Accordingly, the second Piola-Kirchhoff stress tensors can also be further decomposed into three parts, i.e.,

\[
\mathbf{S} = \frac{\partial \mathbf{W}}{\partial \mathbf{E}} = 2 \frac{\partial \mathbf{W}^{\text{EQ}}}{\partial \mathbf{C}} \left( \mathbf{F} \right) + 2 \sum_{n=1}^{N} \frac{\partial \mathbf{W}_{B_n}^{\text{NEQ}}}{\partial \mathbf{C}} \left( \mathbf{F}_{B_n}^e \right) + 2 \frac{\partial U^{\text{vol}}(J)}{\partial \mathbf{C}}
\]  

(3.14)

where \( 2 \frac{\partial \mathbf{W}^{\text{EQ}}}{\partial \mathbf{C}} = \mathbf{S}^{\text{iso}}_{\text{EQ}} \) is the isochoric stress in the equilibrium state, \( 2 \frac{\partial \mathbf{W}_{B_n}^{\text{NEQ}}}{\partial \mathbf{C}} = \mathbf{S}^{\text{iso}}_{B_n} \) is the isochoric stress in the non-equilibrium state for each subnetwork, and the volumetric stress \( \mathbf{S}^{\text{vol}} = 2 \frac{\partial U^{\text{vol}}}{\partial \mathbf{C}} \) is caused by the volumetric change. These stresses can be further expanded as,

\[
\mathbf{S}^{\text{iso}}_{\text{EQ}} = J^{-2/3} \mathbf{P} : \mathbf{S}^{\text{EQ}}
\]  

(3.15a)

\[
\mathbf{S}^{\text{iso}}_{B_n} = \left( J_{B_n}^e \right)^{-2/3} \mathbf{P}_{B_n} : \left( \mathbf{F}_{B_n}^{\text{NEQ}} \mathbf{S}_{B_n}^{\text{NEQ}} \right)
\]  

(3.15b)

\[
\mathbf{S}^{\text{vol}} = \kappa J (J - 1) \mathbf{C}^{-1}
\]  

(3.15c)
where \( \overline{S}_{EQ} = 2 \frac{\partial \overline{W}_{EQ}}{\partial \overline{C}_{iso}} \) and \( \overline{S}_{B_n}^{NEQ} = 2 \frac{\partial \overline{W}_{B_n}^{NEQ}}{\partial \overline{C}_{iso,e}} \) are the direct derivatives of the isochoric free energy density functions with respect to \( \overline{C}_{iso} \) and \( \overline{C}_{iso,e} \). The fourth order tensors \( P \), \( P_{B_n}^{NEQ} \), and \( \overline{P}_{B_n} \) are projection tensors from \( \overline{C}_{iso} \) to \( C \), from \( \overline{C}_{iso,e} \) to \( C_{eB_n} \), and from \( C_{eB_n} \) to \( C \), respectively, which are defined as,

\[
P_{abcd} = J^{2/3} \left( \frac{\partial \overline{C}_{cd}}{\partial C_{ab}} \right) = \delta_{ac} \delta_{bd} - \frac{1}{3} \delta_{cd} \delta_{ab} \]  \hspace{1cm} (3.16a)

\[
\left( P_{B_n}^{NEQ} \right)_{abcd} = (J')^{2/3} \left( \frac{\partial \overline{C}_{eB_n,cd}}{\partial \overline{C}_{eB_n,ab}} \right) = \delta_{ac} \delta_{bd} - \frac{1}{3} \delta_{cd} \delta_{ab} \]  \hspace{1cm} (3.16b)

\[
\left( \overline{P}_{B_n} \right)_{abcd} = \left( \frac{\partial C_{eB_n,cd}}{\partial C_{ab}} \right) = \frac{1}{2} \left[ \left( F_{Bn}^{e} \right)^{-1} \left( F_{Bn}^{e} \right)^{-1} + \left( F_{Bn}^{e} \right)^{-1} \left( F_{Bn}^{e} \right)^{-1} \right] \]  \hspace{1cm} (3.16c)

### 3.3.2 Spatial elasticity tensor (Jacobian matrix)

In addition to the proper definition of the stress tensor, in the FEM framework, the Newton-Raphson iterative process requires the proper relation between the time-discretized stress change and the strain change. The spatial elasticity (Eulerian) tensor, also known as the Jacobian matrix in the UMAT, is used to quantify this relation.

As a common practice in the FE framework, the objective stress rate is needed first to formulate the relation between the stress variation and the strain variation. As an example in the literature [65], the spatial elasticity tensor is derived from the Kirchhoff stress tensor, which is defined in terms of the second Piola-Kirchhoff stress as \( \tau = FSF^T \). Then the time derivative of the Kirchhoff stress tensor is determined as

\[
\dot{\tau} = \dot{FSF}^T + FS\dot{F}^T + F\dot{S}F^T \]  \hspace{1cm} (3.17)

which can be further rewritten as,
\[ \dot{\tau} - L\tau - \tau L^T = F \left( \frac{\partial S}{\partial E} : \dot{E} \right) F^T \] (3.18)

The left side of Eq. (3.18) is called the Truesdell rate of the Kirchhoff stress tensor, which is denoted by \( \dot{\tau}^{(TK)} \). Adopting the Einstein’s notation, Eq. (3.18) is expanded as [65],

\[ \dot{\tau}_{ij}^{(TK)} = F_{in} F_{jp} F_{aq} F_{br} \frac{\partial S_{mp}}{\partial E_{qr}} D_{ab} \] (3.19)

where \( D \) is the symmetric part of the velocity gradient tensor \( L \) defined as \( D = \frac{1}{2} (L + L^T) \). Knowing that \( \frac{\partial S}{\partial E} = 2 \frac{\partial S}{\partial C} \), the spatial elasticity tensor originated from the Truesdell rate is expressed as,

\[ C_{ijab}^{(TK)} = 2F_{in} F_{jp} F_{aq} F_{br} \frac{\partial S_{mp}}{\partial C_{qr}} \] (3.20)

In the Newton-Raphson iteration, the convergence rate relies on the form of the objective stress rate in Eq. (3.19). Other choices of the objective stress rates include the Jaumman rate of the Kirchhoff/Cauchy stress tensor [66, 67], the Biezeno-Hencky rate of the Cauchy stress tensor [67], and some others. Among them, the Jaumman rate of the Cauchy stress is adopted in the built-in material constitutive models in Abaqus. However, it has been reported by Ji et al. [66] that the elasticity tensor derived from the Jaumman rate could lead to divergence and even great errors for problems that require incremental approach to obtain solutions. Therefore, in this study, the Truesdell rate of Kirchhoff stress as shown in Eq. (3.20) is adopted according to the conclusions from the literature [66, 67].

From Eq. (3.14), the material elasticity tensor (elasticity tensor in material description) \( 2 \frac{\partial S}{\partial C} \) can be expanded as,
\[
2 \frac{\partial S}{\partial C} = 2 \left( \frac{\partial S_{EQ}^{iso}}{\partial C} + \sum_{n=1}^{N} \frac{\partial S_{BE}^{iso}}{\partial C} + \frac{\partial S^{vol}}{\partial C} \right) \quad (3.21)
\]

Using the chain rule, the material elasticity tensor \(2 \frac{\partial S_{EQ}^{iso}}{\partial C}\) for the equilibrium ground network in Eq. (3.21) is calculated as [50],

\[
2 \frac{\partial S_{EQ}^{iso}}{\partial C} = \left( \hat{\mathbf{C}} - \frac{1}{3} \mathbf{A}_1 \right) : \mathbf{P}^T - \frac{2}{3} \left( \mathbf{A}_2 + \mathbf{A}_3 + \mathbf{C}^{-1} \otimes S_{EQ}^{iso} \right) \quad (3.22)
\]

where

\[
\hat{\mathbf{C}} = 4J^{-4/3} \frac{\partial^2 \bar{W}_{EQ}^{iso}}{\partial \mathbf{C}^{iso} \otimes \partial \mathbf{C}^{iso}} \quad (3.23a)
\]

\[
\mathbf{A}_1 = \mathbf{C}^{-1} \otimes \mathbf{C} : \hat{\mathbf{C}} \quad (3.23b)
\]

\[
\mathbf{A}_2 = -J^{-2/3} \left( \mathbf{C} : \mathbf{S}^{EQ} \right) \left( \mathbf{C}^{-1} \otimes \mathbf{C}^{-1} \right) \quad (3.23c)
\]

\[
\mathbf{A}_3 = \mathbf{C}^{-1} \otimes J^{-2/3} \mathbf{S}^{EQ} \quad (3.23d)
\]

The operator ‘ \(\otimes\) ’ in Eq. (3.23c) has the definition as: \(A_{ab} \otimes B_{cd} = \frac{1}{2} (A_{ac}B_{bd} + A_{ad}B_{bc})\).

Using the same method, the material elasticity tensor for each non-equilibrium subnetwork can be obtained,

\[
2 \frac{\partial S_{BE}^{iso}}{\partial C} = \mathbf{P}_{BE}^T : \left\{ \left( \hat{\mathbf{C}}_{BE}^e - \frac{1}{3} \mathbf{A}_{BE}^1 \right) \right\} \left( \mathbf{P}_{BE}^{NEQ} \right)^T - \frac{2}{3} \left( \mathbf{A}_{BE}^2 + \mathbf{A}_{BE}^3 \right) \mathbf{P}_{BE}^T - \frac{2}{3} \mathbf{P}_{BE}^T : \mathbf{C}_{BE}^{-1} \otimes S_{BE}^{iso} \quad (3.24)
\]

where,

\[
\hat{\mathbf{C}}_{BE}^e = 4\left(J_{BE}^e\right)^{-4/3} \frac{\partial^2 \bar{W}_{BE}^{NEQ}}{\partial \mathbf{C}_{BE}^{iso,e} \otimes \partial \mathbf{C}_{BE}^{iso,e}} \quad (3.25a)
\]
The material elasticity tensor corresponding to the pure volume change is

\[ \mathbf{A}^1_{B_s} = \mathbf{C}^{-1}_{B_s} \otimes \mathbf{C}^{-1}_{B_s} : \mathbf{C}^e_{B_s} \]  

(3.25b)

\[ \mathbf{A}^2_{B_s} = -\left( \mathbf{J}^e_{B_s} \right)^{-2/3} \left( \mathbf{C}^e_{B_s} \otimes \mathbf{S}^{NEQ} \right) \left( \mathbf{C}^{-1}_{B_s} \otimes \mathbf{C}^{-1}_{B_s} \right) \]  

(3.25c)

\[ \mathbf{A}^3_{B_s} = \left( \mathbf{J}^e_{B_s} \right)^{-2/3} \mathbf{C}^e_{B_s} \otimes \mathbf{S}^{NEQ} \]  

(3.25d)

The material elasticity tensor corresponding to the pure volume change is

\[ 2 \frac{\partial \mathbf{S}^{vol}}{\partial \mathbf{C}} = 2\kappa \left[ \frac{2\mathbf{J}^2 - \mathbf{J}}{2} \mathbf{C}^{-1} \otimes \mathbf{C}^{-1} - \mathbf{J} (\mathbf{J} - 1) \mathbf{C}^{-1} \otimes \mathbf{C}^{-1} \right] \]  

(3.26)

For different constitutive models, the main difference of the spatial elasticity tensors lies in the selection for the strain energy density function to calculate \( \frac{\partial \mathbf{W}^{EQ}}{\partial \mathbf{C}^{iso}} \) and \( \frac{\partial \mathbf{W}^{NEQ}}{\partial \mathbf{C}^{iso,e}} \) as shown from Eqs.(3.22) to (3.25), which need to be determined explicitly in each iteration step. In the end, the general expression of the spatial elasticity tensor (or the Jacobian matrix) is obtained by substituting Eq.(3.22), Eq. (3.25) and Eq. (3.26) into Eq. (3.20).

### 3.3.3 Integration scheme of evolution law of internal variables

In addition to the stress and spatial elasticity tensor, the evolution of internal variables also needs to be numerically calculated in the FE framework. The evolution law of the internal variables has been provided in Section 2 in the form of differential equation as shown by Eq. (3.7). In this section, the integration scheme is presented to illustrate how the evolution equation is solved using the incremental method.

When the deformation gradient tensor \( \mathbf{F} \) is symmetric, the evolution Eq. (3.7) can be easily solved, for example, the cases for uniaxial tension tests as presented by Zhou et. al [48]. However, in general case where \( \mathbf{F} \) is not necessarily symmetric, the evolution of the inelastic deformation can not be fully determined. Without losing the generality for polymers, the simplest possible approach to solve this issue is to discard the spin of the
inelastic deformation, as suggested by Hong [34] and Boyce et al. [22]. For such a case, the skew symmetric part of $\mathbf{L}_{B_n}^i$ is discarded, i.e., $\mathbf{W}_{B_n}^i = 0$, and Eq. (3.7) is re-written as,

$$
\left[ \mathbf{F}_{B_n}^e \mathbf{L}_{B_n}^i \left( \mathbf{F}_{B_n}^e \right)^T \right] \left( \mathbf{B}_{B_n}^e \right)^{-1} = \mathbf{y}^{-1} : \mathbf{\tau}_{B_n}
$$

(3.27)

or,

$$
\dot{\mathbf{F}}_{B_n}^i = \left( \mathbf{F}_{B_n}^e \right)^{-1} \left( \mathbf{y}^{-1} : \mathbf{\tau}_{B_n} \right) \mathbf{B}_{B_n}^e \left( \mathbf{F}_{B_n}^e \right)^{-T} \mathbf{F}_{B_n}^i
$$

(3.28)

With the time discretisation, the evolution law can be expressed using the backward Euler method as,

$$
\left( \dot{\mathbf{F}}_{B_n}^i \right)_{t+\Delta t} = \mathbf{M} \left( \mathbf{F}_{B_n}^i \right)_{t+\Delta t} = \mathbf{M} \left[ \left( \mathbf{F}_{B_n}^i \right)_{t} + \left( \Delta \mathbf{F}_{B_n}^i \right)_{t \rightarrow t+\Delta t} \right]
$$

(3.29)

with the approximation that $\left( \Delta \mathbf{F}_{B_n}^i \right)_{t \rightarrow t+\Delta t} = \left( \mathbf{F}_{B_n}^i \right)_{t+\Delta t} - \left( \mathbf{F}_{B_n}^i \right)_{t} \approx \left( \dot{\mathbf{F}}_{B_n}^i \right)_{t+\Delta t} \Delta t$, the inelastic deformation gradient tensor for the next step can be explicitly calculated as,

$$
\left( \mathbf{F}_{B_n}^i \right)_{t+\Delta t} = \left( \mathbf{F}_{B_n}^i \right)_{t} + \left( \mathbf{I} - \mathbf{M} \Delta t \right)^{-1} \mathbf{M} \left( \mathbf{F}_{B_n}^i \right)_{t} \Delta t
$$

(3.30)

Within this integration scheme, the evolution of the internal variables can be numerically and uniquely determined. To summarize, the algorithm of finite element analysis with the corresponding time discretisation is shown in Fig. 3.2.
3.4 FEM models and simulation results

In this section, the deformation behaviors of three commonly used elastomers, namely VHB4910, carbon black (CB) filled elastomers, and HNBR50 are simulated by using the proposed FE model. Different constitutive models with strain energy density functions being expressed in terms of either strain invariants or principal stretches are adopted for the materials. Comparisons between the computational results and the theoretical analysis
under simple loading conditions for each material are used as the validation of the model accuracy. Further numerical simulations for materials under complex loading conditions demonstrate the capacity and the robustness of the proposed numerical model.

### 3.4.1 Simulation of VHB 4910

In this case study, the viscoelastic deformation of the unfilled elastomer VHB 4910 is simulated under different external loads. The Gent model [30] is taken to describe the hyperelastic behavior of the material and the inelastic deformation. With the adoption of the multiplicative decomposition scheme for the implementation of the material incompressibility, the isochoric part of the Gent strain energy density function is expressed as,

\[
\mathcal{W}^{EQ} = -\frac{G^{EQ}}{2} J_{\text{lim}} \ln \left[ \frac{J_{\text{lim}} - I_{1}^{iso}}{J_{\text{lim}}} + 3 \right] 
\]  

(3.31)

for the equilibrium network, and

\[
\mathcal{W}^{NEQ}_{B_n} = -\frac{G^{NEQ}}{2} J_{\text{lim}} \ln \left[ \frac{J_{\text{lim}} - (I_{1}^{iso,e})_{B_n}}{J_{\text{lim}}} + 3 \right] 
\]

(3.32)

for the non-equilibrium subnetworks. \( T_1 \) is the first invariant of \( \mathbf{C}^{iso} = J^{-2/3} \mathbf{C} \) and \( (I_{1}^{iso,e})_{B_n} \) is the first invariant of \( \mathbf{C}^{iso,e} = (J_{B_n}^{e})^{-2/3} \mathbf{C}^{e}_{B_n} \) for each non-equilibrium subnetwork in the rheological model. \( G^{EQ} \) and \( G^{NEQ}_{B_n} \) are the macro-scale equilibrium shear modulus and non-equilibrium shear modulus, respectively, related to the number of polymer chains per unit volume in the corresponding networks. \( J_{\text{lim}} \) is material extensibility parameter expressed as \( J_{\text{lim}} = 3(N - 1) \) with \( N \) being the polymerization degree of chains.

To update the stress tensor and the spatial elasticity tensor in each step in the FE model, the first and second partial derivatives of the isochoric free energy density functions with
respect to $C^{iso}$ and $C^{iso,e}_{B_n}$ in Eqs. (3.15a), (3.15b), (3.22), and (3.24) need to be determined. Correspondingly,

\[ \overline{S}^{EQ} = \frac{G^{EQ} J_{lim}}{J_{lim} - tr\left(C^{iso}\right) + 3} I \]  

(3.33a)

\[ \overline{S}^{NEQ}_{B_n} = \frac{G^{NEQ}_{B_n} J_{lim}}{J_{lim} - tr\left(C^{iso,e}_{B_n}\right) + 3} I \]  

(3.33b)

\[ \frac{\partial^2 \overline{W}^{EQ}}{\partial C^{iso} \otimes \partial C^{iso}} = \frac{G^{EQ} J_{lim}}{2\left(J_{lim} - tr\left(C^{iso}\right) + 3\right)^2} I \otimes I \]  

(3.34a)

\[ \frac{\partial^2 \overline{W}^{NEQ}}{\partial C^{iso,e}_{B_n} \otimes \partial C^{iso,e}_{B_n}} = \frac{G^{NEQ}_{B_n} J_{lim}}{2\left(J_{lim} - tr\left(C^{iso,e}_{B_n}\right) + 3\right)^2} I \otimes I \]  

(3.34b)

After obtaining the Cauchy stress tensor and second Piola-Kirchhoff stress tensor from the FE simulation results, the nominal stress is then calculated as $\sigma_n = \sigma F^{-1}$. To validate the accuracy of the FE model, when the material is under a uniaxial tension, the nominal stress and the inelastic stretch evolution from the FE model are compared with those from the theoretical modeling. In the theoretical analysis, the nominal stress $\sigma_n$ of the VHB 4910 under a uniaxial tension in the tension direction can be explicitly expressed as [48],

\[ \sigma_n = \frac{G^{EQ} J_{lim} \left(\lambda - \lambda^{-2}\right)}{J_{lim} - 2\lambda^{-1} - \lambda^2 + 3} + \sum_{n=1}^{N} \frac{G^{NEQ}_{n} J_{lim} \left[\lambda \left(\lambda_{B_n}^i\right)^{-2} - \lambda^{-2} \lambda_{B_n}^i\right]}{J_{lim} - 2\lambda^{-1} \lambda_{B_n}^i - \lambda^2 \left(\lambda_{B_n}^i\right)^{-2} + 3} \]  

(3.35)

where $\lambda$ and $\lambda_{B_n}^i$ are the total stretch and the inelastic stretch, respectively. Considering the material incompressibility, the deformation gradient tensors thus take the forms of
The inelastic stretch $\lambda_{B_n}^i$ in the non-equilibrium subnetwork evolves following the thermodynamics evolution law as,

$$\frac{d\lambda_{B_n}^i}{dt} = \frac{G_n^{\text{NEQ}}} {3\eta_n} J_{\text{lim}}\lambda_{B_n}^i \alpha(F)^2 \left[ (\lambda_{B_n}^i)^2 - (\lambda_{B_n}^i)^{-1} + 3 \right]$$

(3.36)

where $\eta_n (n = 1, 2, \ldots N)$ is the viscosity of each undeformed subnetwork. Selecting 3 subnetworks, Zhou et al., [48] conducted data fitting from the experimental measurements of Wang et al. [35] and Hossain et al. [68] to obtain all the material parameters for VHB 4910, which are listed in Table 3.1. These parameters are used as input for the UMAT subroutine for the FE analysis.

| Table 3.1 Material parameters for VHB 4910 [48] |
|-----------------|-----------------|-----------------|-----------------|-----------------|
| Parameter       | $G_1^{\text{EQ}}$ | $G_1^{\text{NEQ}}$ | $G_2^{\text{NEQ}}$ | $G_3^{\text{NEQ}}$ |
| Value           | 14.59 KPa        | 11.87 KPa        | 43.15 KPa        | 19.75 KPa        |
| Parameter       | $J_{\text{lim}}$ | $\eta_1$         | $\eta_2$         | $\eta_3$         |
| Value           | 115.8            | 11.4 MPa s       | 43.17 KPa s      | 381.16 KPa s     |

In the FE model, a cube with dimension of $1\times1\times1$ is meshed with mesh size of $0.1\times0.1\times0.1$. The cube is first stretched with a stretch ratio of 2 and then unloaded to a stress-free state (Fig. 3.3). The stretch rates are set as 0.01/s, 0.1/s and 1/s respectively. The stress-stretch curves for both the FE analysis and theoretical results are presented in Fig. 3.4 for comparison, which demonstrates a good agreement. It shows that the FE
analysis provides excellent quantitative prediction of the loading-unloading responses of the material and can well capture the stretch rate-dependent response.

Figure 3.3 Loading and unloading process of the cube sample.

Figure 3.4 Comparison of loading-unloading stress response curves of VHB4910 under different stretch rates between FE model and theoretical results.

In order to further validate the accuracy of the FE model and explain the variation of stress response, the inelastic stretch evolution during the uniaxial tension with different stretch rates is presented in Fig. 3.5, compared with the theoretical results calculated directly from Eq. (3.36). The large relative errors in the first subnetwork at $|\dot{\lambda}|=1/ s$ are
originated from the precision limit that is set as 0.001 to increase the computation efficiency in the FE analysis. When the stretch rate is high, the inelastic stretch in this subnetwork hardly increases, while the elastic stretch is almost the same as the equilibrium stretch. Thus, the large relative errors in the inelastic stretch will not cause notable error in the elastic stretch and hence the elastic stress in the non-equilibrium subnetworks. As indicated in this figure, due to the viscosity of the elastomers, the inelastic stretch in each subnetwork also depends on the stretch rate. When the stretch rate is lower, there is more time for the non-equilibrium subnetwork to relax, it will thus cause more rapid variation of the inelastic stretch during the loading process. Therefore, the elastic stress in each subnetwork tends to be lower in the loading process, while the residual deformation tends to be larger after the unloading process is completed.

Figure 3.5 Inelastic stretch evolution in VHB 4910 sample under different stretch rates from FE simulation and theoretical analysis.
The capacity of the FE model is also tested for the response of VHB 4910 within a wider range of deformation (up to $\lambda = 9$) and stretching rate (0.01/s to 1/s). Excellent agreement is demonstrated between the numerical simulation and the theoretical results as shown in Fig. 3.6. It is also observed that for relatively low stretching rates, the loading and unloading curves almost overlap when the material undergoes relatively large deformation. As the material viscosity decreases with the increase of the deformation as shown by Eq. (3.9), it will allow the material to relax much faster at larger deformation. However, with the increase of the stretching ratio, a wider gap between the loading and unloading curves is observed as there is less time for the elastomer to relax during this process. The good agreement between the FE simulation results and the theoretical analysis in Fig. 3.4 to Fig. 3.6 have thus validated the accuracy of the developed FE model.

![Figure 3.6 Comparison of stress response curves for finite-deformation of VHB 4910 sample under different stretch rates between FE model and theoretical analysis.](image-url)

The capacity of the FE model in modeling the deformation under non-uniform stress condition is also tested on VHB 4910. A more general case is for a complex geometry, which is subjected to a cyclic simple shear as shown in Fig. 3. 7. The shear is applied with different loading rates $|\nu| = 0.05\,mm/s$ and $|\nu| = 0.5\,mm/s$, respectively. The
distribution of the Cauchy stress $\sigma_{12}$ in the shear direction (denoted as S12 in Abaqus as shown in this figure) within the geometry when the displacement U reaches 5mm is presented in Fig. 3.8. It is observed that the stress distribution patterns are very similar, with the maximum shear stress occurring in the middle of the outer surface. For faster loading, the shear stress amplitude is higher, demonstrating the loading rate effect due to the material viscosity.

Figure 3.7 Model dimensions and load specification.

Figure 3.8 Stress distribution in the model under different shear loading rates.
In order to visualize the effect of the material viscosity during the deformation, the resultant force is presented in Fig. 3.9. Typical features of the viscoelastic behavior of the material are observed in the simulation, i.e., the residual deformation during the unloading process. Also the maximum resultant force due to the applied shear deformation depends on the loading rate, and the maximum difference is about 10 N for this case. From these simulations, it is concluded that the developed FE model is capable of characterizing the viscoelastic response of elastomers with complex geometry and under non-homogenous stress conditions.

![Figure 3.9](image)

**Figure 3.9** Cyclic resultant force evolution under different loading rates from FE simulation.

### 3.4.2 Simulation of carbon black (CB) filled elastomers

To further exhibit the modeling capacity of the UMAT in the FE platform with the adoption of various constitutive models, the stress response of carbon filled elastomers is also simulated. Same as our previous study [48], the Gent strain energy density [30] is taken for the equilibrium network in the rheological model (same as Eq. (3.31)), while the Ogden model [25] is used for the non-equilibrium subnetworks, i.e.,
\[ W_{NEQ}^{B_n} = \sum_{k=1}^{K} \frac{\mu_{k,n}}{\alpha_{k,n}} \left( \lambda_{1_k,n}^{\alpha_{k,n}} + \lambda_{2_k,n}^{\alpha_{k,n}} + \lambda_{3_k,n}^{\alpha_{k,n}} - 3 \right) \]  

(3.37)

where \( \lambda_{1,n}, \lambda_{2,n}, \) and \( \lambda_{3,n} \) are the principal stretches of the \( n \)th viscous subnetwork. \( \mu_{k,n} \) and \( \alpha_{k,n} \) are material constants, and \( K \) is commonly taken as 3. Detailed information regarding these constants can be referred to Ehret [69]. In this study, only one viscous subnetwork is considered with \( n=1 \), and the subscript \( B_n \) is hence omitted for convenience. Different from the Gent model in which the strain invariants are involved, the Ogden strain energy density is in terms of the principal stretches. In order to enforce the material incompressibility, one needs to calculate the stress in Eq. (3.15b) which involves the derivative of the isochoric strain energy density function in the form of eigenvalues of \( \bar{C}^e \). Therefore, the Ogden strain energy density function is then transformed into the following format,

\[ \tilde{W}_{NEQ} = \sum_{k=1}^{3} \frac{\mu_k}{\alpha_k} \left( \bar{\Lambda}_1^{\alpha_k/2} + \bar{\Lambda}_2^{\alpha_k/2} + \bar{\Lambda}_3^{\alpha_k/2} - 3 \right) \]  

(3.38)

where \( \bar{\Lambda}_j = \left( \bar{\Lambda}^e_j \right)^2 \) (\( j=1,2,3 \)) are the eigenvalues of the right Cauchy-Green deformation tensor \( C_{B_n}^{iso,e} \).

The expression of \( \bar{S}_{NEQ} = 2 \frac{\partial \tilde{W}_{NEQ}}{\partial C_{B_n}^{iso,e}} \) adopted from the formulation by Feng et. al. [59] in Eq. (3.15b) is hereby dependent on the existence of the eigenvalues of \( C_{B_n}^{iso,e} \), with different scenarios listed as:

1. When three distinct eigenvalues exist (\( \bar{\Lambda}_1 > \bar{\Lambda}_2 > \bar{\Lambda}_3 \)), \( \bar{S}_{NEQ} \) can be expressed as,

\[ \bar{S}_{NEQ} = \sum_{k=1}^{3} \mu_k \sum_{a=1}^{3} \left( \frac{\alpha_k-2}{\bar{\Lambda}_a^2} \bar{M}_a \right) \]  

(3.39)
where $\mathbf{M}_a$ is the eigenbasis with respect to each unit eigenvector, i.e., $\mathbf{M}_a = \vec{v}_a \otimes \vec{v}_a$. Specifically, QR decomposition iteration algorithm is adopted to numerically determine the eigenvalues and the eigenvectors, instead of deriving the explicit expressions that might take more efforts. Meanwhile, to calculate the material elasticity tensor in Eq. (21), the second order derivative of the strain energy density function with respect to $\bar{\mathbf{C}}^e$ is calculated on the basis of Eq. (3.39), i.e.,

$$\frac{\partial^2 \bar{W}^{\text{NEQ}}}{\partial \mathbf{C}_{B_n}^{\text{iso},e} \otimes \partial \mathbf{C}_{B_n}^{\text{iso},e}} = \frac{1}{2} \sum_{k=1}^{3} \mu_k \left\{ \frac{\alpha_k - 2}{2} \sum_{a=1}^{3} \Lambda_a^{-2} \mathbf{M}_a \otimes \mathbf{M}_a + \sum_{a=1}^{3} \left[ \frac{\alpha_k - 2}{\Lambda_a^{-2} - \Lambda_b^{-2}} \left( \mathbf{M}_a \otimes \mathbf{M}_b + \mathbf{M}_b \otimes \mathbf{M}_a \right) \right] \right\}$$ (3.40)

In this equation, $b$ is the sequential index after $a$ in the circular permutation (1,2,3).

(2) When two distinct eigenvalues exist, ( $\bar{\Lambda}_1 = \bar{\Lambda}_2 > \bar{\Lambda}_3$, or $\bar{\Lambda}_1 > \bar{\Lambda}_2 = \bar{\Lambda}_3$), $\bar{\mathbf{S}}^{\text{NEQ}}$ has the similar expression with Eq. (3.39) as,

$$\bar{\mathbf{S}}^{\text{NEQ}} = \sum_{k=1}^{3} \mu_k \left( \frac{\alpha_k - 1}{\Lambda_{\text{non}}^{-2}} \mathbf{M}_{\text{non}} + \frac{\alpha_k - 1}{\Lambda_{\text{rep}}^{-2}} \mathbf{M}_{\text{rep}} \right)$$ (3.41)

The subscript ‘rep’ stands for the repeated eigenvalues while the ‘non’ is for the non-repeated ones. The rank 2 eigenbasis of repeated eigenvalues can be directly calculated as,

$$\mathbf{M}_{\text{rep}} = 1 - \mathbf{M}_{\text{non}}$$ (3.42)

Based on Eq. (41) and using L’ Hôpital’s rule, the second order derivative of the isochoic strain energy density function with respect to $\bar{\mathbf{C}}^e$ can be formulated as,

$$\frac{\partial^2 \bar{W}^{\text{NEQ}}}{\partial \mathbf{C}_{B_n}^{\text{iso},e} \otimes \partial \mathbf{C}_{B_n}^{\text{iso},e}} = \frac{1}{2} \sum_{k=1}^{3} \mu_k \left\{ \frac{\alpha_k - 2}{2} \sum_{a=1}^{3} \Lambda_a^{-2} \mathbf{M}_a \otimes \mathbf{M}_a + \sum_{a=1}^{3} \left[ \frac{\alpha_k - 2}{\Lambda_a^{-2} - \Lambda_{\text{rep}}^{-2}} \left( \mathbf{M}_a \otimes \mathbf{M}_{\text{rep}} + \mathbf{M}_{\text{rep}} \otimes \mathbf{M}_a \right) \right] \right\}$$ (3.43)
(3) When three identical eigenvalues exist, i.e., $\lambda_1 = \lambda_2 = \lambda_3$, the corresponding tensor $\mathbf{S}^{NEQ}$ and the second derivative of the isochoric strain energy density with respect to $\mathbf{C}^e$ are calculated as,

$$
\mathbf{S}^{NEQ} = \sum_{k=1}^{3} \mu_k \left( \frac{\alpha_k - 1}{2} \right) \mathbf{I} \tag{3.44}
$$

$$
\frac{\partial^2 W^{NEQ}}{\partial \mathbf{C}_{iso,e}^{B_k} \otimes \partial \mathbf{C}_{iso,e}^{B_k}} = \sum_{k=1}^{3} \frac{\mu_k}{2} \left( \frac{\alpha_k - 1}{2} \right) \Lambda^\frac{\alpha_k - 2}{2} \mathbf{I} \otimes \mathbf{I} \tag{3.45}
$$

According to these different situations, the second Piola-Kirchhoff stress tensor in Eq. (3.14) and the spatial elasticity tensor in Eq. (3.20) can thus be updated by using equations Eq. (3.39) to Eq. (3.45). And the nominal stress is thereafter obtained as $\sigma_n = J \mathbf{F}^{-1} \sigma$.

Cyclic uniaxial tension test is first simulated to calibrate the material parameters for CB filled elastomer from the experimental data. The nominal stress $\sigma_n$ is calculated explicitly when the material is uniaxially stretched with a stretch ratio $\lambda$, as,

$$
\sigma_n = \frac{G^{EQ} J_{lim} \left( \lambda - \lambda^{-2} \right)}{J_{lim} - 2 \lambda^{-1} - \lambda^2 + 3} + \sum_{k=1}^{3} \mu_k \left[ \lambda^{\alpha_k - 1} \left( \lambda^i \right)^{-\alpha_k} - \lambda^{-2}\alpha_k - (\lambda^i)^{\alpha_k/2} \right] \tag{3.46}
$$

The inelastic stretch $\lambda^i$ is determined from the thermodynamics evolution law Eq. (3.7),

$$
\frac{d \lambda^i}{dt} = \sum_{k=1}^{3} \lambda^i \alpha \frac{\mathbf{F}}{2} \frac{3 \eta_i}{\eta_k} \mu_k \left[ \lambda^{\alpha_k} \left( \lambda^i \right)^{-\alpha_k} - \lambda^{\alpha_k/2} \left( \lambda^i \right)^{\alpha_k/2} \right] \tag{3.47}
$$

The stress response to the applied stretch with stretch rate of 0.02/s is plotted in Fig. 3.10 in comparison to the experimental measurements [69]. It should be noted that Fig. 3.10 is the correction of Ref. [48]. Both the material parameters of the elastic ground network and the viscous subnetwork are obtained together through data fitting, which are listed in Table 3.2. It should be mentioned that the less satisfactory fitting to the experimental
results may come from the fact that only one viscous subnetwork is modeled in the Ogden model. Meanwhile, Mullins effect [70] observed in the experiments, i.e., the stress softening in the successive loading/unloading cycles due to the change of the material properties, is also captured by the theoretical modeling results.

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Table 3.2 Material parameters for CB filled elastomer

To validate the accuracy of the FE model, uniaxial tension tests with different stretch rates are then simulated by the FE model and are compared with the results from the theoretical analysis. The stress response curves for the first two loading/unloading cycles are plotted in Fig. 3.11. In this simulation, the total uniaxial stretch is prescribed as 3 and the stretch rates are set as 0.02/s, 0.05/s and 0.1/s, respectively. It is observed from this figure, the FE simulated results exhibit an excellent agreement with the theoretical calculations, indicating the capability and accuracy of the FE model. The Mullins softening effect observed in these figures has been explained as the result of energy dissipation during the diffusion process of the polymer chains from the physical perspective [48]. Such a phenomenon can also be intuitively interpreted with the evolution of the inelastic stretch as shown in Fig. 3.12. In the prior loading/unloading process, the inelastic stretch keeps increasing to a relatively large value. Therefore, the elastic stretch in the subsequent reloading process tends to be smaller than that in the previous loading process, and hence the elastic stress is smaller under the same total stretch.
Figure 3.10 Data fitting of material parameters for CB filled elastomers.

Figure 3.11 Stress response curves of CB filled elastomers under different stretch rates from FE simulation and theoretical analysis.
Figure 3.12 Inelastic stretch evolution in the CB filled elastomer sample under different stretch rates during the first loading-unloading cycle from FE simulation and theoretical analysis.

3.4.3 Simulation of HNBR50

Another commonly used material, HNBR50, is also tested to validate the capacity of FE model with the implementation of different constitutive models. The Gent model [30] is applied for the non-equilibrium subnetworks, while the non-affine network model [44] is adopted to describe the elastic response of the material. For this non-affine network model, the strain energy density function is in the form of a mixture of principal stretches and strain invariants, i.e.,

\[
W_{EQ} = \frac{1}{6} G_c I_1 - G_c \lambda_{\text{max}}^2 \ln \left( 3 \lambda_{\text{max}}^2 - I_1 \right) + \sum_{k=1}^{2} G_c \left( \lambda_1^{\alpha_k} + \lambda_2^{\alpha_k} + \lambda_3^{\alpha_k} \right) \tag{3.48}
\]

where \( \alpha_1 = 1, \alpha_2 = -1 \), and \( \lambda_{\text{max}} \) is the material parameter related to the extensibility of the polymer chains [44]. The cross-linking modulus \( G_c \) and the entanglement modulus \( G_e \) are related to the polymer physics parameters. In the FE model, considering the incompressibility constraint by using the multiplicative decomposition scheme, the isochoric strain energy density function is modified as,
\[
\tilde{W}^{EQ} = \frac{1}{6} G_c I_1^{iso} - G_c \lambda_\text{max}^2 \ln \left(3 \lambda_\text{max}^2 - I_1^{iso}\right) + \sum_{k=1}^{2} \frac{G_c}{2} \left(\bar{\Gamma}_1^{a_k/2} + \bar{\Gamma}_2^{a_k/2} + \bar{\Gamma}_3^{a_k/2}\right) 
\]

with \( \bar{\Gamma}_j \) being the eigenvalues of isochoric right Cauchy-Green deformation tensor \( C^{iso} \).

Similar to the CB filled elastomers, the stress tensor and the spatial elasticity tensor are formulated depending on the number of non-repeated eigenvalues of the right Cauchy-Green deformation tensor.

(1) When three distinct eigenvalues exist (\( \bar{\Gamma}_1 > \bar{\Gamma}_2 > \bar{\Gamma}_3 \)), \( \tilde{S}^{EQ} \) is formulated as,

\[
\tilde{S}^{EQ} = \frac{1}{3} G_c I + \frac{2 G_c \lambda_\text{max}^2}{3 \lambda_\text{max}^2 - I_1^{iso}} I + G_c \sum_{a=1}^{3} \left(\bar{\Gamma}_a - \bar{\Gamma}_a^{-1/2} - \bar{\Gamma}_a^{-3/2}\right) M_a 
\]

The second-order derivative of the isochoric strain energy density function for the equilibrium network with respect to \( C^{iso} \) is determined as,

\[
\frac{\partial^2 \tilde{W}^{EQ}}{\partial C^{iso} \otimes \partial C^{iso}} = \frac{G_c \lambda_\text{max}^2}{\left(3 \lambda_\text{max}^2 - I_1^{iso}\right)^2} I \otimes I + G_c \sum_{k=1}^{2} \left(\frac{\alpha_k - 2}{2} \sum_{a=1}^{3} \frac{\alpha_k - 4}{2} M_a \otimes M_a \right) 
\]

\[
+ \sum_{a=1}^{3} \left[\frac{\alpha_k - 2}{2} \frac{\alpha_k - 2}{2} \frac{\Gamma_a - \Gamma_a^{-2}}{\bar{\Gamma}_a - \bar{\Gamma}_b}\right] \left(M_a \otimes M_b + M_b \otimes M_a\right) \right] \}
\]

where \( b \) is the sequential index after \( a \) in the circular permutation (1,2,3).

(2) When two distinct eigenvalues exist (\( \bar{\Gamma}_1 > \bar{\Gamma}_2 = \bar{\Gamma}_3 \), or \( \bar{\Gamma}_1 = \bar{\Gamma}_2 > \bar{\Gamma}_3 \)), the corresponding expressions for \( \tilde{S}^{EQ} \) and the second-order derivative of the isochoric strain energy density function with respect to \( C^{iso} \) are determined as,

\[
\tilde{S}^{EQ} = \frac{1}{3} G_c I + \frac{2 G_c \lambda_\text{max}^2}{3 \lambda_\text{max}^2 - I_1^{iso}} I + G_c \sum_{k=1}^{2} \left(\frac{\alpha_k - 2}{2} \frac{\alpha_k - 4}{2} M_{non} + \alpha_k \frac{\alpha_k - 2}{2} M_{rep}\right) 
\]

(3.51)
\[
\frac{\partial^2 W^{\text{EQ}}}{\partial C^{\text{iso}} \otimes \partial C^{\text{iso}}} = \frac{G_c \lambda_{\text{max}}^2}{(3 \lambda_{\text{max}}^2 - \lambda_1^{\text{iso}})^2} I \otimes I + G_c \sum_{k=1}^{2} \left( \frac{\alpha_k}{2} - 1 \right) \left( \frac{\alpha_k}{2} - 1 \right) \left[ \frac{\lambda_{\text{iso}}}{\lambda_{\text{max}}} \otimes M_{\text{non}} \otimes M_{\text{non}} + \frac{\lambda_{\text{iso}}}{\lambda_{\text{rep}}} M_{\text{rep}} \otimes M_{\text{rep}} \right] \]
(3.53)

(3) When three repeated eigenvalues exist \((\bar{\Gamma}_1 = \bar{\Gamma}_2 = \bar{\Gamma}_3)\), the corresponding expressions are determined as,

\[
\bar{S}^{\text{EQ}} = \frac{1}{3} G_c I + \frac{2 G_c \lambda_{\text{max}}^2}{3 \lambda_{\text{max}}^2 - \lambda_1^{\text{iso}}} I + G_c \sum_{k=1}^{2} \left( \frac{\alpha_k}{2} - 1 \right) \left( \frac{\alpha_k}{2} - 1 \right) \left[ \frac{\lambda_{\text{iso}}}{\lambda_{\text{max}}} \otimes M_{\text{non}} \otimes M_{\text{non}} + \frac{\lambda_{\text{iso}}}{\lambda_{\text{rep}}} M_{\text{rep}} \otimes M_{\text{rep}} \right] \]
(3.54)

\[
\frac{\partial^2 W^{\text{EQ}}}{\partial C^{\text{iso}} \otimes \partial C^{\text{iso}}} = \frac{G_c \lambda_{\text{max}}^2}{(3 \lambda_{\text{max}}^2 - \lambda_1^{\text{iso}})^2} I \otimes I + G_c \sum_{k=1}^{2} \left( \frac{\alpha_k}{2} - 1 \right) \left( \frac{\alpha_k}{2} - 1 \right) \left[ \frac{\lambda_{\text{iso}}}{\lambda_{\text{max}}} \otimes M_{\text{non}} \otimes M_{\text{non}} + \frac{\lambda_{\text{iso}}}{\lambda_{\text{rep}}} M_{\text{rep}} \otimes M_{\text{rep}} \right] \]
(3.55)

In order to validate the accuracy of the FE model, a uniaxial tension test is studied. The explicit expression of the nominal stress of the elastomer under a uniaxial stretch \(\lambda\) can be written as [48],

\[
\sigma_n = \frac{1}{3} G_c (\lambda - \lambda^{-2}) + \frac{2 G_c \lambda_{\text{max}}^2}{3 \lambda_{\text{max}}^2 - \lambda^2 - 2 \lambda^{-1}} + G_c \left( 1 - \lambda^{-2} + \lambda^{-1/2} - \lambda^{-3/2} \right)
\]

(3.56)

Since the same constitutive model is adopted in the non-equilibrium subnetworks as in VHB4910, the evolution of the inelastic stretch also follows Eq. (3.36), but with different material parameters. For modeling HNBR50, five non-equilibrium subnetworks are adopted. The corresponding material parameters were determined by data fitting with the experimental data [48], which are listed in Table 3.3.
Table 3.3 Material parameters for HNBR50 [48]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Parameter</th>
<th>Value</th>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$G_c$</td>
<td>0.08 MPa</td>
<td>$\eta_2$</td>
<td>4.32 MPa s</td>
<td>$G_s^{x\xi}$</td>
<td>0.12 MPa</td>
</tr>
<tr>
<td>$G_e$</td>
<td>0.21 MPa</td>
<td>$\eta_3$</td>
<td>24.96 MPa s</td>
<td>$G_s^{x\xi}$</td>
<td>0.03 MPa</td>
</tr>
<tr>
<td>$\dot{\lambda}_{\text{max}}$</td>
<td>2.44</td>
<td>$\eta_4$</td>
<td>156.5 MPa s</td>
<td>$G_s^{x\xi}$</td>
<td>0.10 MPa</td>
</tr>
<tr>
<td>$J_{\text{lim}}$</td>
<td>14.87</td>
<td>$\eta_5$</td>
<td>12.15 MPa s</td>
<td>$G_s^{x\xi}$</td>
<td>0.15 MPa</td>
</tr>
<tr>
<td>$\eta_t$</td>
<td>0.45 MPa s</td>
<td>$G_t^{x\xi}$</td>
<td>0.45 MPa</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

With different stretch rates of 0.05/min, 0.5/min, and 5/min, the stress response curves from the FE analysis are plotted in Fig. 3.13 in comparison to the theoretical analysis. The FE model is further tested for cyclic uniaxial tension-compression tests [43] under the stretch rate of $|\dot{\lambda}| = 3$/min with a one-hour relaxation after each loading period. The simulated results from FE model are compared with theoretical calculation [48], as illustrated in Fig. 3.14, which has demonstrated an excellent agreement. From this case study considering both cyclic uniaxial tension-compression and loading-relaxation tests, the accuracy and the capability of the UMAT developed in the FE framework in modeling the responses of elastomers under complex loading conditions are validated again. Moreover, the merit of the continuum mechanics framework in adopting various hyperelastic constitutive relations [24] and the easy implementation in the FE platform has also been verified.
Figure 3.13 Nominal stress response of HNBR50 at different stretch rates. (a): 5/min; (b): 0.5/min; (c): 0.05/min.
Figure 3.14 Stress response curves in loading/unloading stretch test with a one-hour relaxation after each loading period from FE simulation and, theoretical analysis. The stretch rate is 0.05/s

3.5 Conclusion

In this work, a finite element framework has been established to simulate the finite deformation viscoelasticity of elastomers with the implementation of a viscoelastic constitutive model into the UMAT subroutine, in which the deformation-dependent material viscosity and the microstructure features are incorporated. The
incompressibility of the elastomers is successfully constrained using the volumetric and isochoric multiplicative decomposition scheme in the FE framework. The general forms of stress tensor and spatial elasticity tensor are formulated in the UMAT subroutine for constitutive relations involving either strain invariants or principal stretches. Therefore, the developed computational framework is capable of adopting most constitutive models for hyperelasticity and thermodynamics evolution laws for viscoelastic materials. Numerical simulations are conducted to predict the stress responses and inelastic deformation evolution of three types of commonly used elastomeric materials under simple uniaxial tension or compression loading conditions. The excellent agreement with the theoretical analysis validates the accuracy of the developed FE model and demonstrates the adaptiveness of the FE framework for most of the constitutive relations of viscoelastic materials. In addition, case studies are also performed for predicting the viscoelastic responses of elastomers with complex configuration and under complex loads. It is observed that the developed FE model exhibits strong capability of capturing the typical viscoelastic response behaviors of elastomeric materials. In summary, this work is expected to provide a general approach and a universal computation platform for characterizing the viscoelastic responses and evaluating the performance of elastomers, which will help to facilitate novel design and optimization of elastomer-based structures in engineering applications.

Reference


Chapter 4

4 A numerical study on the instabilities of viscoelastic dielectric elastomers considering nonlinear material viscosity

Instability has been recognized as one of the major issues restricting the full potential applications of dielectric elastomer (DE)-based devices. While great efforts have been devoted to investigating the instabilities of DE actuators, those studies are limited to using either hyperelastic material models or viscoelastic constitutive models with constant material viscosity. As observed in the experiments, the intrinsic material viscosity of elastomers varies with deformation, which becomes more manifest particularly for DE actuators undergoing large deformation. This work attempts to fill this knowledge gap by developing a finite element (FE) framework that combines the nonlinear field theory with the micro-macro constitutive model incorporating nonlinear material viscosity to investigate the electromechanical responses and the instability of DE actuators. A highly customized user-element subroutine (UEL) in Abaqus is developed for the FE implementation. The effects of the nonlinear material viscosity on a variety of instability modes of DE (VHB 4910) actuators with different configurations are numerically investigated, including electromechanical instability (EMI), buckling, wrinkling, and crumpling. The accuracy and robustness of the FE framework are validated by comparison with existing experimental data and analytical studies. This work provides a general approach for instability analysis of DE actuators with different configurations and can further function as a universal platform for numerical analysis on the electromechanical finite deformation of DE structures with complex configurations, leading to better design and applications of DE-based devices.

4.1 Introduction

Featured by softness, flexibility, large deformation capacity and electromechanical coupling, dielectric elastomers (DEs) have gained great attention in recent years. Such unique properties have facilitated the applications of DEs in many engineering and industrial fields, including artificial muscles [1, 2], sensors [3, 4], soft robots [5-7], and
energy harvesting devices [8, 9]. Nevertheless, the research community has also recognized that the restricted actuation capacity of DEs due to the material and the structural instabilities such as electromechanical instability (EMI), buckling, wrinkling, and crumpling, has become a crucial issue hindering the full potential development and applications of DE-based devices. Under this circumstance, it becomes essential to establish a computational framework, especially one capable of revealing the deformation mechanisms of DEs, to analyze the actuation process and different instability modes of DE structures.

Different instability modes might occur for DE actuators depending on configurations and boundary constraints of the structure [10]. With free boundaries, it is found in the literature that DE actuators may encounter EMI, which is also known as the snap through instability featured by the excessive thinning of the DE plate under the electric field [11]. Early studies on the EMI of DEs were commonly based on the theoretical modeling framework [12, 13] by adding the empirical Maxwell stress into the hyperelasticity theories to characterize the instantaneous response of DEs to an actuation voltage. For example, Wissler and Mazza [14] derived analytical solutions for a circular DE actuator, and systematically investigated the material EMI with different hyperelastic models under certain boundary conditions. Later, with the nonlinear field theory [15], Zhao and Suo [16] modeled the electromechanical behaviors of DEs and concluded that the onset of the EMI can be predicted by the singularities of Hessian matrix from mathematical perspective. Such an approach has paved the way for the systematic studies on the prediction and the suppression of EMI. Adopting the Hessian matrix criterion, efforts have been made to investigate the effects of the material properties on the EMI including the material extension limit [17], the nonlinear dielectric permittivity [18], and the constitutive behaviors of DEs [19, 20]. In addition, some extrinsic approaches have also been explored to enhance the stabilities of the DE actuators for reversible large actuation. For example, Li et al. [21], Zhu [22], Zhou et al. [23, 24] and Su [25] studied the EMI of DE-based actuators with different configurations and they have come to an agreement that the pre-stretch under both dead loads and fixed boundary constraints can help to suppress or even eliminate the EMI as the result of stiffening effects. It is also found that the stability of DE actuators can be improved by the charge-driven actuation [25].
However, when boundary constraints are applied to DEs to suppress EMI, some other instabilities might occur. For example, as reported in Ref. [10], buckling, wrinkling, and crumpling sequentially occur with the increase of the prescribed pre-stretch on the circular DE actuators, which are all featured by the inhomogeneous deformation caused by compression stress. It is worth noting that these instability modes are not always harmful, but sometimes can be used to achieve enhanced deformations [26-28], as long as they are well predicted and harnessed. For the purpose of post-bifurcation prediction and modeling, the incremental approach has been proposed by Dorfmann and Ogden [29] as the Hessian matrix criterion is only applicable for predicting the in-plane instability. The incremental displacement field is commonly formulated as the wave functions, and the out-of-plane bifurcation occurs only when there exist non-trivial solutions of the incremental field. This approach has been successfully applied to predict the post-bifurcation patterns of DE actuators with both plate and tube configurations [30-34]. Due to the complex formulation, it is very difficult, if not impossible, to get analytical solutions for post-bifurcation of DE actuators with other configurations. As the alternative, finite element method provides a feasible tool for the inhomogeneous instability analysis. The linearization and numerical approximation formulations were first derived by Vu et al. [35] for the FE implementation of electroelasticity in simulating the electromechanical finite deformation of DEs. The accuracy and convergence of the Newton-Raphson scheme were also verified in Vu’s work by solving the approximated nonlinear governing equations. Following the similar discretization scheme, Zhou et al. [11] proposed a FE framework incorporating the nonlinear field theory and a hyperelastic constitutive model to study both the homogenous and nonhomogeneous instabilities of DEs. In this FE model, the inhomogeneous instabilities were triggered by introducing imperfections to the original model geometries, while were simulated with the same formulation framework for homogenous instability analysis. Such finite element discretization procedures were also adopted in recent studies of instabilities beyond the simple electrostatic problem, including the instabilities of anisotropic DE actuators by Sharma and Joglekar [36], the dynamic instabilities by Park et al. [37], and even the buckling of swelling gels by Chester et al. [38] with the analogy of chemical free energy to electric free energy.
To better predict and harness the instabilities of DE actuators, accurate constitutive modeling on the electromechanical coupling behavior of DEs is essential. Most DEs are commonly characterized with viscoelasticity, which originates from the diffusion and reptation of polymer chains. In the literature, the viscoelastic constitutive models with constant material viscosity have been incorporated in a few studies for the instability analysis of DE actuators [39-41]. However, as identified experimentally [42, 43], the material viscosity is deformation dependent and thus nonlinear. Based on the theories of polymer dynamics, the tube model [44, 45] has been well recognized as an efficient approach to capture the deformation-dependent viscosity by considering the entanglement effect as the topological constraints of the polymer chains. In this way, the macro deformation of elastomer is connected to the polymer microstructures by assuming that the polymer chains are confined in a tube-like region by the nearby chains. It is natural to believe that the tube diameter will change when DE undergoes large deformation, which will affect the reptation and diffusion ability of the polymer chain confined in the tube and thus leads to the variation of the material viscosity. As DE actuators usually undergo large deformation, the deformation-dependency of the material viscosity becomes more manifest. It is thus essential to consider such nonlinear material viscosity in characterizing the electromechanical behavior of DEs. By modifying the tube model, Li et al. [46] has developed a micro-mechanically based viscoelasticity model, which has been applied to successfully capture the nonlinear material viscosity of both vulcanized and un-vulcanized rubbers through molecular dynamics (MD) simulations [47, 48]. More recently, Zhou et al. [49] incorporated the nonlinear viscosity mechanism into the continuum modeling framework [50] and proposed a micro-macro constitutive model for elastomers and validated the improved accuracy in characterizing the constitutive behaviors of elastomers. This constitutive modeling framework was then implemented into the FE analysis by Feng et al. [51] to study the mechanical response of different elastomeric materials.

To the authors’ best knowledge, the deformation-dependent material viscosity has not yet been considered in the instability analysis of DE structures. It is therefore the objective of the present study to develop a FE framework that implements the nonlinear field theory [15] and the novel micro-macro constitutive model [49] to simulate some common
instability modes of DE actuators. The effects of the nonlinear material viscosity on the instabilities of DE structures are studied for the first time with the proposed FE framework. The robustness and adaptiveness of this FE framework are validated with experimental observations and analytical studies from the existing literatures. Furthermore, this FE framework is also expected to function as a universal platform to conduct numerical simulations on the electromechanical responses of DEs with complex configurations and under general loading conditions, which will help to facilitate novel design and applications of DE-based devices. This chapter is organized as follows: first, the continuum mechanics framework for the nonlinear field theory and the finite-deformation viscoelasticity with the consideration of nonlinear material viscosity is briefly reviewed in Section 4.2; then the finite element discretization procedures are formulated in Section 4.3; in section 4.4, some common instability modes, including EMI, buckling, wrinkling, and crumpling are simulated for VHB 4910 actuators with different configurations using the FE framework; lastly, the conclusions drawn from the current work are summarized in Section 4.5.

4.2 Continuum mechanics framework

In this section, we briefly review the continuum mechanics framework for the constitutive models. In this framework, the nonlinear field theory [15] is adopted to describe the electromechanical coupling of DEs, while the nonlinear material viscosity is incorporated into the finite deformation viscoelasticity theory through the modified tube model originating from the theory of polymer dynamics [49].

4.2.1 Electromechanical field definition with nonlinear field theory

We denote the reference (undeformed) and the current (deformed at time \( t \)) configurations of the DE medium as \( \Omega_0 \) and \( \Omega \), respectively. When subjected to an external load, an arbitrary material particle in DE located by a position vector \( \vec{X} \) in the reference state moves to the current position described by the coordinate \( \vec{x}(\vec{X},t) \) in the current state. Such deformation is described by the deformation gradient tensor \( \mathbf{F} \), defined as,
The nominal electric field $\vec{E}_R$ is defined as the negative gradient of the electric potential $\phi(\vec{X},t)$, i.e.,

$$\vec{E}_R = -\frac{\partial \phi(\vec{X},t)}{\partial \vec{X}} \tag{4.2}$$

The first Piola-Kirchhoff stress tensor $\mathbf{P}$ is defined from the virtual work principle, which is always satisfied for an arbitrary vector test function $\chi$

$$\int_{\Omega_0} \mathbf{P} : \frac{\partial \vec{X}}{\partial \chi} dV = \int_{\Omega_0} \vec{B} \cdot \vec{X} dV + \int_{\partial \Omega_0} \vec{t} \cdot \vec{X} dA \tag{4.3}$$

where $\vec{B}$ is the body force and $\vec{t}$ is the surface traction on the surface $\partial \Omega_0$ of the DE structure. Applying the divergence theorem to Eq. (3), the strong form of the equilibrium equations and the mechanical boundary conditions are derived as,

$$\nabla \mathbf{P} + \vec{B} = \vec{0} \tag{4.4a}$$

$$\mathbf{P} \vec{n} = \vec{t} \text{ on surface } \partial \Omega_0 \tag{4.4b}$$

with $\vec{n}$ being the direction vector normal to the boundary surface $\partial \Omega_0$. Similarly, the nominal electric displacement vector $\vec{D}_R$ also satisfies the virtual work principle, i.e.,

$$-\int_{\Omega_0} \vec{D}_R \frac{\partial \zeta}{\partial \vec{X}} dV = \int_{\Omega_0} q \zeta dV + \int_{\partial \Omega_0} \omega \zeta dA \tag{4.5}$$

for an arbitrary scalar test function $\zeta$. Here $q$ is the body charge density and $\omega$ is the surface charge density applied on the surface $\partial \Omega_0$. Similarly, the strong form of the Gauss's law and the electrical boundary conditions are also obtained as,
\[
\text{div}\vec{D}_R = q 
\quad (4.6a)
\]
\[
\vec{D}_R \cdot \vec{n} = \omega \text{ on surface } \partial \Omega_0 
\quad (4.6b)
\]

From the continuum mechanics perspective, the constitutive equation of DEs is specified by the Helmholtz free energy density function \( W \). The variation of the free energy density function in response to the small change of the mechanical and electrical fields can be written as,

\[
\partial W = \mathbf{P} : \partial \mathbf{F} - \vec{D}_R \cdot \partial \vec{E}_R 
\quad (4.7)
\]

which gives,

\[
\mathbf{P} = \frac{\partial W}{\partial \mathbf{F}} 
\quad (4.8a)
\]
\[
\vec{D}_R = -\frac{\partial W}{\partial \vec{E}_R} 
\quad (4.8b)
\]

For any given free energy density function \( W \), the constitutive relations of DEs can thus be specified according to Eqs. (8).

To capture the electromechanical coupling, the free energy density function \( W \) is decomposed into two terms according to the nonlinear field theory developed by Suo et al. [15], i.e.,

\[
W = W^{\text{mec}}(\mathbf{C}) + W^{\text{ele}}(\vec{E}_R, \mathbf{C}) 
\quad (4.9)
\]

where \( W^{\text{mec}} \) is the free energy density of DE material in the absence of the electric field, which is formulated in terms of the right Cauchy-Green deformation tensor \( \mathbf{C} = \mathbf{F}^T \mathbf{F} \) to ensure the invariance of the free energy density with respect to the rigid body rotation. While \( W^{\text{ele}} \) is the electric free energy density caused by the polarization, defined as
where $\varepsilon$ is the dielectric permittivity of the DE medium, and $J$ is the determinant of the deformation gradient, i.e., $J=\det F$.

### 4.2.2 Finite deformation viscoelasticity theory incorporating nonlinear material viscosity

In this section, we will briefly outline the micro-macro constitutive model proposed by Zhou et al. [49] to describe the viscoelastic behavior of DEs with the consideration of the deformation-dependent material viscosity. As commonly adopted in the literature [49, 50, 52, 53], the viscoelasticity of elastomers is described by the parallel rheological framework (PRF) as illustrated in Fig. 1. Such a concept stems from the idealization of polymer chains with a strong and flexible ground network formed by cross-linked polymer chains and some subnetworks formed by diffusive polymer chains. When DE is subjected to a deformation gradient $F$, the deformation is sustained by both the ground network and the subnetworks, resulting in $F = F_A = F_{B_n} \cdot F_A$ is the pure elastic deformation for the equilibrium ground network $A$, while $F_{B_n}$ is the deformation for each subnetwork which relaxes with time and dissipates energy. Borrowing the concept from the finite deformation plasticity theory [54], the deformation for each subnetwork is commonly multiplicatively decomposed as $F_{B_n} = F_{B_n}^e F_{B_n}^i$ [52]. For each subnetwork, $F_{B_n}^e$ is the elastic deformation gradient with the analogy to the spring deformation and $F_{B_n}^i$ is the inelastic part for the deformation of the dashpot. Accordingly, in each non-equilibrium subnetwork, the elastic and inelastic right Cauchy-Green deformation tensors are thus formulated as $C_{B_n}^e = (F_{B_n}^e)^T F_{B_n}^e$ and $C_{B_n}^i = (F_{B_n}^i)^T F_{B_n}^i$, respectively.
Figure 4.1 Illustration of parallel rheological framework for elastomers.

According to this rheological model, the mechanical Helmholtz free energy density of DE in Eq. (9) is composed of the energy stored in all the springs of the whole network, which can then be expressed as,

$$ W_{mec} \left( C_A, C_{B_1}^e, C_{B_2}^e, \ldots, C_{B_n}^e \right) = W^{EQ} (C) + \sum_{n=1}^{N} W_{B_n}^{NEQ} \left( C_{B_n}^e \right) \quad (4.11) $$

Following Eq. (8a), the first Piola-Kirchhoff stress $\mathbf{P}$ can be formulated by taking the derivative of the total free energy density $W$ with respect to the deformation gradient $\mathbf{F}$ directly. However, the free energy density is commonly expressed in terms of the right Cauchy-Green tensor $\mathbf{C}$ as shown in Eq. (4.11), it will be more convenient to use the second Piola-Kirchhoff stress $\mathbf{S}$ as the alternative stress measure. Following the relation between the first and the second Piola-Kirchhoff stresses, i.e.,

$$ \mathbf{S} = \mathbf{F}^{-1} \mathbf{P} \quad (4.12) $$

the second Piola-Kirchhoff stress is thus derived with the manipulation of Eq. (4.7) as,

$$ \mathbf{S} = 2 \frac{\partial W^{EQ} (\mathbf{C})}{\partial \mathbf{C}} + 2 \sum_{n=1}^{N} \frac{\partial W_{B_n}^{NEQ} \left( C_{B_n}^e \right)}{\partial \mathbf{C}} + 2 \frac{\partial W^{ele} \left( \mathbf{C}, \mathbf{E}_R \right)}{\partial \mathbf{C}} \quad (4.13) $$
Furthermore, the Cauchy stress $\sigma$ can also be obtained following,

$$\sigma = J^{-1} F S F^T$$  \hspace{1cm} (4.14)$$

As the non-equilibrium stress is dependent on the viscous deformation, it is necessary to quantify the inelastic deformation gradient $F_{Bi}^i$ in each subnetwork that evolves following the dissipation inequality [49, 50], i.e.,

$$\left( S - 2 \frac{\partial W}{\partial C} \right) : \frac{1}{2} \dot{C} - \sum_{n=1}^{N} \left( \frac{\partial W^{NEQ}}{\partial C_{Bi}^e} : \frac{\partial C_{Bi}^e}{\partial F_{Bi}^i} \right) \geq 0$$  \hspace{1cm} (4.15)$$

From Eq. (4.13), the dissipation inequality (4.15) can be further rewritten as,

$$\tau_{Bi} \left( B_{Bi}^e \right)^{-1} \left[ F_{Bi}^e L_{Bi}^i \left( F_{Bi}^e \right)^T \right] \geq 0$$  \hspace{1cm} (4.16)$$

where $\tau_{Bi} = 2 F_{Bi}^e \frac{\partial W}{\partial C_{Bi}^e} \left( F_{Bi}^e \right)^T$ is the Kirchhoff stress tensor, $B_{Bi}^e = F_{Bi}^e \left( F_{Bi}^e \right)^T$ is the left Cauchy-Green deformation tensor, and $L_{Bi}^i$ is defined as the velocity gradient tensor with the expression of $L_{Bi}^i = \dot{F}_{Bi}^i \left( F_{Bi}^e \right)^{-1}$. The inequality (4.16) can always be satisfied following the thermodynamics evolution law with the assumption of the material isotropy [49], i.e.,

$$\left[ F_{Bi}^e L_{Bi}^i \left( F_{Bi}^e \right)^T \right] \left( B_{Bi}^e \right)^{-1} = \gamma^{-1} : \tau_{Bi}$$  \hspace{1cm} (4.17)$$

The 4th-order tensor $\gamma^{-1}$ can be expressed as,

$$\gamma^{-1} = \frac{1}{2 \eta_{Bi}} \left( I^4 - \frac{1}{3} I \otimes I \right)$$  \hspace{1cm} (4.18)$$

where $\eta_{Bi}$ is the viscosity for the individual subnetwork $B_i$ and $I^4$ is the 4th-order symmetric identity tensor.
For some simple cases where the deformation gradient $F$ is symmetric, the evolution law of Eq. (4.17) can be easily solved. However, when it comes to the non-homogenous deformation of elastomers, Eq. (4.17) becomes mathematically indeterminate. Following Hong [50] and Boyce et al. [54], it is suggested that the spin of the inelastic deformation gradient can be discarded for polymers without losing the generality. In this case, Eq. (4.17) can be transformed by discarding the skew symmetric part of $L_{B_n}^i$ [51], i.e.,

$$L_{B_n}^i = \left( F_{B_n}^e \right)^{-1} \left( \gamma^{-1} : \tau_{B_n} \right) B_{B_n}^e \left( F_{B_n}^e \right)^{-T} F_{B_n}^i$$

(4.19)

where $M$ is the mobility tensor with respect to the inelastic deformation gradient $F_{B_n}^i$. With such accommodation, the evolution of the internal variables can be numerically integrated and uniquely determined.

Eqs. (4.18) and (4.19) formulate the commonly adopted finite deformation viscoelasticity theory for DEs. In most existing literatures, the material viscosity is treated as a constant in simulating the viscoelastic behavior of elastomers. However, as observed in the experiments [42, 43], the material viscosity is deformation-dependent, which becomes manifest especially when DE undergoes large deformation. According to the micro-macro constitutive model developed by Zhou et al. [49], the viscosity of each subnetwork is affected by the macroscopic deformation of elastomers based on the theory of polymer dynamics [45], i.e.,

$$\eta_{B_n} = \frac{\eta_n}{\alpha \left( F \right)^2}$$

(4.20)

where $\eta_n$ is the initial viscosity of each subnetwork before the deformation. The viscosity in the deformed state is linked to the deformation-related function $\alpha \left( F \right)$ with the expression as,
\[ \alpha(F) = \frac{\langle \overline{R}_{ee}^2 \rangle}{\langle \overline{R}_{ee}^2 \rangle_0 \int \frac{F\ddot{u}_0}{4\pi} d^2\ddot{u}_0} \]  \hspace{1cm} (4.21) 

where \( \ddot{u}_0 \) is the initial unit tangent vector of the primitive chain, and \( \langle \overline{R}_{ee}^2 \rangle \) is the mean square of the end-to-end distance of the primitive chain, defined as,

\[ \langle \overline{R}_{ee}^2 \rangle = \int |F\overline{R}|^2 f(\overline{R}) d^3\overline{R} \]  \hspace{1cm} (4.22) 

The Gaussian distribution function \( f(\overline{R}) \) is adopted for the distribution of the primitive chain vector \( \overline{R} \) in a unit sphere, and the subscript 0 of \( \langle \overline{R}_{ee}^2 \rangle_0 \) represents the quantity before the deformation. The detailed derivation and explanation for the deformation-dependent viscosity can be referred to Zhou et al. [49].

### 4.3 Finite element implementation

In this section, the continuum mechanics framework based on the nonlinear field theory [15] and the micro-macro constitutive model [49] is discretized following the standard FE discretization procedure. The discretized stress tensors and the tangent matrix for the constitutive model considering the electromechanical coupling, the material incompressibility and the nonlinear viscosity are formulated accordingly. Meanwhile, the F-bar method introduced by De Souza Neta et al. [55, 56] is adopted in order to avoid the volume locking in the FE simulations when the incompressibility is enforced.

#### 4.3.1 Finite element discretization

The standard finite element discretization is applied for both the displacement field and the electric potential in the form of shape functions for each element as,

\[ \ddot{u}(\overline{X}, t) = \sum_{d=1}^{M} N^d(\overline{X}) \dddot{u}^d(t) \]  \hspace{1cm} (4.23a)
where \( N^A(\vec{x}) \), \( \vec{u}^A(t) \), and \( \phi^A(t) \) are the shape function, displacement vector and electric potential at the \( A^{th} \) node of the element, respectively. Since the displacement vector and the electric potential are typically in the Hilbert space, we can substitute Eqs. (4.23) into Eqs. (4.3) and (4.5), and apply the Bubnov-Galerkin approximation \([35, 37]\) to convert these equations of the virtual work principle into the weak form, i.e.,

\[
\int_{\Omega} P \frac{\partial N^A}{\partial \vec{x}} dV = \int_{\Omega} \vec{B} N^A dV + \int_{\partial\Omega} \vec{t} N^A dA
\]  

(4.24a)

\[
-\int_{\Omega} \vec{D}_r \frac{\partial N^A}{\partial \vec{x}} dV = \int_{\Omega} q N^A dV + \int_{\partial\Omega} \omega N^A dA
\]  

(4.24b)

In order to numerically solve these equations, the Newton-Raphson iteration procedure is then adopted. The incremental format formulation for Eqs. (4.24) can be written as \([35, 37, 40]\),

\[
\begin{bmatrix}
K_{\vec{u}\vec{u}} & K_{\vec{u}\phi} \\
K_{\phi\vec{u}} & K_{\phi\phi}
\end{bmatrix}
\begin{bmatrix}
\Delta \vec{u}_B \\
\Delta \phi_B
\end{bmatrix} =
\begin{bmatrix}
\vec{R}^u_A \\
\vec{R}^\phi_A
\end{bmatrix}
\]  

(4.25)

where the residual vectors are calculated by,

\[
\vec{R}^u_A = -\int_{\Omega} P \frac{\partial N^A}{\partial \vec{x}} dV + \int_{\Omega} \vec{B} N^A dV + \int_{\partial\Omega} \vec{t} N^A dA
\]  

(4.26a)

\[
\vec{R}^\phi_A = \int_{\Omega} \vec{D}_r \frac{\partial N^A}{\partial \vec{x}} dV + \int_{\Omega} q N^A dV + \int_{\partial\Omega} \omega N^A dA
\]  

(4.26b)

The residual vectors can be numerically integrated as long as the stress tensor \( P \) and the nominal electric displacement \( \vec{D}_r \) are formulated, and the external loads and the boundary conditions are specified. The components of the corresponding tangent matrix on the left-hand side of Eq. (4.25) are defined as,
Adopting the Einstein’s notation, these components can be further expanded with respect to the current configuration as,

\[
K_{AB}^{μ_μ} = -\frac{∂R_μ^μ}{∂u^B}, \quad K_{AB}^{μφ} = -\frac{∂R_μ^φ}{∂φ^B}
\]

\[
(4.27)
\]

\[
K_{AB}^{μφ} = -\frac{∂R_μ^φ}{∂u^B}, \quad K_{AB}^{φφ} = -\frac{∂R_φ^φ}{∂φ^B}
\]

\[
(4.27)
\]

From Eqs. (28), the tangent matrix can be determined numerically using the Gaussian integration scheme if the derivatives \( \frac{∂P}{∂F}, \frac{∂P}{∂E_R}, \frac{∂D_R}{∂E_R}, \) and \( \frac{∂D_R}{∂E_R} \) are formulated.

4.3.2 Implementation of the constitutive models for incompressible materials

In this section, we will present the detailed formulations of the stress tensors and the tangent matrix introduced in Section. 4.3.1 for the implementation of the material constitutive models into the FE framework. With the consideration of the incompressibility of elastomers, we first adopt the multiplicative decomposition scheme to decompose the deformation gradient into the volumetric deformation part and the isochoric deformation part as [55],
\[ F = F^{iso} F^{vol} = \left( J^{-1/3} F \right) \left( J^{1/3} I \right) \]  

(4.29)

where superscripts ‘vol’ and “iso” are used to represent the volumetric part and the isochoric part, respectively. It is obvious that the isochoric part has a constant determinant, i.e., \( \det(J^{-1/3} F) = 1 \). According to this decomposition scheme, the isochoric right Cauchy-Green deformation tensor is formulated as \( C^{iso} = J^{-2/3} C \). Similarly, the elastic isochoric right Cauchy-Green deformation tensor of the subnetwork \( B_n \) is determined as \( C^{iso,e} = (J^{e \cdot B_n})^{-2/3} C^{e \cdot B_n} \). According to this decomposition scheme, the Helmholtz free energy density defined by Eq. (4.9) is rewritten as [51],

\[
W = \tilde{W}^{EQ} \left( C^{iso} \right) + \sum_{n=1}^{N} \tilde{W}^{NEQ}_{B_n} \left( C^{iso,e}_{B_n} \right) + U^{vol} (J) + W^{ele} \left( \tilde{E}_R, C \right) \tag{4.30}
\]

The electric free energy density function \( W^{ele} \left( \tilde{E}_R, C \right) \) takes the form of Eq. (4.10). In the PRF, the selection of the strain energy functions for the isochoric parts \( \tilde{W}^{EQ} \left( C^{iso} \right) \) and \( \tilde{W}^{NEQ}_{B_n} \left( C^{iso,e}_{B_n} \right) \) is quite flexible depending on the material behaviors. While the free energy density function for the volumetric part is expressed in terms of the bulk modulus [57], as,

\[
U^{vol} (J) = \frac{1}{2} \kappa (J - 1)^2 \tag{4.31}
\]

The material incompressibility can be enforced in the FE simulations by using a relatively large bulk modulus \( \kappa \), commonly set within the range of \( 10^4 \text{--} 10^6 \) times of the shear modulus by recommendation [37]. From Eq. (4.13), the second Piola-Kirchhoff stress tensor can thus be further decomposed as,
where $S_{\text{EQ}}^{\text{iso}} = 2 \frac{\partial \bar{W}_{\text{EQ}}}{\partial C}$ is the equilibrium isochoric stress, $S_{\text{nB}}^{\text{iso}} = 2 \frac{\partial \bar{W}_{\text{Bn}}^{\text{NEQ}}}{\partial C}$ is the non-equilibrium isochoric stress for each subnetwork, $S_{\text{vol}} = 2 \frac{\partial U_{\text{vol}}}{\partial C}$ is the volumetric stress caused by the volume change, and $S_{\text{ele}} = 2 \frac{\partial W_{\text{ele}}}{\partial C}$ is the electrostatic stress due to the voltage-induced deformation. These stresses can be further expanded as,

\[ S_{\text{EQ}}^{\text{iso}} = J^{-2/3} \mathbb{P} : \bar{S}_{\text{EQ}} \]  
\[ S_{\text{Bn}}^{\text{iso}} = (J_{\text{Bn}}^e)^{-2/3} \mathbb{P}_{\text{Bn}}^{\text{NEQ}} : \bar{S}_{\text{Bn}}^{\text{NEQ}} \]  
\[ S_{\text{vol}} = \kappa J (J - 1) C^{-1} \]  
\[ S_{\text{ele}} = \varepsilon J \left[ C^{-1} \bar{E}_R \otimes C^{-1} \bar{E}_R - \frac{1}{2} \left( \bar{E}_R \cdot C^{-1} \bar{E}_R \right) C^{-1} \right] \]

where $J_{\text{Bn}}^e$ is the determinant of $F_{\text{Bn}}^e$, $\bar{S}_{\text{EQ}} = 2 \frac{\partial \bar{W}_{\text{EQ}}}{\partial C^{\text{iso}}} \text{ and } \bar{S}_{\text{Bn}}^{\text{NEQ}} = 2 \frac{\partial \bar{W}_{\text{Bn}}^{\text{NEQ}}}{\partial C_{\text{Bn}}^{\text{iso,e}}}$. The fourth order tensors $\mathbb{P}$, $\mathbb{P}^{\text{NEQ}}$, and $\mathbb{P}_{\text{Bn}}$ are projection tensors from $C^{\text{iso}}$ to $C$, from $C_{\text{Bn}}^{\text{iso,e}}$ to $C_{\text{Bn}}^e$, and from $C_{\text{Bn}}^e$ to $C$, respectively, which are formulated as [51],

\[ \mathbb{P}_{abcd} = J^{2/3} \frac{\partial C^{\text{iso}}_{cd}}{\partial C_{ab}} = \delta_{ac} \delta_{bd} - \frac{1}{3} C^{-1}_{ab} C_{cd} \]  
\[ \left( \mathbb{P}^{\text{NEQ}}_{\text{Bn}} \right)_{abcd} = (J_{\text{Bn}}^e)^{2/3} \frac{\partial C^{\text{iso,e}}_{cd}}{\partial C_{ab}} = \delta_{ac} \delta_{bd} - \frac{1}{3} \left( C_{\text{Bn}}^e \right)^{-1}_{ab} \left( C_{\text{Bn}}^e \right)^{-1}_{cd} \]  
\[ \left( \mathbb{P}_{\text{Bn}} \right)_{abcd} = \frac{\partial C_{cd}^e}{\partial C_{ab}} = \frac{1}{2} \left[ \left( F_{\text{Bn}}^i \right)^{-1}_{ac} \left( F_{\text{Bn}}^i \right)^{-1}_{bd} + \left( F_{\text{Bn}}^i \right)^{-1}_{bc} \left( F_{\text{Bn}}^i \right)^{-1}_{ad} \right] \]
After obtaining the stress tensors, the tangent matrix also needs to be updated based on Eqs. (4.28) and (4.30). In Eq. (4.28a), we need to further expand the term \( \frac{\partial P_{ia}}{\partial F_{jn}} \) for integration, which are obtained by applying the chain rule to Eq. (4.12), as,

\[
\frac{\partial P_{ia}}{\partial F_{jn}} = \delta_{ij} s_{ma} + 2F_{ix} F_{jy} \frac{\partial S_{ma}}{\partial C_{yn}}
\] 

(4.35)

In this way, the components of the tangent matrix are directly linked to the 4th-order tensor \( \frac{\partial S}{\partial C} = 4 \frac{\partial^2 W}{\partial C \otimes \partial C} \) and the free energy density functions \( W \). Based on Eq. (4.32), this 4th-order tensor can be expanded as,

\[
2 \frac{\partial S}{\partial C} = 2 \left( \frac{\partial S_{\text{iso}}^{\text{EQ}}}{\partial C} + \sum_{n=1}^{N} \frac{\partial S_{B_n}}{\partial C} + \frac{\partial S_{\text{vol}}}{\partial C} + \frac{\partial S_{\text{ele}}}{\partial C} \right)
\] 

(4.36)

The detailed expressions for the mechanical parts in Eq. (4.36) have been derived in our previous study [51] as,

\[
2 \frac{\partial S_{\text{iso}}^{\text{EQ}}}{\partial C} = \left( \hat{\mathbf{C}} - \frac{1}{3} A_1 \right) : \mathbb{P}^T \frac{2}{3} \left( A_2 + A_3 + \mathbf{C}^{-1} \otimes S_{\text{iso}}^{\text{EQ}} \right)
\] 

(4.37a)

\[
2 \frac{\partial S_{B_n}}{\partial C} = \frac{\hat{\mathbb{P}}_{B_n}}{B_n} : \left( \hat{C}_{B_n} - A_{B_n}^1 \right) \left( \mathbb{P}_{B_n}^{\text{NEQ}} \right)^T \frac{2}{3} \left[ \left( A_{B_n}^2 + A_{B_n}^3 \right) \right] \frac{\hat{\mathbb{P}}_{B_n}}{B_n} \frac{2}{3} \mathbf{C}_{B_n}^{-1} \otimes S_{B_n}^{\text{iso}} \] 

(4.37b)

\[
2 \frac{\partial S_{\text{vol}}}{\partial C} = 2 \kappa \left[ \frac{2J^2 - J}{2} \mathbf{C}^{-1} \otimes \mathbf{C}^{-1} - J (J - 1) \mathbf{C}^{-1} \otimes \mathbf{C}^{-1} \right]
\] 

(4.37c)

where

\[
\hat{\mathbf{C}} = 4J^{-4/3} \frac{\partial^2 \tilde{W}^{\text{EQ}}}{\partial \mathbf{C}^{\text{iso}} \otimes \partial \mathbf{C}^{\text{iso}}}
\] 

(4.38a)

\[
A_{\lambda} = \mathbf{C}^{-1} \otimes \mathbf{C} : \hat{\mathbf{C}}
\] 

(4.38b)
\[ \mathbf{A}_{2} = -J^{-2/3} \left( \mathbf{C} : \mathbf{S}^{EQ} \right) \left( \mathbf{C}^{-1} \odot \mathbf{C}^{-1} \right) \] (4.38c)

\[ \mathbf{A}_{3} = \mathbf{C}^{-1} \otimes J^{-2/3} \mathbf{S}^{EQ} \] (4.38d)

and,

\[ \mathbf{\hat{C}}_{B_{n}}^{e} = 4 \left( J_{B_{n}}^{e} \right)^{-4/3} \frac{\partial^{2} \mathbf{W}_{B_{n}}^{NEQ}}{\partial \mathbf{C}_{B_{n}}^{iso,e} \otimes \partial \mathbf{C}_{B_{n}}^{iso,e}} \] (4.39a)

\[ \mathbf{\hat{A}}_{B_{n}}^{1} = \mathbf{C}_{B_{n}}^{e-1} \otimes \mathbf{C}_{B_{n}}^{e} : \mathbf{\hat{C}}_{B_{n}}^{e} \] (4.39b)

\[ \mathbf{\hat{A}}_{B_{n}}^{2} = -\left( J_{B_{n}}^{e} \right)^{-2/3} \left( \mathbf{C}_{B_{n}}^{e} : \mathbf{S}_{B_{n}}^{NEQ} \right) \left[ \left( \mathbf{C}_{B_{n}}^{e} \right)^{-1} \odot \left( \mathbf{C}_{B_{n}}^{e} \right)^{-1} \right] \] (4.39c)

\[ \mathbf{\hat{A}}_{B_{n}}^{3} = \left( J_{B_{n}}^{e} \right)^{-2/3} \mathbf{C}_{B_{n}}^{e-1} \otimes \mathbf{S}_{B_{n}}^{NEQ} \] (4.39d)

The operator ‘\( \odot \)’ in Eq. (4.39c) has the definition as: \( A_{ab} \odot B_{cd} = \frac{1}{2}(A_{ac}B_{bd} + A_{ad}B_{bc}) \).

In addition, the 4th-order tensor in Eq. (4.36) with respect to the electrostatic stress can be expressed as,

\[ 2 \frac{\partial S_{ea}^{ele}}{\partial C_{yn}} = S_{xe}^{ele} C_{yn}^{-1} + 2\varepsilon J \tilde{E}_{R,m} \tilde{E}_{R,L} \left[ -\frac{1}{2} C_{sm}^{-1} \left( C_{sa}^{-1} C_{nl}^{-1} + C_{na}^{-1} C_{ly}^{-1} \right) - \frac{1}{2} C_{al}^{-1} \left( C_{sy}^{-1} C_{mu}^{-1} + C_{xm}^{-1} C_{my}^{-1} \right) \right] \]

\[ -\varepsilon J \tilde{E}_{R,m} \tilde{E}_{R,L} \left[ -\frac{1}{2} C_{ml}^{-1} \left( C_{sy}^{-1} C_{am}^{-1} + C_{xn}^{-1} C_{ay}^{-1} \right) - \frac{1}{2} C_{sa}^{-1} \left( C_{my}^{-1} C_{ln}^{-1} + C_{mn}^{-1} C_{ly}^{-1} \right) \right] \] (4.40)

Similarly, the remaining parts in the tangent matrix (4.27) as formulated by Eqs. (4.28b), (4.28c) and (4.28d) are derived following the chain rule, as,

\[ \frac{\partial P_{a}}{\partial E_{R,m}} = \varepsilon J \left( F_{m}^{-1} C_{al}^{-1} E_{R,L} + F_{m}^{-1} F_{Li}^{-1} E_{m,L} - F_{a}^{-1} C_{ml}^{-1} E_{R,L} \right) \] (4.41a)

\[ \frac{\partial D_{R,m}}{\partial E_{Ib}} = \varepsilon J \left( F_{hi}^{-1} C_{mn}^{-1} E_{R,n} - F_{mi}^{-1} C_{bn}^{-1} E_{R,n} - C_{mh}^{-1} F_{Li}^{-1} E_{L} \right) \] (4.41b)
where the spatial electric field vector is expressed as $\bar{E} = \left(F^{T}\right)^{-1} \bar{E}_r$. With the proposed FE framework, different electro-viscoelasticity constitutive models can be easily implemented since the formulations of the tangent matrix relies on the direct derivatives $\frac{\partial^2 \bar{W}^{EQ}}{\partial C_{iso}^{i} \otimes \partial C_{iso}^{i}}$ and $\frac{\partial^2 \bar{W}^{NEQ}}{\partial C_{B_{iso}}^{j} \otimes \partial C_{B_{iso}}^{j}}$.

### 4.3.3 F-bar method for alleviation of volumetric locking

To mitigate the volumetric locking behaviors of the elements when the incompressibility limit is approached, we adopt the F-bar method [55, 56] to accommodate the isochoric-volumetric decomposition scheme. F-bar method gains its popularity in FE analysis for the nearly incompressible materials due to the ease of implementation. It can be simply implemented by substituting the deformation gradient at each integration point with a modified one, which has the same determinant of the deformation gradient at the centroid of the element in the form of,

$$
\bar{F} = \left(\frac{\det F_c}{\det F}\right)^{1/3} F
$$

where $F_c$ is the deformation gradient tensor at the centroid of the element. The volumetric locking is relieved as the average volumetric change throughout the element instead of the point-wise change at each integration point, is adopted for the incompressibility constraint. The expressions of the residuals in Eqs. (4.26) remain the same except that the original deformation gradient tensor $F$ is replaced by $\bar{F}$, while the material tangent in Eq. (4.28a) needs to be modified according to Refs. [38, 56], i.e.,

$$
K_{AB}^{\alpha\beta} = \int_{\Omega} \frac{\partial N^A}{\partial x_b} \frac{\partial N^B}{\partial x^L} d\nu + \int_{\Omega} \frac{\partial N^A}{\partial x_b} \left( \frac{\partial N^B}{\partial x^L} - \frac{\partial N^B}{\partial x^L} \right) d\nu
$$

(4.43)
with \( x^i_L \) being the coordinate of the centroid of the element. The 4th-order tensors in this equation take the forms of,

\[
A_{ijkl} = J^{-1} F_{ba} F_{Le} \frac{\partial P}{\partial F_{jn}}
\]  

(4.44)

and,

\[
Q = \frac{1}{3} \mathbf{A} : (\mathbf{I} \otimes \mathbf{I}) - \frac{2}{3} \mathbf{\sigma} \otimes \mathbf{I}
\]  

(4.45)

With the assumption that the voltage induced deformation is always isochoric, the other three parts in the tangent matrix remain unchanged. After the element-level tangent matrix and the residuals are determined in Eq. (4.25), the global residual vectors and material tangent matrix can be assembled and finally the displacement and potential increments can be solved using the Newton-Raphson iteration scheme in the FE simulations.

To conclude, the numerical scheme to solve the displacement field and the electric potential is outlined as follows. This scheme is coded in a user-element subroutine (UEL) in Abaqus for simulating different instability modes of the DE structures.
4.4 Finite element simulation results and discussions

In this section, the developed finite element (FE) framework is adopted to simulate some common instabilities of VHB 4910 structures, including electromechanical instability (EMI), buckling, wrinkling, and crumpling using the user-element subroutine (UEL) implemented in Abaqus. Different boundary conditions, i.e., pre-stretches with dead loads and fixed boundaries with different pre-stretches, are applied on the DE actuators to investigate different instability modes of DE structures. In order to apply the parallel rheology model in Fig. 4.1, the Helmholtz free energy densities of the springs in
the network denoted by $W_A(C_A)$ and $W_{B_n}(C_{B_n}^e)$ must be first prescribed. Following the work of Zhou et al. [49], the Gent free energy density function is adopted to characterize the behavior of DE for both equilibrium and non-equilibrium states. Following the decomposition scheme in Eq. (4.30), the corresponding isochoric Gent free energy density functions can be written as

$$W^{EQ} = -\frac{G^{EQ}_n J_{lim}}{2} \ln \left( \frac{J_{lim} - I_1^{iso,e} + 3}{J_{lim}} \right)$$ (4.46a)

$$W^{NEQ}_{B_n} = -\frac{G^{NEQ}_n J_{lim}}{2} \ln \left( \frac{J_{lim} - \left(I_1^{iso,e}\right)_{B_n} + 3}{J_{lim}} \right)$$ (4.46b)

where $G^{EQ}$ is the macroscale equilibrium shear modulus of the ground network, while $G^{NEQ}_n$ is the non-equilibrium shear modulus of each subnetwork; $J_{lim}$ is the material parameter for the extensibility that can be expressed as $J_{lim} = 3(N-1)$ with N being the polymerization degree of polymer chains; $I_1^{iso}$ is the first invariant of $C^{iso} = (F^{iso})^T F^{iso}$ and $\left(I_1^{iso,e}\right)_{B_n}$ is the first invariant of $C^{iso,e}_{B_n} = (F^{iso,e}_{B_n})^T F^{iso,e}_{B_n}$ for each non-equilibrium subnetwork. Through data fitting with experimental data, it is found that three non-equilibrium subnetworks are accurate to capture the viscoelastic behavior of VHB 4910 with the material parameters listed in Table. 4.1 [49]. Along with the dielectric permittivity $\varepsilon$ set as $4.1 \times 10^{-11}$ F/m, these parameters are used as the input material properties of the constitutive model of VHB 4910 in the current FE analysis. The simulation results will be compared with experimental or analytical results from the existing literatures to validate the accuracy and the adaptiveness of the FE framework.
Table 4.1 Material parameters for VHB 4910 according to the PRF with three subnetworks [49]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>$G_{EQ}$</th>
<th>$G_{NEQ}^1$</th>
<th>$G_{NEQ}^2$</th>
<th>$G_{NEQ}^3$</th>
</tr>
</thead>
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<td>11.87 KPa</td>
<td>43.15 KPa</td>
<td>19.75 KPa</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Parameter</th>
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<th>$\eta_1$</th>
<th>$\eta_2$</th>
<th>$\eta_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Value</td>
<td>115.8</td>
<td>11.4 MPa s</td>
<td>43.17 KPa s</td>
<td>381.16 KPa s</td>
</tr>
</tbody>
</table>

4.4.1 EMI of VHB 4910 plate under dead loads

Electromechanical instability (EMI), also known as a snap-through instability, is one of the most common instability modes that DE actuators may encounter in the actuating process. The suppression of EMI using pre-stretch under dead loads has been widely explored in the literature to achieve large deformation for DE actuators under applied electric field [21-25]. For this configuration, an external mechanical load $\mathbf{P}_{pre} = \int_{\partial \Omega} \mathbf{t} dA$ ($\mathbf{t}$ is the surface traction) is applied on the boundaries of the DE plate to introduce strain stiffening effect to help suppressing the instability.

For case study, a VHB 4910 plate with dimensions of 20 mm × 20 mm × 2 mm is modeled in Abaqus as shown in Fig. 4.3. The equi-biaxial loads are applied to the plate to reach different pre-stretches $\lambda_{pre}$ in a long duration (one hour). Since the pre-loading rate is rather low, the VHB 4910 plate can be considered in the equilibrium state free of inelastic stress before the voltage-induced actuation. The voltage $\varphi$ is then applied with an incremental step of 0.5 kV/10s at 10s intervals in the plate thickness direction, inducing an equi-biaxial stretch $\lambda$. Each loading step is then discretized into $10^3$ increments. The FE model is the same as the experimental setup in Ref. [58]. The pre-stretches induced by the constant bi-axial stresses are set as 1, 1.6, 2, 2.5, and 3 respectively.
It should be mentioned that the body force, surface charge and body charge are all neglected for the simulations in the present study. To apply the surface tractions on the boundaries of the DE plate in the FE simulation, we need to further formulate the integration \( \int_{\partial \Omega} \vec{t} N^4 dA \) for the residual vector in Eq. (4.26a). Assuming that the surface traction is applied in the direction normal to the local surface \( \zeta = 1 \) as shown in Fig. 4.4, the residual caused by the surface traction can be transformed as [59],

\[
\int_{\partial \Omega} \vec{t} N^4 dA = \int_{\partial \Omega} \vec{f} | N^4 d\vec{A} = \int_{\partial \Omega} \vec{f} | N^4 \left( d\vec{\xi} \times d\vec{\eta} \right) \tag{4.47}
\]

in which the vector increments can be further expressed as,

\[
d\vec{\xi} = \left( i \frac{\partial X}{\partial \xi} + j \frac{\partial Y}{\partial \xi} + k \frac{\partial Z}{\partial \xi} \right) d\xi \tag{4.48a}
\]

\[
d\vec{\eta} = \left( i \frac{\partial X}{\partial \eta} + j \frac{\partial Y}{\partial \eta} + k \frac{\partial Z}{\partial \eta} \right) d\eta \tag{4.48b}
\]

with \( i \), \( j \), and \( k \) being the basis vectors of the global coordinate system. In Eqs. (4.48), the derivatives of the global coordinates with respect to the local coordinates can be
obtained from the coordinate system transformation (Jacobian) matrix with the expression as,

\[
[J] = 
\begin{bmatrix}
\frac{\partial X}{\partial \xi} & \frac{\partial X}{\partial \eta} & \frac{\partial X}{\partial \zeta} \\
\frac{\partial Y}{\partial \xi} & \frac{\partial Y}{\partial \eta} & \frac{\partial Y}{\partial \zeta} \\
\frac{\partial Z}{\partial \xi} & \frac{\partial Z}{\partial \eta} & \frac{\partial Z}{\partial \zeta}
\end{bmatrix} = 
\begin{bmatrix}
\sum_{A=1}^{8} \frac{\partial N_A^X}{\partial \xi} X^A & \sum_{A=1}^{8} \frac{\partial N_A^Y}{\partial \xi} Y^A & \sum_{A=1}^{8} \frac{\partial N_A^Z}{\partial \xi} Z^A \\
\sum_{A=1}^{8} \frac{\partial N_A^X}{\partial \eta} X^A & \sum_{A=1}^{8} \frac{\partial N_A^Y}{\partial \eta} Y^A & \sum_{A=1}^{8} \frac{\partial N_A^Z}{\partial \eta} Z^A \\
\sum_{A=1}^{8} \frac{\partial N_A^X}{\partial \zeta} X^A & \sum_{A=1}^{8} \frac{\partial N_A^Y}{\partial \zeta} Y^A & \sum_{A=1}^{8} \frac{\partial N_A^Z}{\partial \zeta} Z^A
\end{bmatrix} \tag{4.49}
\]

Then the residuals caused by the external loads can be numerically integrated following Eq. (4.47).

Figure 4.4 Illustration of coordinate system transformation

The FE simulated results of the voltage-induced in-plane deformation of the DE plate under different pre-stretches \(\lambda_{pre}\) with the incorporation of the nonlinear material viscosity are plotted in Fig. 4.5, in comparison with the experimental data from Ref. [58]. Featured by the dramatic in-plane expansion in response to the infinitesimal increment in voltage as shown in Fig. 4.5, the EMI phenomenon can be identified when the simulations diverge as marked by cross. Attributed to the stiffening effect of the large pre-stretches (for example when \(\lambda_{pre} > 2\)), the EMI is suppressed, and a finite value of the voltage-induced deformation can be achieved as indicated in the finite element analysis, which is in consistence with the experimental observations. From this figure, it is concluded that the developed FE framework can successfully predict the EMI and reflect the suppression effect of the pre-stretches. The discrepancy of the critical voltages for
EMI between the simulation results and the experiment data may arise from the potential error in measurement or selection of the material property parameters.

Figure 4.5 Voltage-induced in-plane deformation of a VHB 4910 plate with different pre-stretches. The FE simulated results are represented in solid lines and the experimental data are plotted as symbols.

To investigate the effect of the nonlinear material viscosity on the voltage-induced deformation and the EMI of the DE plate, viscoelastic constitutive model with constant material viscosity and hyperelastic model (i.e., the equilibrium state of the viscoelastic DE) are also adopted in the FE framework for comparison purpose. The voltage-induced deformation for the DE plate with EMI and without EMI is plotted in Fig. 4.6 and Fig. 4.7, respectively. It is evident from Fig. 4.6 that the material viscosity delays the EMI as expected. It means that the critical electric voltage for the EMI becomes larger due to the material viscosity. As observed in these two figures, the nonlinear material viscosity, compared with the constant material viscosity, leads to smaller in-plane deformation of the DE plate under the same electric voltage. This is due to the fact that the material viscosity decreases with the increase of the in-plane stretch of the DE plate which was analytically predicted in our previous research [49]. It is worth noting that when there is no pre-stretch (see Fig. 4.6 (c) \( \lambda_{\text{pre}} = 1 \)), the difference between the simulated results with
constant and nonlinear material viscosity can be ignored since the variation of viscosity is negligible when the DE plate is under small actuation.

Figure 4.6 Effect of material viscosity on electromechanical response of a VHB 4910 plate with EMI under different pre-stretches.
Figure 4.7 Effect of material viscosity on electromechanical response of a VHB 4910 plate free from EMI under different pre-stretches.

The effect of the voltage loading rate on the voltage-induced deformation and the onset of the EMI is also investigated through the FE simulation with the consideration of the deformation-dependent viscosity. The loading rates of the applied voltage are set as 0.5 kV/1s at 1s intervals, 0.5 kV/10s at 10s intervals, and 0.5 kV/100s at 100s intervals respectively. The finite voltage-induced deformation of the VHB 4910 plate is plotted in Fig. 4.8 for the cases with EMI, while in Fig. 4.9 for the cases without EMI. It is observed from Fig. 4.8, the critical voltage for the onset of the EMI increases with the
electrical loading rate. On the other hand, for the cases where the VHB 4910 plate is free of the EMI, the material viscosity effect becomes weak especially when the stretch limit of the material is approached. This is due to the fact that the in-plane deformation rate experiences a drastic decrease in response to the applied electric voltage, which allows the material to have sufficient time to relax and approach the equilibrium state.

Figure 4.8 Effect of voltage loading rate on electromechanical response of a VHB 4910 plate with EMI under different pre-stretches.
Figure 4.9 Effect of voltage loading rate on electromechanical response of a VHB 4910 plate free from EMI under different pre-stretches

4.4.2 EMI and buckling of VHB 4910 plate with fixed end constraints

As reported in the literature [23], the fixed boundary constraints are also capable of helping to suppress the EMI. In the present work, we revisit this problem with the consideration of the material viscosity. The dimension of the plate is set as 4 mm × 2 mm × 0.1 mm with the fixed constraint in the x-direction, as shown in Fig. 4.10. Due to the symmetry of this configuration, a quarter-symmetry model is established to reduce the computational cost in the FE simulation with the corresponding symmetric boundary
conditions in the planes of \( x=0 \) and \( y=0 \). The electric voltages \( \varphi \) is applied through the electrodes coated on the upper and lower surfaces of the plate. The voltage is normalized as \( \bar{\varphi} = \frac{\varphi}{H} \sqrt{\frac{\varepsilon}{G^{EQ}}} \) in demonstrating the simulation results for simplicity purpose. For this case study, the normalized voltage is applied at the rates of \( \bar{\varphi}'=1/s, 0.1/s, 0.01/s, \) and \( 0.001/s \), respectively, to show the rate-dependent effect caused by the material viscosity. Meanwhile, the voltage-induced in-plane deformation in the equilibrium state is also numerically simulated by the FE framework and analytically calculated based on the formulation in Ref. [23] for validation.

Reference state

![Reference state](image)

**Figure 4.10** Model setup of a VHB 4910 plate with fixed constraint. Voltages are applied on the upper surface and bottom surface of the plate.

Fig. 4.11 summarizes the FE simulated results for the DE plate subjected to the applied voltage with different loading rates for the cases with constant (denoted as \( \eta_n \)) and nonlinear (denoted as \( \eta_B \)) material viscosity. The accuracy of the proposed FE framework is validated with the excellent agreement between the FE simulation results and the analytical solutions on the DE response in the equilibrium state. As outlined in this figure, the EMI is successfully eliminated with the fixed boundary constraint and a large deformation is thus achieved through the actuation of the applied electric field. The delay effect of the material viscosity on the actuation of the DE plate is also observed,
i.e., the actuation lags with the higher loading rate of the applied electric voltage. It is also observed that the nonlinear material viscosity plays an important role in the actuation response of the DE plate, as demonstrated by the discrepancy for the material with constant and nonlinear viscosity, particularly when the DE plate undergoes relatively large deformation. This is expected as the material viscosity decreases with the deformation. However, similar to the simulations for the DE plate subjected to dead loads, when the voltage-induced deformation continues to increase until approaching the extension limit, the effect of material viscosity on the finite deformation becomes imperceptible as the material can sufficiently relax when the deformation rate is low.

![Figure 4.11 Out-of plane deformation of a VHB 4910 plate with fixed ends under applied voltage with different loading rates of 0.001/s, 0.01/s, 0.1/s and 1/s.](image)

Despite of the successful suppression of the EMI by the boundary constraints, another instability issue arises for this configuration. During the actuation process, the compression stresses will be generated due to the constrained in-plane expansion in $x$-direction with the fixed ends. As the compression stresses accumulate, buckling may occur in the thin plate with low bending stiffness. To trigger the buckling instability, all the elements on the top surface are prescribed by a random geometric imperfection in the thickness direction on the order of $10^{-3}$ of the original plate thickness. Numerical
simulations are performed for the cases with different loading rates of 1/s, 0.1/s, 0.01/s, and 0.001/s, respectively. The buckling mode in the equilibrium state has also been simulated for comparison with the numerical results by Sharma [36].

Figure 4.12 Displacement contours of buckled patterns of a VHB 4910 plate with the voltage loading rate of 1/s before failure in: (a) thickness direction, and (b) expansion direction

The displacement contours in the thickness direction (z-axis) and the expansion direction (y-axis) of the DE plate are plotted in Figs. 4.12 with the applied voltage rate of 1/s as an example, with which the capacity of the FE framework in predicting the post-buckling patterns is validated. It should be advised that Figs. 4.12 are plotted by mirroring the deformation contours of the quarter-symmetry model for better demonstration. The relation between the maximum out-of-plane deflection and the applied voltage at different loading rates is plotted in Fig. 4.13. As detailed in this figure, when the applied voltage is low, the in-plane expansion dominates, and the out-of-plane deflection can be ignored. While with the increase of the applied voltage, the plate starts to buckle out of the plane until it fails at a large deflection. Similar to the EMI, the delay effect of the material viscosity is also noticed by the significant discrepancy among the curves for different loading rates. The critical voltages for the onset of the buckling as well as the final failure of the DE plate are plotted in Fig. 4.14. It is concluded from these figures that it is also crucial to incorporate the material viscosity in characterizing the buckling and the post-buckling of DE structures. However, by contrast, the nonlinearity of the material viscosity plays a minor role in the buckling of the DE plate, which can be explained by the negligible decay of the viscosity with small stretch as evident from the slight expansion in Fig. 4.12 (b), notwithstanding the rather large out-of-plane
displacement. This can be further validated by the small variation of the material viscosity with the applied voltage as plotted in Fig. 4.15, taking normalized loading rate of 1/s for the applied voltage as an example.

Figure 4.13 Effect of voltage loading rate on the maximum out-of-plane deflection of a VHB 4910 plate with fixed ends
Figure 4.14 Variation of critical voltages for onset of buckling and failure with the loading rate of the applied voltage.
4.4.3 Wrinkling and crumpling of VHB 4910 circular membranes under fixed pre-stretch

In addition to the EMI and the buckling, the DE membranes are also vulnerable to some other instability modes such as wrinkling and crumpling when subjected to a fixed pre-stretch. Such instabilities commonly occur when the tension stress in the membrane is lost. The membranes with large radius-thickness ratio can hardly sustain the compression stresses after the loss of tension. Following the terminology convention introduced in Ref. [10], we define the wrinkling as the in-plane wave-like deformation while crumpling as the out-of-plane inhomogeneous deformation with vertices and ridges. In the present study, wrinkling and crumpling instabilities of a VHB 4910 circular membrane are simulated in comparison with the experimental data from Ref. [10] to validate the robustness and accuracy of the proposed FE framework.

Figure 4.15 Variation of material viscosity in each subnetwork of the PRF with applied voltage at normalized loading rate of 1/s.
The simulations follow three steps as shown in Fig. 4.16, which are the same as the experimental procedures in [10]. Firstly, the membrane is stretched from a radius of \( R \) to \( r = \lambda R = 30 \text{ mm} \), in which the initial radius \( R \) is determined according to the prescribed pre-stretch ratio. Then the DE membrane is fixed by a rigid frame and relaxed for one hour so that the inelastic stresses caused by the pre-stretch are fully relaxed. In the end, the voltage is applied with a rate of 20 V/s for the actuation. The initial thickness \( H \) is set as 1 mm with the random imperfection on the order of 0.001 mm to aid in triggering the onset of instability. For the simulation of the wrinkling, the bottom surface of the membrane is fixed as the boundary constraint. While in simulating the crumpling instability, there is no constraint in the thickness direction so that the membrane can bulge out of the plane freely [10].

As reported in the reference [10], wrinkles were observed experimentally in a pre-stretched VHB 4910 membrane with the increase of the applied voltage. With the same pre-stretch of \( \lambda_{\text{pre}} = 5 \) applied in the experiment, random variation of the membrane thickness with the order of \( 10^{-4} \text{ mm} \) is introduced as the geometric imperfection to trigger the wrinkling instability in the FE simulation. It is observed that the wave-like in-plane deformation starts to appear when \( \varphi = 5.5 \text{ kV} \) in the simulation, and then propagates rapidly to the whole membrane. This observation is illustrated by the logarithmic strain

\[ h = \frac{H}{\lambda^2} \]

**Figure 4.16 Illustration of the FE simulation process for wrinkling and crumpling of a circular VHB 4910 membrane.**
contours in the thickness direction of the membrane as plotted in Fig. 4.17 when the applied voltage reaches 5.5 kV and 6.5 kV, respectively. The critical voltage for the onset of the wrinkling in the experiment was found as 5.96 kV, which demonstrates a good agreement with the FE simulation. It should be noted that different from the EMI and the buckling, the delay effect of the viscosity is not observed in the wrinkling analysis, i.e., there is no difference in the critical voltages of wrinkling for DE membrane modelled as viscoelastic and hyperelastic media. This is attributed to the fact that when the boundaries are fully fixed and the membrane is sufficiently relaxed, the pre-stretched material can be considered as in the equilibrium state under the electric field since there is no further deformation until the membrane wrinkles locally. When the electrostatic stress exceeds the pre-stretch induced equilibrium stress, loss of tension occurs which dominates the onset of the wrinkling. As the electrostatic stress is also in the equilibrium state, the wrinkling process can thus be considered as independent of the material viscosity.

\[
\phi = 5.5 \text{ kV} \quad \phi = 6.5 \text{ kV}
\]

![Figure 4.17 Logarithmic strain contours of the wrinkled patterns of a circular VHB 4910 membrane](image)

We further increase the pre-stretch to \(\lambda_{\text{pre}} = 6\) to simulate the crumpling instability that was reported to occur with larger pre-stretch in the experiments [10]. The formation of crumples due to the loss of tension in the membrane is observed when the applied voltage reaches \(\phi = 4.95 \text{ kV}\) in the FE simulation, which is in a satisfactory agreement with the critical voltage \(\phi = 6.1 \text{ kV}\) in the experiments [10]. The simulated out-of-plane
displacement contour of the DE membrane at the onset of crumpling is plotted in Fig. 4.18 (a). With the increase of the applied voltage, the crumpling propagates till a fully crumpled pattern is reached at $\phi = 6.24$ kV once the FE simulation diverges. This fully crumpled pattern is demonstrated in Fig. 4.18 (b). Compared with the wrinkling, crumpling occurs in the DE membrane with larger pre-stretch which leads to the remarkable strain-stiffening effect and increases the bending rigidity. Meanwhile, due to the small thickness, large compressive stresses are generated in the membrane by the voltage-induced expansion after loss of tension. Then the membrane tends to bulge accompanied by large local deformations due to the concentration of stresses near the geometric imperfections [10]. As a result, crumpling is featured by the finite out-of-plane displacement similar to the buckling, but with singularities such as vertices and ridges. As discussed before, the critical voltage of the onset of the loss of tension is mainly governed by the voltage-induced stresses for the fully relaxed membrane, and the effect of the material viscosity can thus be neglected in the initiation and the rapid propagation of crumpled.

$$\phi = 4.95$ $KV \quad \phi = 6.24$ $KV$$$

![Displacement contours of crumpled patterns of a VHB 4910 membrane](image)

Figure 4.18 Displacement contours of crumpled patterns of a VHB 4910 membrane

4.5 Conclusions

In this work, a finite element (FE) framework has been established to simulate some common instability modes of DE structures including EMI, buckling, wrinkling, and crumpling. The nonlinear field theory is adopted in the FE framework to describe the electromechanical coupling, while the micro-macro constitutive model [49] is
implemented to capture the intrinsic deformation-dependent material viscosity as well as the microstructure features. The material incompressibility is successfully constrained by multiplicatively decompositing the deformation into volumetric and isochoric parts. Meanwhile, the F-bar approach is adopted in the FE framework to mitigate volumetric locking of elements when incompressibility limit is approaching. With the developed FE framework, the effects of the nonlinear material viscosity on various instability modes of DE structures with different boundary constraints have been investigated numerically. The comparison between the simulated results and the experimental data as well as the analytical calculations from the existing studies demonstrates the accuracy and robustness of the proposed FE framework. The simulation results show that the material viscosity delays the onset of the EMI and the mechanical buckling especially at high loading rates of electric voltage. Due to the decay of material viscosity with deformation, the constitutive model with nonlinear material viscosity predicts lower critical voltage for EMI compared with the viscoelastic constitutive model with constant material viscosity. However, the difference between the simulation results with constant and nonlinear viscosity is negligible in buckling analysis of the DE plate due to the small strains despite of the large out-of-plane displacement. For wrinkling and crumpling where the equilibrium voltage-induced stresses play a dominating role, the delay effect of the material viscosity can be neglected as long as the boundaries of the DE membrane are fully fixed, and DE is sufficiently relaxed. To conclude, the proposed FE framework provides a general approach for instability analysis of DE actuators, and can further function as a universal platform for accurately predicting the performance of DE-based devices with the implementation of electromechanical coupling and viscoelastic constitutive models incorporating nonlinear viscosity.

Reference


Chapter 5

5 Phase field modeling on fracture behaviors of elastomers considering deformation-dependent and damage-dependent material viscosity

Elastomers have attracted great research interests owing to their high compliance and large deformation capacity. However, with inevitable flaws, elastomers are susceptible to rupture, leading to reduction in stretchability and loss of functionality. Though phase field model (PFM) provides a feasible tool in fracture simulation on brittle elastic materials, numerical study on the fracture of elastomers is still at a tentative stage with challenges stemming from the extremely large deformation and the nonlinear material properties. In addition, recent experimental observations on the insensitivity of the onset of rupture to loading rates suggest another knowledge gap in revealing the role of the rate-dependent viscoelasticity in fracture. To capture such phenomenon and fill the research gap, this work will propose a finite element (FE) framework by incorporating the polymer dynamics into the PFM. For the first time, the driving force to the fracture of viscoelastic elastomers is identified and the micro-mechanism of the material viscosity is further elucidated with the consideration of polymer chain breakage. The disentanglement of polymer chains in the fracture process is considered in the model to release the constraints on polymer chain reptation from nearby chains, capturing the inherent damage-dependent as well as deformation-dependent material viscosity. The simulation results demonstrate good agreement with existing experimental data in both loading rate sensitivity tests and geometry sensitivity tests. With the variation of loading rates, obvious differences in stress responses are observed, while the critical fracture stretches almost keep constant. This work provides a deeper understanding on the fracture mechanisms of viscoelastic materials and is expected to provide guidance for the better design and full potential applications of elastomer-based devices.

5.1 Introduction

Over the past decades, elastomers have been widely utilized in many engineering fields [1-4] due to their unique properties including softness, flexibility, and large deformation
capacity. To facilitate better design and optimization of elastomer-based devices, the research community has devoted great efforts in establishing various constitutive models [5-9] and developing robust numerical algorithms [10-12] for the simulation of their finite deformation as well as failure behaviors [13-15]. It has been recognized that the overall performance of elastomer-based devices is dictated not only by constitutive behaviors but also by fractures especially when elastomers function as supporting and actuating components in soft robots [16] and energy harvesting devices [17]. With inevitable flaws, elastomers are susceptible to rupture, leading to the reduction in stretchability and the loss of functionality [18,19]. To fulfill the potential applications and maintain the structural integrity of elastomer-based devices, it is essential to investigate the fracture behaviors of elastomers under various loading conditions, including the prediction of crack initiation, propagation, and fracture path.

The pioneering work on the fracture mechanics can be dated back to 1920s by Griffith to explain the failure of brittle materials [20], establishing the foundation of linear elastic fracture mechanics (LEFM). The LEFM was further complemented by Irwin [21] and Rice [22] to form the essential framework, which has been widely adopted to predict the fracture behaviors of elastic bodies with sharp cracks. Elastomers usually exhibit more complicated fracture behaviors due to large deformation, nonlinear hyperelasticity and viscoelasticity, which necessitates the further development of fracture mechanics modeling. In line with the Griffith’s theory, the Lake-Thomas model [23] provided feasible measures for fracture properties of polymeric materials by formulating the critical energy release rate accounting for the total energy required to rupture the chains lying in the crack plane. Within the framework of the Lake-Thomas model, different dissipation mechanisms have been introduced to characterize the fracture behaviors of viscoelastic polymers [24, 25]. Despite the successful applications of those classical models based on the LEFM, their inherent limitations are non-negligible, i.e., the crack nucleation and propagation path need to be carefully prescribed and it becomes very challenging, if not impossible, to model the merging and branching of cracks. Meanwhile, the discontinuity of the displacement fields poses great challenges in the numerical context for the implementation of those classical fracture mechanics models.
In recent years, the research community has attempted to approach the fracture problems in an alternative way. The phase field model, also known as a variational approach, was originally proposed by Francfort and Marigo [26] by minimizing the potential energy based on the Griffith’s theory without the required information of the pre-defined crack nucleation and path. Based on the assumption that the total free energy must always remain stationary for every admissible crack-displacement pair, the crack path can be self-consistently determined. By introducing an auxiliary variable, the numerical implementation of this variational model in brittle fracture analysis was reported by Bourdin et al., [27] with convergence approximation. Such an auxiliary variable, commonly interpreted as the phase-field variable in literature, varies from 0 to 1 interpolating between an intact state and a cracked state. Another milestone research in the development of the phase field methods was conducted by Miehe et al., [28] in which this variational model was reformulated based on the framework of continuum mechanics and thermodynamics without the mathematical demanding of the convergence regulations. Owing to the solid theoretical foundation laid by those pioneering work, the research community has witnessed the prosperous development of the phase field fracture modeling across the disciplines in recent years. By revisiting the characteristics of the crack dissipation zone, some researchers proposed a set of crack surface functions with various choices of geometric crack functions for the purpose of characterizing different crack topologies, including but not limited to linear [29], quadratic [28], and double-well forms [30]. Those geometric crack functions depict how the phase field fracture variable distributes in the diffusive damage zone. From the material constitutive perspective, a degradation function was introduced to capture the effect of fracture on the degradation of material stiffness. The degradation function directly links the mechanical behavior with the phase-field variable and determines how much deformation energy is involved in crack evolution and how the material damage affects the mechanical properties of the material. The most commonly used degradation function is the quadratic function first proposed by Bourdin et al. [27]. Similar degradation functions were developed later in the forms of cubic [31] and quartic polynomials [32], and rational-type functions [33] to characterize both brittle and cohesive fracture.
The implementation of numerical techniques in solving the coupled deformation-phase field problem has, however, encountered a great challenge due to the non-convexity of the free energy functional and non-symmetricity of the stiffness matrix considering the crack irreversibility constraint. The traditional fully-monolithic solver becomes very unstable especially when the crack propagates. To tackle such difficulties, Heister et al. [34] adopted a primal-dual active set strategy by treating the phase field fracture problem as a constrained optimal control problem. Later Gerasimov and Lorenzis [35] used a line-search assisted monolithic Newton method to improve the convergence of the non-convex minimization by allowing for the negative search direction in the Newton-Raphson iterations. Borrowing the concept from the nonlinear fluid mechanics, Wick [36] further modified the Jacobian matrix by separating the ‘bad’ terms from the well-posed terms with a control parameter. More recently, Wu et al. [37] for the first time introduced the quasi-Newton scheme, namely the Broyden-Fletcher-Goldfarb-Shanno (BFGS) algorithm, to solve the coupled deformation-phase field problem. The benefit of this approach lies on the much easier implementation in commercial software packages, which has advanced the applicability of the monolithic modeling on the phase field fractures [38, 39]. On the other hand, the alternate minimization (AM) [40-43] or staggered solver also provides a feasible approach to the non-convex problems. Compared with the monolithic solvers, the advantage of the staggered algorithm is manifested by its inherent robustness: the coupled non-convex problem is decoupled into iterative linear problems by holding either the displacement or the phase field as constant while solving the other one [41]. The Jacobian matrix hence becomes always symmetric and positive definite in the staggered algorithm. Therefore, this numerical solver can be easily implemented in the existing platforms and commercial software with only certain attention to the step size [41,42].

With the development of feasible numerical algorithms, the phase field models have been widely adopted over the last decade in the simulation of the fracture behaviors of various materials ranging from isotropic to anisotropic [44, 45], brittle [35, 36, 41, 42], and ductile materials [32, 46]. However, the phase field modeling on the fracture behaviors of viscoelastic polymers is still at a tentative stage. Featured by incompressibility, large deformation and viscoelasticity, numerical modeling on the fracture behaviors of
elastomeric materials becomes highly nonlinear and requires specific attention. The material incompressibility constraint tends to cause severe volumetric locking especially at finite deformation and thus undermines the solver convergence. So far, several strategies have been reported to relieve the volumetric locking in the phase field modeling. Among those, the mixed displacement-pressure formulation framework was implemented by treating the hydrostatic pressure as another freedom of the element [47-49]. In their modeling framework, both the volumetric and the deviatoric deformation energies were considered to contribute to the cracking of materials. In the very recent years, Ye et al. [50] and Alessi et al. [51] adopted the enhanced strain approach and selective reduced integration method, respectively, in the phase field modeling and distinguished the sole contribution of the deviatoric deformation energy to fracture. In addition to those methodologies, the F-bar method [52], though not yet adopted in the phase field modeling, has gained its popularity in tackling the incompressibility constraint in the FE analysis for its easy implementation. Particularly, it has been proved feasible and efficient to treat the other highly nonlinear behaviors of polymers such as buckling and wrinkling [15]. Therefore, in the present work, we adopt the F-bar method as a solution to eliminating the volumetric locking of FE analysis of polymers at large deformation.

The material viscoelasticity further complicates the fracture behaviors of polymers. A question may be naturally raised: what is the role of material viscosity playing in the fracture of polymers? To answer this question, it is essential to reveal the driving force for fracture. Recently researchers have revisited the deformation mechanisms of the elastomeric materials to reveal the fracture origin. For example, Li and Bouklas [47] adopted the eight-chain model and attributed the cracking of elastomers to the stretch of polymer chains and Kuhn segments. The fracture energy was formulated based on the elongation of Kuhn segments with the consideration of chain distribution, which is also consistent with the bond rupture criterion. Though without accounting for the material viscosity, this work attempted to define the driving force and criteria of fracture in the phase field model to reveal the fracture mechanisms. From the continuum mechanics perspective, Yin and Kaliske [53] adopted the parallel rheology model with standard maxwell elements and accounted the nonequilibrium deformation energy from polymer
diffusion as an extra portion for the crack driving force. With this assumption, the constitutive behaviors as well as the cracking of materials become deformation rate dependent. However, some recent experimental observations [18, 54] pointed out that under tension test and pure shear test, the critical crack stretch of precut elastomers is insensitive to the stretch rates regardless of the rate dependence of the stress response. Moreover, some other research [55, 56] revealed that as the breakage of crosslinks accumulates, re-orientation of the entangled chains also known as disentanglement can be observed, which will relieve the constraints on the diffusion of confined chains and hence reduce the material viscosity. However, to the authors’ best knowledge, there is no numerical research on the fracture behaviors of elastomers that is capable of capturing such phenomena. This motivates us to develop a FE framework that integrates the micro-macro viscoelasticity constitutive model and the phase field model to accurately predict the fracture behaviors of elastomers. We start to present the formulation of the micro-macro constitutive model that incorporates the deformation-dependent material viscosity [9, 57]. With further consideration of the polymer chain breakage, we connect the phase field variable to the topology constraint on the diffusive polymer chains. Therefore, the material viscosity is also dependent on phase field variable, which is then adjusted with the evolution of the material damage. The driving energy that contributes to the fracture is also identified in the phase field model based on the rate-insensitive fracture behaviors from experimental observations. The novel constitutive model together with the modified phase field formulations are numerically discretized following the standard finite element procedure, and iteratively solved with the staggered algorithm. Finally, the numerical modeling is verified with experimental observations from the literature and the numerical results are summarized in the conclusions.

The major contributions of the current work are summarized as follows: 1. A finite element framework is established to simulate the fracture behaviors of viscoelastic elastomers with the novel incorporation of the polymer dynamics into the phase field model. 2. A new degradation scheme of the free energy density is proposed to identify the driving force for viscoelastic fracture in the phase field model. With the proposed scheme, the rate-insensitivity of critical fracture stretch is first captured with the FE simulation in the literature. 3. The deformation-dependent material viscosity is further
modified to capture the effect of polymer chain disentanglement in fracture. With the solid foundation of continuum mechanics and polymer dynamics, the proposed model is expected to provide a deeper insight into the delicate mechanisms governing the fracture behaviors of viscoelastic elastomers.

5.2 Model Formulations and Finite Element Framework

In this section, the foundations and scheme of the developed numerical framework for the simulation of the fracture behaviors of viscoelastic elastomers are presented, which is organized as follows. The classic phase field modeling is briefly reviewed in Section 5.2.1. Then a new energy degradation scheme together with a micro-macro constitutive model are described in Section 5.2.2. The evolution of the nonlinear viscosity in response to material damage and deformation is modeled in Section 5.2.3 based on the polymer dynamics to capture the disentanglement of polymer chains. In Section 5.2.4, the standard finite element discretization procedure and the staggered iteration algorithm for the implementation of the proposed framework in ABAQUS user element (UEL) subroutine will be outlined.

5.2.1 Phase field modeling

The phase field modeling assumes that the geometry discontinuity of a crack is described by a continuous auxiliary scalar function [28]. Then a phase field damage variable $\phi \in [0,1]$ is introduced with $\phi = 0$ representing an intact state and $\phi = 1$ for a fully broken state as shown in Fig. 5.1. We denote the reference (undeformed) and the current (deformed at time $t$) configurations of the elastomeric medium as $\Omega_0$ and $\Omega$, respectively. The material point is deformed from $\tilde{X}$ in reference configuration $\Omega_0$ to $\tilde{x}$ in current configuration $\Omega$. Accordingly, the initial crack configuration $\Gamma_0$ grows and expands to a newly cracked configuration $\Gamma$ after the deformation of the medium.

We first start with one-dimension simplification. In the literature, the diffusive crack topology is commonly approximated with an exponential function as [28],

$$\phi(x) = e^{-\gamma x}$$

(5.1)
where $l_0$ is the regularization constant related to the damage domain. When $l_0$ approaches 0, the diffusive crack becomes sharp. Note that such a distribution function is the solution to the following diffusion differential equation,

$$\phi(\bar{x}) - l_0^2 \phi''(\bar{x}) = 0 \text{ in } \Omega_0$$  \hspace{1cm} (5.2)

Then the crack surface functional is introduced as,

$$\Gamma_{l_0}(\phi) = \int_{\Omega_0} \gamma d\Omega_0 = \frac{1}{2l_0} \int_{\Omega_0} \left( \phi^2 + l_0^2 \phi'^2 \right) d\Omega_0$$  \hspace{1cm} (5.3)

As Eq. (5.2) is the result from the minimization of this scaled functional, it gives the solution of the regularized crack topology $\phi$, i.e., $\phi = \text{Arg inf} \{ \Gamma(\phi) \}$. From the physics perspective, $\gamma$ in this functional can be interpreted as the crack surface density function per unit volume. Accordingly, the crack surface density function can be extended to multi-dimension as,

$$\gamma(\phi, \nabla \phi) = \frac{1}{2l_0^2} \phi^2 + \frac{l_0}{2} |\nabla \phi|^2$$  \hspace{1cm} (5.4)

It is worth noting that the choice of $\gamma$ is not unique, but instead, depends on the selection of the diffusive crack topology $\phi$ in Eq. (5.4). Detailed discussions can be found in Ref. [58]. With the phase-field damage variable at hand, the crack surface energy can be formulated as,

$$\Psi_c = \int_{\Gamma} G_c \gamma(\phi, \nabla \phi) dA \approx \int_{\Omega_0} G_c \gamma(\phi, \nabla \phi) dV$$  \hspace{1cm} (5.5)

where $G_c$ is the material critical energy release rate in the classic LEFM theory.
5.2.2 Material constitutive model and energy degradation

With the consideration of the existence of cracks, the total potential energy takes the form of,

$$
\Pi = \int_{\Omega_0} W(F, \phi) dV + \int_{\Omega_0} G_c \gamma(\phi, \nabla \phi) dV - \Pi_{\text{ext}}
$$

(5.6)

where $W(F, \phi)$ is the deformation energy density depending on both the deformation gradient $F$ and the phase field variable $\phi$, and $\Pi_{\text{ext}}$ is the work done by traction $\vec{t}$ and body force $\vec{B}$. The variational form of Eq. (5.6) is then written as,

$$
\delta \Pi = \int_{\Omega_0} \frac{\partial W(F, \phi)}{\partial F} : \delta F dV - \int_{\Omega_0} \vec{t} \cdot \delta \vec{u} dV - \int_{\partial \Omega_0} \vec{n} \cdot \delta \vec{u} dA + G_c \int_{\Omega_0} \frac{\partial \gamma(\phi, \nabla \phi)}{\partial \phi} \delta \phi dV + G_c \int_{\Omega_0} \frac{\partial \gamma(\phi, \nabla \phi)}{\partial \phi} \cdot \delta \phi dV
$$

(5.7)

Applying the divergence theorem, we can further re-organize the variation of the total potential energy as,

$$
\delta \Pi = \int_{\Omega_0} \left[ \left( \frac{\partial W(F, \phi)}{\partial F} \right) \cdot \vec{n} - \vec{t} \cdot \delta \vec{u} \right] dA - \int_{\Omega_0} \left[ \nabla \cdot \left( \frac{\partial W(F, \phi)}{\partial F} \right) : \delta \vec{u} + \vec{b} \delta \vec{u} \right] dV
$$

$$
+ \int_{\Omega_0} \left[ \frac{\partial W(F, \phi)}{\partial \phi} + G_c \frac{\partial \gamma(\phi, \nabla \phi)}{\partial \phi} - G_c \nabla \cdot \left( \frac{\partial \gamma(\phi, \nabla \phi)}{\partial \phi} \right) \right] \delta \phi dV + \int_{\partial \Omega_0} \left( \frac{\partial \gamma(\phi, \nabla \phi)}{\partial \phi} \cdot \vec{n} \delta \phi \right) G_c dA
$$

(5.8)
The energy conservation gives that $\delta \Pi = 0$ for the quasi-static process. The two coupled balance equations can thus be derived as,

$$\nabla \cdot P + \tilde{b} = 0$$  \hspace{1cm} (5.9a)

$$\frac{\partial W(F, \phi)}{\partial \phi} + G_c \frac{\partial \gamma(\phi, \nabla \phi)}{\partial \phi} - G_c \nabla \cdot \frac{\partial \gamma(\phi, \nabla \phi)}{\partial \nabla \phi} = 0$$  \hspace{1cm} (5.9b)

in domain $\Omega_0$, and the boundary conditions are derived as,

$$P \cdot \bar{n} = \bar{i}$$  \hspace{1cm} (5.10a)

$$\frac{\partial \gamma}{\partial \nabla \phi} \cdot \bar{n} = 0$$  \hspace{1cm} (5.10b)

on $\partial \Omega_0$. \( P = \frac{\partial W(F, \phi)}{\partial F} \) is the first Piola-Kirchhoff stress. In Eq. (5.9b), the first term $\frac{\partial W(F, \phi)}{\partial \phi}$ can be considered as the external driving force for the evolution of the phase field variable,

Due to the loss of energy in the diffusive crack regime, the deformation energy needs to be degraded accordingly. A commonly used degradation function $g(\phi) = (1 - \phi)^2$ in the literature is also adopted in the current work. Taking an elastic body as an example, its deformation energy density in response to cracking is degraded directly from the pure elastic deformation energy $W^{\text{elastic}}(F)$ as [28],

$$W(F, \phi) = g(\phi) W^{\text{elastic}}(F)$$  \hspace{1cm} (5.11)

As elastomers intrinsically possess viscoelasticity, the degradation strategy should be modified accordingly. We start with the constitutive modelling of elastomeric materials, which are commonly described using the parallel rheological framework [9] from the mathematical perspective as shown in Fig. 5.2. Network $A$ is a purely elastic network
formed by cross-linked chains, while $N$ parallel viscous subnetworks (subnetworks $B_1$ to $B_N$) are formed by entangled chains. For the undamaged material, the deformation energy density is stored in all the springs of the whole network, therefore, it can be expressed as,

$$ W^{mec} \left( F_A, F_{B_1}^i, F_{B_2}^i, \ldots, F_{B_n}^i \right) = W^{EQ} (F) + \sum_{n=1}^{N} W^{NEQ}_{B_n} (F, F_{B_n}^i) \quad (5.12) $$

where $F = F_A = F_{B_n}$ is the global deformation acting on all the parallel networks. The deformation gradient $F_{B_n}$ in each network is commonly decomposed in a multiplicative form as, $F_{B_n} = F_{B_n}^e F_{B_n}^i$, with $F_{B_n}^e$ and $F_{B_n}^i$ representing the elastic and inelastic deformation of each individual subnetwork. In the numerical implementation, considering the material incompressibility, the global deformation gradient is further decomposed into an isochoric part $F^{iso}$ and a volumetric part $F^{vol}$ as $F = F^{iso} F^{vol} = (J^{-1/3} F) (J^{1/3})$ with $J = \det(F)$ being the volumetric change. Accordingly, the deformation energy density is decomposed as,

$$ W^{mec} \left( F^{iso}, F_{B_1}^i, F_{B_2}^i, \ldots, F_{B_n}^i, J \right) = \overline{W}^{EQ} (F^{iso}) + \sum_{n=1}^{N} \overline{W}^{NEQ} (F^{iso}, F_{B_n}^i) + U^{vol} (J) \quad (5.13) $$

where $\overline{W}^{EQ}$ and $\overline{W}^{NEQ}$ stand for the isochoric parts of the equilibrium and non-equilibrium deformation energy density. $U^{vol} (J)$ is the volumetric deformation energy density with an expression of,

$$ U^{vol} (J) = \frac{1}{2} \kappa (J - 1)^2 \quad (5.14) $$

When a relatively large bulk modulus $\kappa$ is selected, the material incompressibility is satisfied numerically.

To apply the phase field modeling to the viscoelastic elastomers with such energy decomposition, the following assumptions and experimental observations are considered:
1) Elastomers are assumed to be incompressible as commonly treated in the literature, it means that only the deviatoric deformation contributes to the cracking of materials for the numerical simulation.

2) The experimental results [18] showed that the cracking of elastomeric materials such as VHB 4910/4905 was irrelevant to the strain rate.

3) The entangled polymer chains are reported to be disentangled with the accumulation of material damage which relieves constraints on polymer chain diffusion. Therefore, the material viscosity can be assumed to vary with the evolution of damage and vanish in the fully cracked zone.

![Parallel rheological framework with crosslinked chains as ground network and entangled chains as non-equilibrium subnetworks](image)

Figure 5.2 Parallel rheological framework with crosslinked chains as ground network and entangled chains as non-equilibrium subnetworks

It is obvious that the adoption of assumption (3) leads to necessary modification on the material viscosity, which will be discussed later in the following section. Combining assumption (1) and experimental observation (2) and based on Eq. (5.9b), we propose a new energy degradation strategy to calculate the deformation energy density of the damaged medium as,

\[
W\left( F_{iso}^{vol}, F_{in}^{i}, F_{in}^{i}, ..., F_{in}^{i}, J, \phi \right) = g(\phi)W_{EQ}^{iso}\left( F_{iso}^{vol} \right) + \sum_{n=1}^{N} W_{NEQ}^{vol}\left( F_{iso}^{vol}, F_{in}^{i} \right) + U^{vol}(J) \tag{5.15}
\]
Based on the proposed energy decomposition and degradation scheme, the driving force term in Eq. (5.9b) is derived as,

\[ \frac{\partial W(F, \phi)}{\partial \phi} = g'(\phi) \bar{W}_{EQ}(F) \]  

(5.16)

It indicates that the only the isochoric elastic deformation energy density contributes to driving the crack growth. Considering the irreversibility of the crack, it is reasonable to replace the driving energy density \( \bar{W}_{EQ}(F_{\text{iso}}) \) with a historical field \( \mathcal{H} \) that can be treated as the maximum isochoric elastic energy density that has ever occurred during the deformation [59]. Therefore, the governing equation for the phase field variable can be rewritten as,

\[ g'(\phi) \mathcal{H} + G_c \frac{\partial \gamma(\phi, \nabla \phi)}{\partial \phi} - G_c \nabla \cdot \frac{\partial \gamma(\phi, \nabla \phi)}{\partial \nabla \phi} = 0 \]  

(5.17)

### 5.2.3 Nonlinear material viscosity

In this section, we briefly recall the formulation of the nonlinear material viscoelasticity of elastomers in our previous research and further adjust it with the consideration of material damage. As discussed before, the material viscoelasticity is commonly quantified with the parallel rheological framework that originating from the idealization of polymer chains with strong cross-linked ground network and diffusive entangled subnetworks as indicated in Fig. 5.2. For each nonequilibrium subnetwork, the total deformation can be decomposed as \( F_{B_n} = F_{B_n}^c F_{B_n}^i \). As the non-equilibrium stress is dependent on the viscous deformation \( F_{B_n}^i \), it is necessary to quantify the viscous reptation of the polymer chains. In this paper, we adopt the thermodynamics evolution equation based on the dissipation inequality with the assumption of the material isotropy as [9],

\[ \left[ F_{B_n}^c F_{B_n}^i \left( F_{B_n}^c \right)^T \right] \left( B_{B_n}^c \right)^{-1} = \sqrt{\gamma}^{-1} : \tau_{B_n} \]  

(5.18)
where $\mathbf{L}_{B_n}^i = \mathbf{F}_{B_n}^i \left( \mathbf{F}_{B_n}^i \right)^{-1}$ is the viscous velocity gradient tensor, $\mathbf{\tau}_{B_n} = 2\mathbf{F}_{B_n}^e \frac{\partial W}{\partial \mathbf{C}_{B_n}^e} \left( \mathbf{F}_{B_n}^e \right)^T$ is the Kirchhoff stress tensor and $\mathbf{B}_{B_n}^e$ is the elastic left Cauchy-Green deformation tensor in the nonequilibrium subnetwork. The 4th-order tensor $\gamma^{-1}$ is formulated as,

$$\gamma^{-1} = \frac{1}{2\eta_{B_n}} \left( \mathbb{I}^4 - \frac{1}{3} \mathbf{I} \otimes \mathbf{I} \right)$$

(5.19)

with $\eta_{B_n}$ being the material viscosity for the individual subnetwork $B_n$. For detailed derivation of the evolution law, please refer to reference [9]. Without losing generality, Eq. (5.18) is transformed by discarding the spin of the inelastic deformation gradient as,

$$\hat{\mathbf{F}}_{B_n}^i = \left( \mathbf{F}_{B_n}^e \right)^{-1} \left( \gamma^{-1} : \mathbf{\tau}_{B_n} \right) \mathbf{B}_{B_n}^e \left( \mathbf{F}_{B_n}^e \right)^{-T} \mathbf{F}_{B_n}^i$$

(5.20)

so that the inelastic deformation gradient can be explicitly solved.

At large deformation, the material viscosity $\eta_{B_n}$ manifesting the diffusion capability of polymer chains is no longer constant according to experimental observations [60, 61]. Such behavior has been numerically modeled in the previous research based on the theory of polymer dynamics [12]. It was assumed that the reptation of polymer chains was constrained in a tube-like region formed by nearby chains. The diffusion ability of the confined polymer chain relies on the tube diameter which is tightly related to the macro-scale deformation. The nonlinear material viscosity was reported to depend on the deformation as [9],

$$\eta_{B_n} = \frac{\eta_n}{\alpha \left( \mathbf{F} \right)^2}$$

(5.21)

where $\eta_n$ is the initial viscosity of each subnetwork. The deformation related function $\alpha \left( \mathbf{F} \right)$ depends on the mean value of the diameter $\left\langle a_n \right\rangle$ of the deformed tube as,
\[
\alpha(F) = \frac{\langle a_n \rangle}{\langle a_0 \rangle} = \frac{\int |\mathbf{F}\mathbf{R}|^2 f(\mathbf{R}) d^3 \mathbf{R}}{\left( \int \mathbf{R}^2 f(\mathbf{R}) d^3 \mathbf{R} \right) \int \mathbf{F}\tilde{u}_0 d^2 \tilde{u}_0 \frac{\mathbf{F}\tilde{u}_0}{4\pi}}
\] (5.22)

where \(\langle a_0 \rangle\) is the mean value of the tube diameter in the reference state, \(\bar{R}\) is the unit end-to-end vector of the polymer chain and \(\bar{u}_0\) is the initial unit tangent vector of the primitive chain. Each session of the polymer chain is assumed to distribute randomly, leading to the random-walk distribution of the end-to-end vector with Gaussian distribution function \(f(\mathbf{R})\).

In addition to the deformation-dependence, the material viscosity is also justified as damage-dependent in the current work as the polymer chains are disentangled in the diffusive damage zone when the crack is approaching. Since the tube model is the reflection of the constraints of the nearby polymer chains on the reptation of the polymer chain confined in the tube, the polymer chain disentanglement due to the rupture of crosslinks will relieve such constraints. It is thus natural to make a new connection between the tube diameter and the breakage of crosslinks and the disentanglement of polymer chains. Here we perceive that the degradation function \(g(\phi)\) reflects the fraction of the unbroken chains, and it is rational to assume that the mean value of the tube cross area is reciprocal to the degradation function. Accordingly, the mean value of the tube diameter is modified as,

With such an assumption, for the fully damaged zone with cracking, the tube diameter becomes infinitely large with no topology constraints to the primitive chain confined in the tube as illustrated in Fig. 5.3, the material viscosity thus vanishes.
5.2.4 Finite element implementation and iteration scheme

In this section, the phase field modeling of the fracture of elastomers is implemented into the FE simulation following the standard FE discretization procedure. The staggered integration is adopted to solve the two-field (displacement and phase field) system by holding one constant while solving the other one at each iteration. The residuals and the tangent matrices are then formulated for the finite element implementation in the Abaqus UEL subroutine.

Firstly, the displacement and the phase field variable are both discretized with shape functions $N^A(\bar{X})$ as,

$$\bar{u}(\bar{X}, t) = \sum_{A=1}^{M} N^A(\bar{X}) \bar{u}^A(t)$$  \hspace{1cm} (5.24a)$$

$$\phi(\bar{X}, t) = \sum_{A=1}^{M} N^A(\bar{X}) \phi^A(t)$$  \hspace{1cm} (5.24b)$$

where $\bar{u}^A$ is the nodal displacement and $\phi^A$ is the nodal phase field variable at node $A$.

Applying Bubnov-Galerkin approximation and substituting Eqs. (5.24) into Eq. (5.8) result in the weak form of the governing equations, as,

$$\int_{\Omega} p \frac{\partial N^A}{\partial \bar{X}} dV = \int_{\Omega} \bar{B} N^A dV + \int_{\partial \Omega} \bar{t} N^A dA$$  \hspace{1cm} (5.25a)$$
\[
\int_{\Omega_0} \left[ g' (\phi) \mathcal{H} \right] N^{d} dV + \int_{\Omega_0} \phi D (\phi, \nabla \phi) G_{c} N^{d} dV + \int_{\Omega_0} \phi D \left( \phi, \nabla \phi \right) G_{u} \frac{\partial N^{d}}{\partial X} dV = 0 \tag{5.25b}
\]

The governing equations (5.25) can be solved using the Newton-Raphson scheme, i.e.,

\[
\begin{bmatrix}
K_{u,u,j}^{AB} & K_{u,\phi,j}^{AB} \\
K_{\phi,u,j}^{AB} & K_{\phi,\phi,j}^{AB}
\end{bmatrix}
\begin{bmatrix}
\delta u_{j}^{B} \\
\delta \phi_{j}^{B}
\end{bmatrix}
= \begin{bmatrix}
R_{u,j}^{A} \\
R_{\phi,j}^{A}
\end{bmatrix}
\tag{5.26}
\]

with residuals,

\[
R_{u,j}^{A} = - \int_{\Omega_0} P_{ij} \frac{\partial N_{j}^{a}}{\partial X} dV + \int_{\Omega_0} b_{j} N_{j}^{a} dV + \int_{\Omega_0} t_{j} N_{j}^{a} dA
\tag{5.27a}
\]

\[
R_{\phi,j}^{A} = - \int_{\Omega_0} \left[ g' (\phi) \mathcal{H} \right] N_{j}^{d} dV - \int_{\Omega_0} \phi D (\phi, \nabla \phi) G_{c} N_{j}^{d} dV - \int_{\Omega_0} \phi D \left( \phi, \nabla \phi \right) G_{u} \frac{\partial N_{j}^{d}}{\partial X} dV
\tag{5.27b}
\]

and the corresponding tangents,

\[
K_{u,u,j}^{AB} = - \frac{\partial R_{u,j}^{A}}{\partial u_{j}^{B}}, \quad K_{u,\phi,j}^{AB} = - \frac{\partial R_{u,j}^{A}}{\partial \phi_{j}^{B}},
\]

\[
K_{\phi,u,j}^{AB} = - \frac{\partial R_{\phi,j}^{A}}{\partial u_{j}^{B}}, \quad K_{\phi,\phi,j}^{AB} = - \frac{\partial R_{\phi,j}^{A}}{\partial \phi_{j}^{B}}
\tag{5.28}
\]

However, it should be noted that the convexity of the free energy functional cannot be guaranteed especially when cracks propagate. The direct use of the Newton-Raphson algorithm, namely the monolithic solver, becomes unstable. In this case, we adopt the staggered algorithm to solve the two-field system as indicated in Fig. 5.4. In the odd iteration attempt of the current step \( N \), the displacement is held constant using the results from the previous step \( N-1 \), so that only the phase field variable is to be solved. While in the even iteration, the phase field from the previous step \( N-1 \) is fixed to solve the displacement. Using such an algorithm, only two tangent matrices need to be formulated as displacement and phase field variable are decoupled and can be solved linearly in each iteration. The corresponding tangent matrices are then formulated as,
To alleviate the potential volumetric locking of incompressible materials undergoing large deformation, we adopt the F-bar [15, 52] method to adjust the volumetric-isochoric multiplicative decomposition in Eq. (5.15). Replacing the deformation gradient at each integration point with the modified one having the same volumetric change of the element centroid, i.e.,

$$\bar{F} = \left( \frac{\det F_c}{\det F} \right)^{1/3} F$$  \hspace{1cm} (5.30)

where $F_c$ stands for the deformation gradient tensor at the centroid of the element. With such a treatment, the volumetric incompatibility throughout the element can be relieved. Correspondingly, the material tangent is modified referring to [52, 62], as,

$$K^{A_B} = \int_{\Omega} \frac{\partial N_A^B}{\partial \phi} N^B d\Omega + \int_{\Omega} G_{ij} \frac{\partial N_A^B}{\partial x_i} \frac{\partial N^B}{\partial x_j} d\Omega$$  \hspace{1cm} (5.29a)

$$K^{A_B} = \int_{\Omega} \frac{\partial N_A^B}{\partial x_b} \left( \int_{\Omega} F_b \frac{\partial P_{in}}{\partial F_{in}} d\Omega \right) \frac{\partial N^B}{\partial x_L} d\Omega$$  \hspace{1cm} (5.29b)

where $\bar{u}$ is the coordinate of element centroid and the 4th order tensors are,
\[ \mathbf{A}_{ijkl} = J^{-1} F_{ba} F_{La} \frac{\partial P_{ul}}{\partial F_{jn}} \]  

(5.32)

and,

\[ Q = \frac{1}{3} \mathbf{A} : (\mathbf{I} \otimes \mathbf{I}) - \frac{2}{3} \mathbf{\sigma} \otimes \mathbf{I} \]  

(5.33)

The phase field and displacement are then solved in the user element (UEL) subroutine with the corresponding residuals and the tangent matrices. For the display purpose, all the nodal information is mapped onto the third layer elements defined in the user material (UMAT) subroutine with very small elastic modulus (10^{-15} MPa) so that the solution is not affected. The detailed derivation for the residuals and tangent matrices to be coded in UEL subroutine is given in the Supplementary Materials.

![Element assembly with three layers sharing the same nodal information](image)

**Figure 5.5** Element assembly with three layers sharing the same nodal information

### 5.3 Results and Discussion

In this section, we will present several case studies to demonstrate the robustness and accuracy of the proposed numerical model. Both geometry and rate sensitivity of the fracture behaviors of VHB 4905 is investigated and the simulation results are compared with experimental data. The evolution of material viscosity is also characterized to demonstrate the deformation and damage-dependent property. Following the work of Zhou et al [9] and Feng et al [12], the Gent free energy density function is adopted for
both the equilibrium and the nonequilibrium subnetworks in the parallel rheology model as shown in Fig. 5.2. Following the decomposition algorithm as introduced in Eq. (5.15), the corresponding free energy density functions can be expressed as,

\[
\begin{align*}
\mathcal{W}_{\text{EQ}} &= -\frac{G_{\text{EQ}}^{\text{EQ}}}{2} J_{\text{lim}} \ln \left( \frac{J_{\text{lim}} - I_{1}^{\text{iso}} + 3}{J_{\text{lim}}} \right) \\
\mathcal{W}_{\text{NEQ}} &= -\frac{G_{n}^{\text{NEQ}}}{2} J_{\text{lim}} \ln \left( \frac{J_{\text{lim}} - (I_{1}^{\text{iso,e}})_{B_n} + 3}{J_{\text{lim}}} \right)
\end{align*}
\]

(5.34a)

(5.34b)

where \( G_{\text{EQ}} \) and \( G_{n}^{\text{NEQ}} \) are the shear moduli of the ground equilibrium network and the nonequilibrium subnetworks, respectively; \( J_{\text{lim}} \) is the material extensibility limit depending on the polymerization degree \( N \) of polymer chains with \( J_{\text{lim}} = 3(N - 1) \); \( I_{1}^{\text{iso}} \) and \( (I_{1}^{\text{iso,e}})_{B_n} \) are the first invariant of the isochoric elastic deformation in the equilibrium and the non-equilibrium subnetworks, respectively. The material parameters used in the simulations are listed in Table 5.1.

<table>
<thead>
<tr>
<th>Table 5.1 Material properties used for VHB 4905</th>
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<td>Parameter</td>
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The first case study focuses on the geometry sensitivity of the fracture behaviors of elastomer VHB 4905 observed in the existing experiments [18]. The onset of fracture from the FE results is compared with the experimental data to validate the accuracy of the FE model. The geometry of the model is illustrated in Fig. 5.6 with different heights \( H \). The width-height ratios are large enough to ensure the ‘pure shear’ mode with width \( B \) as 152 mm. The samples precut with a 20 mm long initial crack. The thickness of the
samples $L$ is set as 0.5 mm. In the experiments [18], the height $H$ was varied from 10 mm to 50 mm with an increment of 10 mm. To avoid slippery, the samples were wrapped around rods with a diameter of 3.2 mm. In consistency with the experimental setup, the diameter of the rod is also added in the FE model, leading to the variation of the sample height from 16.4 mm to 56.4 mm with an increment of 10 mm. The experimental loading rate 1/min is selected for each FE simulation. Due to the symmetry, half of the sample is modeled and discretized to reduce computational cost. The length scale of the phase field model $l_0$ is set as 0.75 mm. The mesh of the damaged region along the potential crack path is refined to ensure that the ratio of the length scale to mesh size is greater than 5 for the purpose of convergence [58]. The total element numbers are approximately 60,000 to 100,000. As the staggered algorithm is adopted, very small time step is used with around 20,000 steps in total.

![Figure 5.6 Geometry of single notched VHB 4905 sample and FE model setup](image)

In the present study, the fracture threshold is set with the phase field variable $\phi \geq 0.95$ for a totally damaged state of the material. Once the maximum phase field variable at the precut tip reaches the threshold, the sample is considered to be ruptured. The deformed configurations of the samples at the onset of fracture are then depicted in Fig. 5.7. The totally damaged elements at the precut tip are hidden in the postprocess with full transparency. It can be easily found that fracture occurs earlier for samples with larger height, which is consistent with experimental observations [18]. To further validate the
accuracy of the proposed numerical model, the critical stretches for the fracture onset of the samples in FE simulation are plotted in Fig. 5.8, showing an excellent agreement with the experimental data.

Figure 5.7 Deformed configurations of precut VHB 4905 samples at the onset of fracture with different heights from 16.4 mm to 56.4 mm with an increment of 10 mm.
Figure 5.8 Comparison of critical stretches between experimental data and FE simulation.

As VHB 4905 exhibits highly rate-dependent viscoelasticity, the effects of strain rate on the fracture are investigated with different loading rates on the precut VHB 4905 samples. With the same geometry setup as illustrated in Fig. 6, the height of the samples keeps a constant of H=16.4mm. Different loading rates are applied at 1/min, 10/min, and 100/min, respectively. Holding the same fracture threshold $\phi \geq 0.95$, the stress responses of the samples before fracturing at different loading rates are plotted in Fig. 5.9 in comparison with the experimental data [18]. The stress responses from the FE simulation show a similar trend as the experimental observations under high loading rates, i.e., the results lie within the error band of the experimental data. With the higher loading rate, the reptation of the polymer chain is limited due to the shorter relaxation time, leading to higher stress response as the elastic deformation remains constrained by the entanglement. Such phenomenon has been commonly recognized by the research community. However, when the loading rate is rather low (Fig. 5.9c), observable discrepancy exists in the stress responses between the FE results and the experimental data. The peak value of the nominal stress of the FE simulation occurs prior to the fracture threshold. This can be attributed to the joined contribution of the reduced material stiffness of the Gent model together with the softening effect of the degradation
function \( g(\phi) = (1 - \phi)^2 \). Such softening effect becomes more predominant especially with a relatively large diffusive zone prescribed by the dimension scale \( l_0 \). Therefore, the discrepancy between the FE simulation results and the experimental data is expected to be reduced with the decrease of \( l_0 \), while it will significantly increase the computational cost.
Figure 5.9 Stress responses of precut VHB 4905 samples under different loading rates of: (a) 100/min (b) 10/min, and (c) 1/min.

We further investigate the effects of deformation rates on the fracture behaviors of VHB 4905. The onset of fracture with different loading rates are illustrated in Fig. 5.10. As
observed from the simulation results, there is no significant change in the critical stretch for the fracture, which is in agreement with the experimental data in Ref. [18]. The detailed comparison with the experimental data is shown in Fig. 5.11. It can be concluded from these results that regardless of the highly rate-dependent viscoelasticity of VHB 4905, the fracture behaviors of the samples can be roughly considered as rate-insensitive. It could be interpreted from the physical perspective that the macro-scale fracture is considered as a result of the accumulation of the breakage of the crosslinks, not the diffusion of the entangled chains. Based on such an assumption, in the continuum mechanics treatment, only the elastic deformation energy in the ground network contributes to the fracture driving force in the phase field model, while the free energy of the entangled chains is fully dissipated.

Figure 5.10 The deformed configurations of VHB 4905 samples at the onset of fracture with different loading rates of 1/min, 10/min and 100/min.
Figure 5.11 Comparison of critical stretches between experimental data [18] and FE simulation for precut VHB 4905 samples under different loading rates.

The effects of the material damage and disentanglement of polymer chains on the material viscosity are further elucidated. In the previous studies [9, 12], the deformation-dependence of the material viscosity has been depicted with the topology constraints by the tube-like region. With the accumulation of material damage accompanied with the breakage of crosslinks, the entangled polymer chains are gradually disentangled. The disentanglement of polymer chains will release the constraints on the reptation of the polymer chain confined in the tube model. Therefore, the topology of the tube needs to be modified, which is directly linked to the degradation function as expressed in Eq. 5.23. With the phase field variable $\phi$ approaching to 1, the degradation function decreases quadratically to 0, leading to an infinitely large tube radius with no constraints on the confined chain. At this stage, the polymer chains are fully disentangled with the material viscosity vanishing in the fully damaged zone where fracture occurs. With this hypothesis in the current numerical model, the simulation results successfully capture the disentanglement effect on the material viscosity as depicted in Fig. 5.12. The material viscosity evolution of the first non-equilibrium subnetwork at the tip of the precut is also plotted in Fig. 5.13. It can be seen that the material viscosity drops dramatically to almost
0 when the critical stretch is reached to propagate the crack at the precut tip. The material viscosity of the other two non-equilibrium subnetworks exhibits the similar pattern since they share the same global deformation.

Viscosity of subnetwork 1

Figure 5.12 Material viscosity distribution at the onset of fracture of the VHB 4905 sample

Figure 5.13 Material viscosity evolution at the tip of the precut.
It is worth to mention that the damage-dependent viscosity on the other hand ensures the plausibility of the proposed model. Without the modification of the viscosity to incorporate the damage effect, the degradation strategy proposed to capture the rate-insensitivity of fracture only guarantees the decrease of the elastic deformation energy in the equilibrium ground network with the increase of phase field variable. By adopting the damage-dependence adjustment, in the damaged zone with fracture, the topology constraints on the confined chain in the tube no longer exist and therefore the corresponding elastic deformation energy vanishes. To conclude, the self-consistence of the established model lies in the fact that the decrease of the deformation energy to 0 in the non-equilibrium subnetworks is inherently the result of the disentanglement of chains.

5.4 Conclusions

A FE numerical framework is developed to investigate the fracture behaviors of viscoelastic elastomers by implementing the phase field model in ABAQUS with staggered algorithm in this work. The material viscosity is formulated as deformation-dependent and damage-dependent with the adoption of the tube model from the polymer dynamics and the involvement of the degradation function. The disentanglement of polymer chains observed in the fracturing process is also characterized by the present model. With the accumulation of material damage, the disentanglement of polymer chains loosens the topology constraints on the reptation of the confined polymer chain gradually, therefore, at the onset of the fracture, the material viscosity vanishes. The results of the geometry sensitivity simulation test present a good agreement with the experimental data, validating the accuracy and the feasibility of the proposed numerical model. The sensitivity test on loading rates exhibits high rate-dependence of the stress responses but rate-insensitivity of the fracture behaviors, which also agrees well with the experimental observations. The rationale for such behavior is that the elastic deformation energy of the crosslinked polymer network is the sole contribution to the global fracture of viscoelastic elastomers. To conclude, the developed FE model exhibits strong capability in capturing the typical fracture behaviors of viscoelastic polymers. It is expected to work as a universal computation platform for characterizing the fracture behaviors and evaluating the mechanical integrity of elastomers, which will help to
facilitate the novel design and fulfill the potential applications of elastomer-based devices.

Reference


Numerical simulation on time-dependent behaviors of self-healing hydrogels based on a generalized recursive algorithm

Self-healing hydrogels exhibit enhanced toughness due to their unique microstructures consisting of an extensible and loose crosslinked polymer network, and a brittle but healable network with tightly connected sacrificial bonds. Due to the time-dependent breaking-healing kinetics of the sacrificial network, the constitutive models of self-healing hydrogels are commonly in the form of convolution-like integral, which imposes great challenges to the numerical simulation with intolerable computational cost. In this study, we propose to solve this issue by incorporating a generalized recursive integration algorithm into the finite element (FE) framework for the simulation of the convolution-like constitutive behaviors. With the recursive algorithm, the time-dependent breaking-healing kinetics are numerically solved only based on the results from the previous last time step. The deformation behaviors of one typical self-healing hydrogel with ionic sacrificial bonds are then modeled with the proposed FE framework by adopting the full Newton-Raphson formulation, in which the recursive algorithm is expanded into the tensor space to determine the convolution-like stress and material stiffness tensors. The merits of this numerical framework in simulation capabilities and accuracy are witnessed by different case studies under both transient and equilibrium loading conditions. This proposed FE framework can also be expanded to incorporate different self-healing mechanisms of hydrogels, which is expected to act as a general avenue to numerically simulate the time/history-dependent constitutive behaviors of soft materials.

Introduction

Over the last decade, hydrogels, a typical type of soft and wet materials consisting of polymeric network and interstitial water, have drawn great research interests in tissue engineering [1-3] and biomechanical engineering [4-6] due to the merits of biocompatibility and water absorption capacity. The demands in the applications of hydrogels with long lasting usage and large load bearing capacity have motivated
extensive research efforts for the improvement of their mechanical properties. One of the pioneering examples is the development of double-network (DN) hydrogels, first synthesized by Gong et al. [7]. To obtain high mechanical strength, a loose and extensible polymer network with low ratio of cross-linking agents was incorporated into the tightly crosslinked polymer network to form an interpenetrating polymer structure. When subjected to large loads, some bonds of the tightly crosslinked chemical network also called as the sacrificial bonds will break to dissipate energy while the extensible network retains the material conformation, leading to greatly enhanced toughness and stiffness [8, 9]. By replacing the crosslinks of the sacrificial network with weak but reformable bonds such as ionic [10, 11], hydrogen [12, 13], dynamic covalent [14, 15] and metal coordination [16,17] bonds, the toughness and fatigue resistance of the DN hydrogels were further improved owing to the healing capability of the sacrificial bonds [18]. Those hydrogels, also known as self-healing hydrogels, exhibit highly nonlinear viscoelasticity due to the time-dependent and history-dependent behaviors of the re-attached bonds, which unfortunately raises great obstacles in material characterization and future structure design of self-healing hydrogels considering the computational cost in numerical simulations. Therefore, it becomes essential to establish an efficient numerical model to simulate such transient deformation behaviors.

Though the constitutive behaviors may vary among the self-healing hydrogels with different dynamic breaking-healing mechanisms, the preliminary task for material characterization is always to quantify the breaking-healing kinetics of the sacrificial network. Hui and Long [19] formulated the dynamic breaking-healing process by assuming that the debonding rate of the sacrificial ionic bonds depends on the maximum deformation that the current bonds have ever experienced, while the healing rate is related to the number of the broken bonds as well as the deformation history. Similar kinetics was also adopted for modeling the self-healing behaviors of hydrogels with metal-coordinated bonds [20] and other dynamic bonds [21,22]. Later on, with further experimental investigation [23], the breaking-healing kinetics of the self-healing of hydrogels with ionic sacrificial bonds was modified by Long et al. [24] and Venkata et al [25] to be only sensitive to the time history excluding the effects of the imposed deformation. A more physical interpretation of such kinetics was given by Lamont et al.
[26] based on statistical damage mechanics to provide a deeper insight into the self-healing mechanisms. With those established time-dependent breaking-healing kinetics framework, the contributions of the two networks of hydrogels to the sustained stresses are distinguished and the material constitutive behaviors can be accurately characterized.

Featured by time/history-dependence, most of the existing constitutive models take the convolution-like integral form, which however imposes great challenges to the corresponding numerical simulations. As the stress sustained by the reborn bonds depends on the reattachment time history, from the numerical perspective, the traditional way using trapezoidal integration method for the constitutive modeling requires the storage of all the time history and will result in intractable computational cost in the simulation of hydrogels with complex deformation. To reduce the computational cost, Guo et al. [27] used the recursive approximation for the breaking function of the sacrificial network by Prony series to numerically investigate the stabilized self-healing of hydrogels. Similarly, Ghorbanoghli and Narooei [28] also adopted the Prony series approximation to model the equilibrium hyper-viscoelasticity of the self-healing hydrogels from the macro-continuum aspect. It should be noted that such models with simplifications for the equilibrium simulation becomes ill-defined when characterizing the transient behaviors of the newly synthesized hydrogels due to the rapid change of the material constitutive behaviors. With such consideration, Liu et al. [29] recently incorporated an averaging algorithm into the FE simulation on the transient behaviors of self-healing hydrogels, while the separate contributions of the reborn and the original sacrificial bonds were still not distinguished. For the complete transient behavior simulation, the numbers of the survived sacrificial bonds as well as the total broken bonds need to be explicitly determined following a Volterra integral equation (VIE) [24] in order to characterize the effects of the time history of the reborn network. However, to the authors’ best knowledge, there is a lack of efficient numerical simulation framework on modeling the transient breaking-healing process of self-healing hydrogels due to the aforementioned challenges.

To fill this knowledge gap, we will establish a finite element (FE) framework for the simulation of the transient deformation behaviors of self-healing hydrogels by
implementing a more generalized recursive integration scheme [30] to solve the convolution-like integrals. The recursive integration algorithm can be considered as a more generalized approach of the Prony series approximation. In the developed FE framework, the full Newton-Raphson algorithm is formulated with a well-defined convolution-like material stiffness tensor, so that the deformation behaviors of both the newly synthesized and the stabilized self-healing hydrogels can be accurately characterized. For case study, we adopt the material constitutive model of one typical self-healing hydrogel with ionic sacrificial bonds, while the full capacity of the proposed integration scheme and the Newton-Raphson formulation are still reserved for different breaking-healing mechanisms formulated by similar convolution-like integrals. The accuracy and the robustness of the proposed model are verified by comparing with experimental data and cross-checking with existing models.

6.2 Model formulation and finite element discretization

In this section, we will briefly revisit the constitutive model for self-healing hydrogels considering the history-dependent breaking-healing mechanism. For numerical implementation of the self-healing thermodynamics, we will then introduce a generalized recursive integration algorithm based on the approximation for the decay function of the sacrificial bonds to solve the corresponding convolution-like integrations. On this basis, an exemplary constitutive model of the self-healing hydrogels is then implemented into the FE framework, within which the convolution-like stress tensor as well as the generalized material stiffness tensor are formulated.

6.2.1 Breaking-healing kinetics of self-healing hydrogels

The kinetics of the breaking and self-healing process is briefly summarized in this subsection. A typical poly(vinyl alcohol) (PVA) hydrogel with dual cross-linked networks is used as the example, and the detailed derivation and formulation of the constitutive model is referred to Ref. [24]. The permanent extensible (‘primary’) cross-linked bonds illustrated as circle symbols in Fig. 1(a) and Fig. 1(b) can sustain rather large deformation and therefore is considered as undamaged during the deformation without material rupture. The sacrificial (‘secondary’) bonds as visualized by rhombus
symbols breaks and reattaches dynamically. The newly reattached bonds only sustain the deformation from the time of birth while the breakage of sacrificial bonds dissipates all the stored deformation energy and assures the high toughness of the self-healing hydrogels.

Figure 6.1 Schematic illustration of breaking-healing kinetics of self-healing hydrogels: (a-b) Bonds deformation with re-bonding and breakage of sacrificial bonds. Blue symbol represents bonds without deformation and red symbol stands for bonds sustaining deformation; circle, rhombus, and triangle symbols stand for permanent bonds, connected sacrificial bonds, and broken sacrificial bonds, respectively. (c) Mathematical description of breaking-healing dynamics.

To characterize the kinetics of the breaking-healing process, we denote the total number of polymer chains as $N_0$, the number of the primary chains as $N_1$, and the number of the original secondary chains that exist at $t=0$ and remain undamaged at the current time $t$ as $N_{20}(t)$. Following the formulation by Long et al. [24], the breaking rate of the original secondary chains at the current time depends on the remaining chains in the original secondary network as,

$$
\frac{d\rho(t)}{dt} = \frac{\left(\rho(t)\right)^\alpha}{T_\alpha}
$$

(6.1)
where \( \alpha \) is a material constant and \( T_\alpha \) is the characteristic breaking time for the original secondary network. The decay function \( \rho(t) \) stands for the proportion of the remaining original secondary chains defined as \( \rho(t) = N_{20}(t)/N_{20}(0) \), which is determined by solving Eq. (6.1), i.e.,

\[
\rho(t) = \left[1 + (\alpha - 1)\frac{t}{T_\alpha}\right]^{\frac{1}{1-\alpha}} \tag{6.2}
\]

Similarly, the breaking rate of the reborn sacrificial chains that are formed at time \( \tau \) and remain connected till the current time \( t \) is formulated as,

\[
\frac{d\varphi(\tau,t)}{dt} = \frac{(\varphi(t-\tau))^\beta}{T_\beta} \tag{6.3}
\]

where \( \beta \) is a material constant and \( T_\beta \) is the characteristic breaking time for the reborn chains of the secondary network. The decay function of the reborn secondary network is derived from Eq. (6.3) as,

\[
\varphi(t-\tau) = \left[1 + (\beta - 1)\frac{t-\tau}{T_\beta}\right]^{\frac{1}{1-\beta}} \tag{6.4}
\]

Correspondingly, the total number \( N_{r2} \) of the reborn chains of the secondary network at the current time \( t \) can be formulated by the integration of,

\[
N_{r2} = \int_0^t \varphi(t-\tau)\gamma(\tau)d\tau \tag{6.5}
\]

where \( \gamma(\tau) \) is the healing rate at any time \( \tau \) that is proportional to the total broken chains \( n_h \) from both the original and the reborn secondary networks, as,

\[
\gamma(\tau) = \frac{n_h(\tau)}{T_{H}} \tag{6.6}
\]
with $T_p$ being the characteristic healing time. The conservation of the chains of the secondary network requires,

$$n_b(t) = N_{20}(0) - N_{20}(t) - \int_0^t \varphi(t-\tau)\gamma(\tau) d\tau \quad (6.7)$$

Therefore, the number of the broken chains and the reborn chains of the secondary network can be determined from the above Volterra integral equation (VIE).

### 6.2.2 Constitutive model of self-healing hydrogels

We denote a material point of the hydrogel at position $\bar{X}$ in the reference configuration $\Omega_0$, which moves to position $\bar{x}$ in the deformed configuration $\Omega$. The deformation gradient is defined as $\mathbf{F} = \partial \bar{X}/\partial \bar{x}$. When subjected to external loads, the virtual work principle states that

$$\int_{\Omega_0} \frac{\partial W(\mathbf{F})}{\partial \mathbf{F}} : \delta \mathbf{F} dV = \int_{\Omega_0} \bar{B} \cdot \delta \mathbf{x} dV + \int_{\partial \Omega_0} \bar{t} \cdot \delta \mathbf{x} dA \quad (6.8)$$

where $\bar{B}$ is the body force, $\bar{t}$ is the surface traction, and $W(\mathbf{F})$ is the free energy density function. The first Piola-Kirchhoff stress is then defined as $\mathbf{P} = \partial W/\partial \mathbf{F}$. The free energy density function for self-healing hydrogels is commonly assumed to consist of the corresponding free energy density for both primary and secondary networks while following the breaking-healing kinetics as,

$$W(t) = \frac{N_1 + N_{20}(t)}{N_0} W_0(I_1) + \frac{1}{N_0} \int_0^t \varphi(t-\tau)\gamma(\tau) W_0(I_1^{-\tau}) d\tau \quad (6.9)$$

where $W_0(I_1)$ is the free energy density for the intact polymer network, $I_1$ is the first invariant of the global deformation, and $I_1^{-\tau}$ is the first invariant of the deformation sustained by the sacrificial bonds that are reattached at instant $\tau$ and still remain connected at the current time $t$. As the reborn bonds only sustain deformation from the time it is reborn, i.e., $\tau$, the deformation gradient sustained by the reborn bonds is thus
formulated as $F^{\tau \rightarrow t} = F\left(F^{0 \rightarrow \tau}\right)^{-1}$. With the decomposition of the free energy density in Eq. (6.9), the first Piola-Kirchhoff stress can then be expressed as,

$$P = \frac{N_1 + N_{20}(t)}{N_0} \frac{\partial W_0(I_1)}{\partial F} + \frac{1}{N_0} \frac{\partial}{\partial F} \int_0^t \varphi(t - \tau) \gamma(\tau) W_0(I_1^{\tau \rightarrow t}) d\tau$$  \hspace{1cm} (6.10)

It is noted from Eqs. (6.9) and (6.10) that the constitutive model of self-healing hydrogels possesses the convolution-like behavior due to the introduction of the dynamic breaking-healing process.

### 6.2.3 Recursive algorithm for convolution integration

Although the constitutive behaviors as well as the kinetics of breaking-healing process for self-healing hydrogels are well formulated, the effective implementation of the constitutive model into numerical simulations is still challenged by solving the convolution-like integrals and the VIE in the breaking-healing kinetics. The commonly used trapezoidal method requires the results at each previous time step to be stored for summation in the current step, which however, will lead to intractable computational cost in numerical simulations with large geometry and time domain as illustrated in Fig. 6.2.

The dashed lines connect the variables to be multiplied in order to calculate the convolution integrals at different time steps. Obviously, such integration algorithm will lead to intractable computational cost in the numerical simulation particularly for cases with large geometry and increased time domain. To tackle this issue, we will adopt a generalized recursive integration algorithm [30] to model the dynamic breaking-healing process. The recursive algorithm is expanded to the tensor space to formulate the convolution-like stress tensors and material stiffness tensors.
Figure 6.2 Schematic illustration of the trapezoidal method

From the mathematical perspective, the convolution integral

\[ Y(t) = \int_0^t \gamma(\tau) \phi(t-\tau) d\tau \]

with a given function \( \gamma(\tau) \) can be numerically approximated with the recursive iteration method if the kernel function can be expanded into the following form \[30\] with total terms of \( V \),

\[
\phi(t) = \sum_{v=1}^{V} \phi_v^{(D_v)}(t) \tag{6.11}
\]

For each term,

\[
\phi_v^{(D_v)}(t) = B_v \exp(-C_v t) t^{D_v} \cos(E_v t + F_v) \tag{6.12}
\]

where the superscript \((D_v)\) stands for the power of the term \( t^{D_v} \). When \( D_v = E_v = F_v = 0 \), it is reduced to the Prony series as adopted by Guo \[27\]. With such expansion of the kernel function, the convolution integral can be expanded accordingly, as,

\[
Y(t) = \sum_{v=1}^{V} Y_v(t) \tag{6.13}
\]
Adopting the trapezoidal integration method, the numerical convolution integration for each term in Eq. (6.13) at $t + \Delta t = (N + 1)\Delta t$ ($\Delta t$ is the discretized time step) can be formulated as,

$$Y_v (t + \Delta t) = \Delta t \sum_{m=0}^{N+1} b_m \gamma (m\Delta t) \phi_v^{(D_v)} ((N + 1 - m)\Delta t)$$

(6.14)

where $b_m$ is the weighting constant at time $m\Delta t$, $N$ is the number of the discretized time steps within time $t$. For the purpose of convenient notation, Eq. (6.14) is rewritten as,

$$Y_{v,N+1} = \Delta t \sum_{m=0}^{N+1} b_m \gamma_m \phi_v^{(D_v)}_{N+1-m}$$

(6.15)

It is obvious that all the information at each time step needs to be explicitly stored for the direct integration, which is almost impossible for most numerical techniques.

Before we proceed to calculate the convolution integral, we rewrite the kernel function $\phi_v^{(D_v)}(t)$ with the recursive approximation. Firstly, the kernel function for any instant $t = N\Delta t$ is rewritten with the discretized time as,

$$\phi_v^{(D_v)} = B_v \exp(-C_v N\Delta t)(N\Delta t)^{D_v} \cos(E_v N\Delta t + F_v)$$

(6.16)

Similarly, for next time step $t + \Delta t = (N + 1)\Delta t$, the kernel function is determined as,

$$\phi_v^{(D_v)}_{N+1} = \sum_{k=0}^{D_v} \binom{D_v}{k} \left[(\Delta t)^{D_v-k} \frac{1}{(k!)^2} \left[\exp(-C_v N\Delta t)\exp(-C_v \Delta t)(N\Delta t)^{D_v-2k} \times \right.\right.$$

$$\left.\left[\cos(E_v N\Delta t + F_v)\cos(E_v \Delta t) - \sin(E_v N\Delta t + F_v)\sin(E_v \Delta t)\right] \right]$$

(6.17)

where $\binom{D_v}{k} = \frac{D_v (D_v - 1) (D_v - 2) \ldots (D_v - k + 1)}{k!}$ is the binomial coefficient. For further simplification purpose, we denote the following variables,

$$R_{v1} = \exp(-C_v \Delta t), \quad S_{v1} = \cos(E_v \Delta t), \quad \text{and} \quad S_{v1}^{\text{conj}} = \sin(E_v \Delta t)$$

(6.18)
and a conjugate pair of the kernel function,
\[ \varphi_{v,N}^{(D_v)} = B_v \exp(-C_v N \Delta t)(N \Delta t)^{D_v} \sin(E_v N \Delta t + F_v) \] (6.19)

With those defined variables, the kernel function at \( t + \Delta t = (N+1) \Delta t \) is written as,
\[ \varphi_{v,N+1}^{(D_v)} = R_v \sum_{k=0}^{D_v} \left( \frac{D_v}{k} \right) \left( \Delta t \right)^{D_v-k} \left( S_{v1} \varphi_{v,N}^{(k)} - S_{v1} \varphi_{v,N}^{\text{conj.}(k)} \right) \] (6.20)

where \( \varphi_{v,N}^{(k)} \) is the recursive kernel function with power of \( k \) at time step \( t = N \Delta t \) and \( \varphi_{v,N}^{\text{conj.}(k)} \) is the corresponding conjugate pair of \( \varphi_{v,N}^{(k)} \). Similarly, the conjugate kernel function with power of \( D_v \) at \( t + \Delta t = (N+1) \Delta t \) is formulated as,
\[ \varphi_{v,N+1}^{\text{conj.}(D_v)} = R_v \sum_{k=0}^{D_v} \left( \frac{D_v}{k} \right) \left( \Delta t \right)^{D_v-k} \left( S_{v1} \varphi_{v,N}^{\text{conj.}(k)} + S_{v1} \varphi_{v,N}^{(k)} \right) \] (6.21)

Now, we can reformulate the convolution integral at \( t + \Delta t = (N+1) \Delta t \) with the recursive kernel function. Eq. (6.15) is thus transformed as,
\[ Y_{v,N+1} = \Delta t Q_{v,N+1}^{(D_v)} + \Delta t b_{N+1} \gamma_{N+1} \varphi_0 \] (6.22)

with \( Q_{v,N+1}^{(D_v)} = \sum_{m=0}^{N} b_m \gamma_m \varphi_{v,N+1-m}^{(D_v)} \). Substituting \( \varphi_{v,N+1}^{(D_v)} \) from Eq. (20) into the expression of \( Q_{v,N+1}^{(D_v)} \), a similar recursive formulation is obtained as,
\[ Q_{v,N+1}^{(D_v)} = \left( \sum_{k=0}^{D_v} \left( \frac{D_v}{k} \right) \left( \Delta t \right)^{D_v-k} \left( R_{v1} S_{v1} Q_{v,N}^{(k)} - R_{v1} S_{v1} Q_{v,N}^{\text{conj.}(k)} \right) \right) + b_{N} \gamma_{N} \varphi_{v,1} \] (6.23)

with \( Q_{v,N}^{\text{conj.}(k)} = \sum_{m=0}^{N-1} b_m \gamma_m \varphi_{N-m}^{\text{conj.}(k)} \) as the corresponding conjugate pair with power of \( k \) at time \( t \). Similarly, the conjugate pair \( Q_{v,N+1}^{\text{conj.}(D_v)} \) at \( t + \Delta t = (N+1) \Delta t \) is determined as,
\[ Q_{v,N+1}^{\text{conj}(D_v)} = \left[ \sum_{k=0}^{D_v} \binom{D_v}{k} (\Delta t)^{D_v-k} \left( R_{v1} S_{v1} Q_{v,N}^{\text{conj}(k)} + R_{v1} S_{v1} Q_{v,N}^{(k)} \right) \right] + b_N \gamma_N \varphi_{v,1}^{\text{conj}(D_v)} \] (6.24)

Therefore, the convolution integral can be recursively approximated by using the variables from the previous step without storing all the history information at each step, i.e.,

\[ Y_{N+1} = \Delta t \sum_{v=1}^{Y} \left[ \sum_{k=0}^{D_v} \binom{D_v}{k} (\Delta t)^{D_v-k} \left( R_{v1} S_{v1} Q_{v,N}^{(k)} - R_{v1} S_{v1} Q_{v,N}^{\text{conj}(k)} \right) \right] + b_N \gamma_N \varphi_{v,1}^{(D_v)} + b_{N+1} \gamma_N \varphi_{v,0}^{(D_v)} \] (6.25)

Each pair of \( Q_{v,N}^{(k)} \) and \( Q_{v,N}^{\text{conj}(k)} \) \((k \text{ from } 0 \text{ to } D_v)\) can be recursively updated from the previous step with the initial conditions,

\[ Q_{v,N}^{(k)} = Q_{v,N}^{\text{conj}(k)} = 0 \] (6.26)

With the recursive integration scheme at hand, the breaking-healing kinetics following VIE in Eq. (6.7) can be recursively solved to determine the evolution of the number of the broken chains and the healing rate. The VIE at \( t + \Delta t = (N + 1) \Delta t \) is transformed as,

\[
n_{b,N+1} = N_{b20}(0) - N_{b20}(t + \Delta t) \frac{\Delta t}{T_H} n_{b,N+1} \phi_{v,0}^{(D_v)} - \frac{\Delta t}{T_H} \sum_{v=1}^{Y} \left[ \sum_{k=0}^{D_v} \binom{D_v}{k} (\Delta t)^{D_v-k} \left( R_{v1} S_{v1} Q_{v,N}^{(k)} - R_{v1} S_{v1} Q_{v,N}^{\text{conj}(k)} \right) + b_N n_{b,N} \phi_{v,1}^{(D_v)} \right] \] (6.27)

Therefore, the number of the broken chains at \( t + \Delta t = (N + 1) \Delta t \) can be explicitly determined as,

\[
n_{b,N+1} = \frac{T_H \left[ (1 - \rho_{v,1}) N_{b20}(0) \right]}{T_H + \Delta t b_{N+1} \sum_{v=1}^{Y} \phi_{v,0}^{(D_v)}} - \frac{\Delta t}{T_H + \Delta t b_{N+1} \sum_{v=1}^{Y} \phi_{v,0}^{(D_v)}} \sum_{v=1}^{Y} \left[ \sum_{k=0}^{D_v} \binom{D_v}{k} (\Delta t)^{D_v-k} \left( R_{v1} S_{v1} Q_{v,N}^{(k)} - R_{v1} S_{v1} Q_{v,N}^{\text{conj}(k)} \right) + b_N n_{b,N} \phi_{v,1}^{(D_v)} \right] \] (6.28)
As all the variables in this equation are obtained directly from the results in the previous last step and the initial step, the number of the broken chains can be recursively determined without excessive information storage.

### 6.2.4 Numerical implementation of the constitutive model and the recursive integration algorithm

In this section, the constitutive model as well as the kinetics of the breaking-healing process of self-healing hydrogels are implemented into the finite element framework following the standard discretization procedure. The full Newton-Raphson algorithm is formulated with the detailed expressions of residuals and tangent matrices by expanding the recursive algorithm to tensor space in ABAQUS user element (UEL) subroutine for FE implementation [31].

The standard FE discretization requires the displacement to be discretized with shape function \( N^A(\overline{X}) \) as,

\[
\ddot{u}(\overline{X}, t) = \sum_{A=1}^{M} N^A(\overline{X}) \ddot{u}^A(t)
\]  

(6.29)

where \( \ddot{u}^A \) is the nodal displacement at node A and M is the total number of nodes in each element. Applying the Bubnov-Galerkin approximation, the thermodynamics law in Eq. (6.8) is transformed as,

\[
\int_{\Omega^o} \rho \frac{\partial N^A}{\partial X} dV = \int_{\Omega^o} \overline{B} N^A dV + \int_{\partial \Omega^o} \overline{t} N^A dA
\]  

(6.30)

This weak form of the governing equation can be iteratively solved using the Newton-Raphson scheme as,

\[
K^{AB} \delta \ddot{u}^B = \ddot{R}^A
\]  

(6.31)

with residual

\[
\ddot{R}^a = \int_{\Omega^o} \rho \frac{\partial N^A}{\partial X} dV - \int_{\Omega^o} \overline{B} N^A dV - \int_{\partial \Omega^o} \overline{t} N^A dA
\]  

(6.32)
and tangent matrix

\[ K^{AB} = -\frac{\partial \tilde{R}^d}{\partial u^B} \]  

(6.33)

In order to model the constitutive behavior of the self-healing hydrogels, the stress tensor as well as the tangent matrix that are also in the form of convolution-like integrals need to be recursively formulated. As dry hydrogels are commonly incompressible, the deformation energy density is further modified based on the decomposition of the deformation gradient into an isochoric part \( F^{iso} \) and a volumetric part \( F^{vol} \) as \( F = F^{iso} F^{vol} = (J^{-1/3} F)(J^{1/3} I) \) with \( J = \det(F) \). Accordingly, the total free energy density in Eq. (6.9) is decomposed as,

\[
W(t) = \left( \frac{N_1 + N_20(t)}{N_0} \right) \left[ W^{iso}_0 \left( I_1^{iso} \right) + U(J) \right] \\
+ \frac{1}{N_0} \int_0^t \gamma(t) \varphi(\tau,t) W^{iso} \left( I_1^{iso,\tau \rightarrow t} \right) d\tau + \frac{1}{N_0} \int_0^t \gamma(t) \varphi(\tau,t) U(J^{\tau \rightarrow t}) d\tau
\]

(6.34)

with \( I_1^{iso} \) being the first invariant of the isochoric right Cauchy-Green tensor \( C^{iso} = F^{iso} \left( F^{iso} \right)^T \). The superscript "\( \tau \rightarrow t \)" stands for the time history of the variables from instant \( \tau \) to the current time \( t \). The variables without time history specification are all formulated based on the intact configuration from the starting time \( t=0 \) to the current time \( t \). The free energy density function caused by volumetric change commonly takes the form of,

\[
U(J) = \frac{1}{2} \kappa (J - 1)^2
\]

(6.34)

With relatively large bulk modulus \( \kappa \), the material incompressibility can be imposed for both the original network and the reborn network without considering the swelling behaviors of hydrogels. In this paper, following the constitutive modeling work by Long et al. [24], the Neo-Hookean hyperelasticity model is adopted for the unbroken networks subjected to isochoric deformation, i.e.,
\[ W_0^{iso}(I_0^{iso}) = \frac{1}{2} \mu (I_1^{iso} - 3) \]  \hfill (6.36)

Correspondingly, the Cauchy stress can be derived following the relation \( \sigma = J^{-1}PF^T \), as,

\[
\sigma(t) = \left( \frac{N + N_{20}(t)}{N_0} \right) \sigma_0 + \sigma_{rb}^{iso} + \sigma_{rb}^{vol} 
= \left( \frac{N + N_{20}(t)}{N_0} \right) \left[ 2F \frac{\partial W_0^{iso}(I_1^{iso})}{\partial C} F^T + 2F \frac{\partial U(J)}{\partial C} F^T \right] 
+ \frac{\mu}{N_0} \int_0^t \gamma(\tau,t) \phi(\tau,t) F \left( \frac{\partial C^{\tau\rightarrow\tau}}{\partial C} \right) F^T d\tau
+ \frac{\kappa}{J_0^{\tau\rightarrow\tau}} \int_0^t \gamma(\tau,t) \phi(\tau,t) \left( \frac{J}{J_0^{\tau\rightarrow\tau}} - 1 \right) \frac{J}{J_0^{\tau\rightarrow\tau}} d\tau I \tag{6.37}
\]

\( \sigma_0 \) is the Cauchy stress for the intact networks, which can be directly calculated with tensor calculus following a similar procedure in the previous research \( [32] \). The second term \( \sigma_{rb}^{iso} \) in Eq. (6.37) is the isochoric stress in the reborn secondary network, which can be further rewritten as,

\[
\sigma_{rb}^{iso}(t) = \sigma_{ri}^{iso}(t) + \sigma_{ri}^{iso}(t)
= \frac{\mu}{T_H J N_0} \int_0^t n_b(t) \phi(\tau,t) J^{-1/3} B^{\tau\rightarrow\tau} d\tau - \frac{1}{3} tr(\sigma_{ri}^{iso}(t)) I \tag{6.38}
\]

It is obvious that the determination of the isochoric stress relies on the deformation history, leading to a convolution-like integral from the mathematical perspective. To numerically calculate the convolution-like integral for a tensor, the kernel function is also expanded into the same format as shown in Eq. (6.11) with the same expressions for each term in Eq. (6.12) as for the convolution integral of scalar functions. At time \( t + \Delta t = (N + 1) \Delta t \), each term of \( \sigma_{ri}^{iso}(t) \) can be approximated as,

\[
\sigma_{ri, N+1}^{iso} = \frac{\mu \Delta t}{J T_H N_0} \left( J^{N\rightarrow N+1} \right)^{2/3} F^{N\rightarrow N+1} \left( \sum_{m=0}^N b_m v_m I \phi_s^{(\text{D},)} \left( J^{m\rightarrow N} \right)^{-2/3} F^{m\rightarrow N} \left( F^{m\rightarrow N} \right)^T \right) \left( F^{m\rightarrow N} \right)^T \tag{6.39}
\]

\[
+ \frac{\mu \Delta t}{J T_H N_0} b_{N+1} v_{N+1} \phi_s^{(\text{D},)} I
\]
The superscript "m \rightarrow N" stands for the relative change of the variables from time \( t = m\Delta t \) to \( t = N\Delta t \).

In Eq. (6.39), we further denote a new summation tensor corresponding to the power of \( D_v \) as \( Q^{(D_v)}_{v,N+1} = \sum_{m=0}^{N} b_m n_h, m \phi^{(D_v)}_{v,N+1-m} \left( J^{m\rightarrow N} \right)^{-2/3} F^{m\rightarrow N} \left( F^{m\rightarrow N} \right)^T \) which can be recursively determined as,

\[
Q^{(D_v)}_{v,N+1} = \left( J^{N-1\rightarrow N} \right)^{-2/3} F^{N-1\rightarrow N} \left[ \sum_{k=0}^{D_v} \left( \frac{D_v}{k} \right)(\Delta t)^{D_v-k} \left( R_{v_{1}}S_{v_{1}}^{(k)}Q^{(k)}_{v,N} - R_{v_{1}}S_{v_{1}}^{\text{conj.(k)}}Q_{v,N}^{\text{conj.(k)}} \right) \right] \left( F^{N-1\rightarrow N} \right)^T + b_n n_h, m \phi^{(D_v)}_{v,N+1}^1 I
\]

(6.40)

The conjugate form \( Q_{v,N}^{\text{conj.}(D_v)} \) of the summation function is defined as,

\[
Q_{v,N}^{\text{conj.}(D_v)} = \sum_{m=0}^{N-1} b_m n_h, m \phi_{v,N-m}^{\text{conj.}(D_v)} \left( J^{m\rightarrow N-1} \right)^{-2/3} F^{m\rightarrow N-1} \left( F^{m\rightarrow N-1} \right)^T
\]

(6.41)

which can also be recursively updated with,

\[
Q_{v,N+1}^{\text{conj.}(D_v)} = \left( J^{N\rightarrow N-1} \right)^{-2/3} F^{N\rightarrow N-1} \sum_{k=0}^{D_v} \left( \frac{D_v}{k} \right)(\Delta t)^{D_v-k} \left( R_{v_{1}}S_{v_{1}}^{\text{conj.}(k)}Q_{v,N}^{(k)} + R_{v_{1}}S_{v_{1}}^{\text{conj.}(k)} Q_{v,N}^{\text{conj.(k)}} \right) \left( F^{N\rightarrow N-1} \right)^T + b_n n_h, m \phi^{(D_v)}_{v,N+1}^\text{conj.}
\]

(6.42)

Similarly, the volumetric stress \( \sigma_{rb}^{\text{vol}} \) in Eq. (6.37) is also approximated with the recursive algorithm with formulation for each term as,

\[
\sigma_{rb,v}^{\text{vol,}N+1} = \frac{\kappa \Delta t}{T_{H}N_0} X_{N+1}^{(D_v)} - \frac{\kappa \Delta t}{T_{H}N_0} Y_{N+1}^{(D_v)}
\]

\[
= \frac{\kappa \Delta t}{T_{H}N_0} \left\{ \sum_{k=0}^{D_v} \left( \frac{D_v}{k} \right)(\Delta t)^{D_v-k} \left( R_{v_{1}}S_{v_{1}}^{(k)}X_N^{(k)} - R_{v_{1}}S_{v_{1}}^{\text{conj.(k)}} X_N^{\text{conj.(k)}} \right) + b_n n_h, m \phi^{(D_v)}_{v,N+1} \left( J_0^{0\rightarrow N+1} \right)^{-2} \right\} - \frac{\kappa \Delta t}{T_{H}N_0} \left\{ \sum_{k=0}^{D_v} \left( \frac{D_v}{k} \right)(\Delta t)^{D_v-k} \left( R_{v_{1}}S_{v_{1}}^{(k)}X_N^{(k)} - R_{v_{1}}S_{v_{1}}^{\text{conj.(k)}} X_N^{\text{conj.(k)}} \right) + b_n n_h, m \phi^{(D_v)}_{v,N+1} \left( J_0^{0\rightarrow N} \right)^{-2} \right\}
\]

(6.43)
With two pairs of recursive summation \((X_{N+1}^{(Dv)}), (Y_{N+1}^{(Dv)})\) and \((X_{N+1}^{(conj,Dv)}), (Y_{N+1}^{(conj,Dv)})\), the volumetric stress tensor is explicitly determined for each time step following the same procedure as elaborated for the convolution integral of scalar functions in the previous Section.

To obtain good convergence of the Newton-Raphson scheme, in addition to the stress tensor, the tangent matrix also needs to be carefully formulated with the recursive algorithm. For each element, the tangent matrix as defined in Eq. (6.33) is further expanded as,

\[
K_{u,ij}^{AB} = \int_{\Omega} \frac{\partial N^A}{\partial x_m} J^{-1} F_{ma} \frac{\partial P_{ia}}{\partial F_{jb}} F_{nb} \frac{\partial N^B}{\partial x_n} dV \tag{6.44}
\]

Here we denote the material stiffness tensor in the current configuration as

\[
K_{imjn} = J^{-1} F_{ma} \frac{\partial P_{ia}}{\partial F_{jb}} F_{nb}.
\]

Following the decomposition scheme of the free energy density, the material stiffness tensor can also be decomposed as,

\[
K = \left( \frac{N + N_{20}(t)}{N_0} \right) K^0 + K_{iso}^{0} + K_{vol}^{0} \tag{6.45}
\]

where \(K^0\) is the material stiffness for the case without bond breakage and is formulated as,

\[
K_{imjn}^0 = (\sigma_0)_m \delta_{nj} - (\sigma_0)_n \delta_{mj} + F_{nb} \frac{\partial (\sigma_0)_m}{\partial F_{jb}} \tag{6.46}
\]

This 4th-order tensor can be directly calculated without further convolution-like integral treatment following our previous research [32]. The isochoric and the volumetric parts of the material stiffness for the reborn network in Eq. (6.45) are expressed as,
\[
\begin{align*}
\mathbf{K}_{iso}^{rb}(\delta_{in}) &= \left(\sigma_{in}^{iso} + \sigma_{in}^{iso}\right) \delta_{nj} - \left(\sigma_{in}^{iso} + \sigma_{in}^{iso}\right) \delta_{mj} \\
&+ F_{nb} \left(\frac{\partial \sigma_{in}^{iso}}{\partial F_{jb}} + \frac{\partial \sigma_{in}^{iso}}{\partial F_{jb}}\right) \\
\mathbf{K}_{vol}^{rb}(\delta_{in}) &= \left(\sigma_{in}^{vol} \delta_{nj} - \left(\sigma_{in}^{vol} \delta_{mj} + F_{nb} \frac{\partial \sigma_{in}^{vol}}{\partial F_{jb}}\right) \right)
\end{align*}
\]

With the obtained expressions for Cauchy stresses from Eq. (6.38), Eq. (6.39) and Eq. (6.43) and after tedious derivation with the recursive algorithm, the material stiffness tensors for the isochoric and the volumetric deformation of the reborn network at time \(t + \Delta t = (N + 1) \Delta t\) are finalized as,

\[
\begin{align*}
\mathbf{K}_{iso}^{rb,N+1}(\delta_{in}) &= \left(\sigma_{I,N+1}^{iso} \delta_{ij} - \frac{2}{3} \left(\sigma_{I,N+1}^{iso} + \sigma_{II,N+1}^{iso}\right) \delta_{mi}\right) \\
&- \frac{2}{3} \left(\sigma_{I,N+1}^{iso} \delta_{mj} - \left(\sigma_{II,N+1}^{iso}\right) \right) \\
\mathbf{K}_{vol}^{rb,N+1}(\delta_{in}) &= \left(\sigma_{I,N+1}^{vol} \delta_{nj} - \left(\sigma_{I,N+1}^{vol} \delta_{mj}\right) + \frac{\kappa \Delta t \delta_{mj} \delta_{nj}}{T_{H}N_{0}} X_{N+1}^{(Dv)}\right)
\end{align*}
\]

in which the summation \(X_{N+1}^{(Dv)}\) has been formulated recursively in Eq. (6.43). With the well-defined stiffness tensors, the tangent matrix can be assembled in Eq. (6.44) for the good convergence of the Newton-Raphson scheme.

### 6.3 Results and Discussion

In this section, several case studies are performed on the transient responses of a newly synthesized self-healing hydrogel and the deformation behaviors of a stabilized self-healing hydrogel. The simulation results are compared with experimental data and other numerical methods for the validation purpose, which also demonstrate the accuracy and the robustness of the proposed recursive integration algorithm and the FE framework in tackling both the equilibrium and transient behaviors of self-healing hydrogels. As we
mainly focus on the performance of the proposed numerical framework, the material behaviors are only briefly depicted without detailed discussion.

### 6.3.1 Transient responses of newly synthesized self-healing hydrogels

Long et. al. [24] synthesized a poly(vinyl alcohol) (PVA) hydrogel with dual cross-links and conducted different loading tests to characterize the kinetics of the breaking-healing process. In this paper, we adopted the same constitutive model as well as the material parameters obtained from Long’s research so that the simulation results can be directly compared with the experimental data for validation. For the implementation of the recursive algorithm, the decay function is approximated with Eq. (6.11). For the transient simulation, from our practical experience, the fitted decay equation with 3 fitting terms ($V=3$) are accurate enough as demonstrated in Fig. 6.3. The fitted constants in Eqs. (6.11) and (6.12) are listed in Table 6.1.

<table>
<thead>
<tr>
<th>$v$</th>
<th>$B_v$</th>
<th>$C_v$</th>
<th>$D_v$</th>
<th>$E_v$</th>
<th>$F_v$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.4889</td>
<td>1.419</td>
<td>1</td>
<td>0.03132</td>
<td>-0.07015</td>
</tr>
<tr>
<td>2</td>
<td>0.9365</td>
<td>2.786</td>
<td>0</td>
<td>0.04621</td>
<td>0.009149</td>
</tr>
<tr>
<td>3</td>
<td>0.06219</td>
<td>0.1816</td>
<td>0</td>
<td>0.01266</td>
<td>-0.1647</td>
</tr>
</tbody>
</table>
Using the same loading conditions as Ref. [24], tensile tests are simulated with different loading/unloading rates: 0.3 s\(^{-1}\) as the loading rate with the unloading rate changing from 0.1 s\(^{-1}\) to 0.01 s\(^{-1}\) and 0.001 s\(^{-1}\); 0.01 s\(^{-1}\) as the loading rate with the unloading rate changing from 0.1 s\(^{-1}\), 0.01 s\(^{-1}\), and 0.001 s\(^{-1}\). The relaxation test is also simulated to capture the healing behavior of the hydrogel. The samples are stretched to initial stretch \(\lambda = 1.1, 1.2,\) and 1.5 in 1 s, and then are held for a long relaxation time. The comparison between the simulation results and the experimental data are plotted in Fig. 6.4 and Fig. 6.5 for tensile tests and relaxation test, respectively. The newly synthesized self-healing hydrogels exhibit highly viscoelastic deformation behaviors due to the dynamic breaking-healing of the sacrificial bonds. Within the relaxation time, the stresses keep decreasing until it becomes stable. The detailed interpretation of the test results can be referred to Ref. [24]. The good agreement indicates that the recursive algorithm can well characterize the dynamic breaking-healing behaviors of the self-healing hydrogels.
Figure 6.4 Stress responses in loading-unloading tests with different loading rates in (a) and different unloading rates in (b). Solid lines are obtained from simulation and symbols are from experiments [24].

Figure 6.5 Stress responses in relaxation test: materials are first stretched and then held with constant stretch ratios of 1.1, 1.2 and 1.5. Solid lines are obtained from simulation and symbols are from experiments [24].
6.3.2 Equilibrium behavior of stabilized self-healing hydrogels

The constitutive behaviors of the stabilized self-healing hydrogels become rather different from the newly synthesized samples. The breaking and reattaching of the physical sacrificial bonds will achieve dynamic equilibrium soon after the gel is synthesized. Such behavior can also be simulated with the proposed recursive algorithm. To do so, the transient approach is adopted, with which the breaking-healing kinetics is modeled from the time when the material is newly synthesized, and the external load is not applied until the equilibrium state is achieved. In this section, we conduct several simulations to verify the accuracy and the robustness of our proposed algorithm in the equilibrium simulation within a long-time range. The material properties are directly adopted from Guo’s work [27, 33] for the stabilized hydrogels. Without losing accuracy and generality, we use the same material parameters for the reborn sacrificial network and the original sacrificial network with the assumption that the original sacrificial network totally breaks before loading.

For comparison purpose, the numerical model proposed by Guo et. al. [27] that mainly focuses on the equilibrium state for the stabilized materials is also presented. In Guo’s stable approach, the healing rate of the broken bonds in the steady state is assumed to be constant and then explicitly determined as the new initial conditions for the equilibrium simulation as [24],

\[
\gamma_s = \frac{N_0 - N_{10}}{t_H + t^\beta/(2 - \beta)} \quad (6.51)
\]

The stable number of the reborn bonds before loading is hence approximated as 

\[ n_b^s = \gamma_s t_H \].

Meanwhile, as the survived reborn bonds have already experienced time/damage history before loading, the decay function for the newly bonded network is no longer applicable in the equilibrium-state simulation. Guo et. al [27] used the integral of the decay function to characterize the kinetics of the physical sacrificial bonds as,
\[
\varphi^\sigma (t) = \int_{-\infty}^{t} \varphi(t-\tau) d\tau = \frac{t^\beta}{2-\beta} \left(1 + (\beta - 1) \frac{t}{t^\beta}\right)^{2-\beta} \tag{6.52}
\]

With those simplifications, the stable approach can be adopted in modeling the stabilized self-healing hydrogels while losing the capability in the simulation of the transient behaviors. The differences of the two integration approaches are illustrated in Fig. 6.6.

**Figure 6.6 Schematic comparison between the transient integration approach and the stable approach.**

The stabilization process as well as the following uniaxial test of the stabilized self-healing hydrogels are then simulated based on both the proposed transient approach and the stable approach [27] for a special case of the natural stabilization of self-healing hydrogels without applying external loads. The evolution of the healing rate of the hydrogel during the stabilization process is first examined to investigate the accuracy of our complete transient scheme. Within this time region, as no load is applied to the homogenous material, the classical trapezoidal integration with both fitted decay function and the original decay function can be utilized for comparison due to relatively low storage cost. In addition, as the stabilization process only involves the healing kinetics, the theoretical solution by Laplace transform can also be obtained if the kernel function...
in the VIE can be expanded as the exponential functions (Prony series). Once the equilibrium state is achieved, the deformation of the stabilized hydrogel is then simulated under the loading/unloading rate of 0.01 s\(^{-1}\) with the transient scheme, which is compared with the results from the stable approach [27].

The evolution of the healing rate during the stabilization is plotted in Fig. 6.7 with different fitting terms for the decay function. After a long-time rest, the healing rate becomes stable converging to a theoretical limit which can be calculated from Eq. (6.51). Surprisingly, even though 3 terms (V=3) of the fitting equation for the decay function are accurate enough for the transient simulation within a short period as discussed in the previous section, observable error still exists after a long rest time in the equilibrium simulation. Similar accumulated error is also observed in Fig. 6.7(b) for the classical trapezoidal integration method if the fitted decay function instead of the original decay function is used. The reason behind that is the local fitting performance is not satisfied especially when the decay function decreases to a rather small value despite the good overall fitting achieved with fewer fitting terms. Such error is then exaggerated in the following loading simulation on the stabilized material. The stress response of the stabilized material under a loading-unloading rate of 0.01/s is plotted in Fig. 6.8 using the proposed transient scheme. The simulation result by using the stable approach [27] is also provided for comparison. It is observed that simulation result with 5 fitting terms (V=5) for the decay function almost overlaps with that of the stable approach, while the fitting function with 3 terms leads to a relative error around 10% in the maximum stress. It is thus concluded that a good fitting of the decay function plays a significant role in reducing the simulation error in the transient algorithm.
Figure 6.7 Healing rate evolution with different fitting terms V in the decay function by (a) recursive integration algorithm and (b) trapezoidal integration algorithm.

Figure 6.8 Stress-stretch curves simulated with the recursive algorithm using different fitting terms V in the decay function, and the result of stable approach is reproduced from [27].

As the stabilization process always takes rather long time, the complete transient recursive algorithm often comes with a cost of great computational time in simulating the stabilized materials. In order to accelerate the simulation of the transient-equilibrium transition, a relatively large time step is needed. With this consideration, the robustness of the recursive scheme with the increased time increment is investigated by comparing the
simulation results with time increment-insensitive Laplace solution. The evolution of the healing rate is studied to demonstrate the stabilization process without applied loads. The decay function is fitted with 5 terms to eliminate the potential error caused by the data-fitting. Compared with the trapezoidal integration method, the recursive algorithm is much more stable with large time increment as shown in Fig. 6.9. That is because the direct trapezoidal integration method accumulates the errors from all the previous steps while the accuracy of the recursive algorithm only depends on the results from the last step. Another merit of the recursive algorithm is the reduction of computational cost, as demonstrated by the comparison of the CPU time of the recursive and the trapezoidal methods with different time step increments in Fig. 6.10. With the reduced time step size, the elapsed time of the trapezoidal solver increases significantly compared with the recursive algorithm. Realizing the advantage and the robustness of the recursive algorithm, a large time increment can be used to reduce computational cost in the FE simulation as long as the Gaussian-Raphson iteration converges. Based on the simulation results, the capabilities of different schemes for characterizing the self-healing behaviors of hydrogels are outlined in Table 6.2, showing that the proposed transient approach is more robust with capability in capturing transient deformation behaviors of hydrogels with affordable computational cost due to the insensitivity to the time increment.

<table>
<thead>
<tr>
<th></th>
<th>Transient</th>
<th>Stable</th>
<th>Laplace</th>
<th>Fitted Trapezoidal</th>
<th>Original Trapezoidal</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stabilization without loading</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>Equilibrium loading</td>
<td>✓</td>
<td>✓</td>
<td>×</td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Transient loading</td>
<td>✓</td>
<td>×</td>
<td>×</td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Time increment insensitivity</td>
<td>✓</td>
<td>N/A</td>
<td>✓</td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Fitting insensitivity</td>
<td>×</td>
<td>N/A</td>
<td>N/A</td>
<td>×</td>
<td>✓</td>
</tr>
</tbody>
</table>
In order to demonstrate the capability of the proposed framework in modeling the complex deformation behaviors of self-healing hydrogels, we also conduct 3D simulations on the pre-cut samples with high stress concentration. The FE model follows the experimental setup by Guo [27], with symmetric simplification as shown in Fig. 6.11(a). The pre-cut sample is first stabilized and then stretched under loading rate of 0.01/s. The total element number is approximately 32,000 and the time step increment is fixed as 0.1s. The transient recursive algorithm is applied in this case study. By contrast,
the trapezoidal integration method leads to excessive data storage which is greater than the allocated memory in Fortran compiler, and hence is not presented. Because the fracture mechanism of the permanent network is not considered in the present research, we only compare the crack-opening profiles with the experimental observations without material rupture. A good convergence performance is observed as shown in Fig. 6.11(b) in the FE simulation, which implies the accuracy of the formulation of material stress-stiffness relations. The simulated profiles are mapped onto the experimental results captured by CCD camera from Guo’s research [27]. The overall opening profiles of the pre-cut agree well with the experimental images under different stretches as shown in Fig. 6.12. The good agreements indicate the accuracy and capability of the established model in the simulation of practical applications with complex geometry and loading conditions.

![Figure 6.11 (a) FE model setup (b) convergence rate of recursive algorithm](image)

Figure 6.11 (a) FE model setup (b) convergence rate of recursive algorithm
6.4 Conclusions

In this study, a FE framework is established with the incorporation of a generalized recursive integration algorithm to simulate both the transient and the stabilized deformation behaviors of self-healing hydrogels. The recursive algorithm is implemented to numerically solve the convolution-like integrals in constitutive models so that the time and history dependent behaviors of self-healing hydrogels can be simulated with affordable computational cost. With the well-defined material stiffness from the complete Newton-Raphson scheme, the highly nonlinear transient deformation is successfully modeled with guaranteed convergence. The accuracy and the robustness of the proposed numerical framework have been demonstrated by comparison with experimental data and existing numerical schemes in the literature. The merits of the current research can be concluded as:

1. Compared with the existing numerical equilibrium models in the literature [27], the proposed numerical framework can be adopted to simulate both the transient and equilibrium deformation behaviors of self-healing hydrogels.
2. The accuracy of the recursive integration algorithm less sensitive to time step increment, which will help to reduce the computational cost for a long-time simulation.

Figure 6.12 Comparison of crack opening profiles from simulation with images from experiments [27].
3. The proposed numerical model exhibits good robustness in simulating complex deformation behaviors, which is guaranteed by following the complete Newton-Raphson scheme.

The formulation in the current work is derived based on adopting the Neo-Hookean model as the constitutive model. Future research may be extended to other material constitutive models and the implementation of the recursive integration algorithm in the FE framework to capture more sophisticated material behaviors. This proposed integration algorithm also paves the way for the global fracture simulation of the self-healing polymeric materials, which however still requires future investigation on the rupture mechanism of the primary polymer network.

References


Chapter 7

7 Conclusions and future work

7.1 Conclusions and main contributions

This thesis focuses on developing a finite element framework with the implementation of different constitutive models of polymers to investigate their deformation and failure behaviors with the consideration of microstructure evolution of polymer networks. This work is expected to provide a general approach and a universal computation platform for characterizing and evaluating the performance of soft and smart material-based devices, which will help to facilitate novel design and optimization of these polymeric materials in engineering applications. To be specific, main contributions of this thesis are summarized as follows:

1. In Chapter 3, a FE framework is established to simulate the viscoelastic behaviors of polymeric materials with the implementation of different forms of constitutive models into the UMAT subroutine in Abaqus. General forms of stress tensors and material stiffness tensors are derived that are applicable for constitutive relations involving either strain invariants or principal stretches. Meanwhile, the intrinsic deformation-dependent material viscosity is also incorporated into the FE framework based on polymer dynamics.

2. In Chapter 4, some common instability and bifurcation modes of one typical stimuli-response polymeric material, i.e., dielectric elastomers VHB 4910, are numerically investigated by the incorporation of the nonlinear field theory and the micro-macro constitutive model in UEL subroutine. The effect of material viscosity and the loading conditions on different failure modes are identified. The established FE model can also be expanded to characterize other bifurcation modes for different stimuli-responsive polymeric materials.

3. In Chapter 5, the phase field method is incorporated into the FE framework with staggered algorithm to simulate the fracture behavior of one typical polymeric
material VHB 4905. The material viscosity is formulated to be both deformation and damage dependent during finite deformation. The rate-insensitive fracture behaviors observed in experiments for highly entangled polymeric materials are captured with the proposed free energy degradation scheme.

4. In Chapter 6, the breaking-healing mechanism of self-healing hydrogels is incorporated into the proposed FE framework. A generalized recursive integration algorithm is proposed to numerically solve the convolution-like integrals in the history-dependent constitutive model with reduced computational cost. With the well-defined material stiffness from the complete Newton-Raphson scheme based on the recursive algorithm, the highly nonlinear transient deformation is successfully modeled with guaranteed convergence.

The accuracy and robustness of the proposed numerical models and theories are validated by comparison with experimental data and other existing models in literature. Based on the modeling work and simulation results, some concluding remarks are outlined as below:

1. Different constitutive relations involving either strain invariants or principal stretches are discretized in a generalized form within which only certain intermediate variables need to be specified.

2. The simulation results on VHB 4910 show that material viscosity delays the electromechanical instability and buckling behaviors, while for wrinkling and crumpling where the equilibrium voltage-induced stresses play a dominating role, the delay effect of the material viscosity can be neglected.

3. The rationale for the rate-insensitive fracture behavior of viscoelastic elastomers can be justified by the fact that elastic deformation energy of the crosslinked polymer network is the sole contribution to the global fracture of viscoelastic elastomers. With the proposed free energy degradation scheme, viscoelastic deformation and rate-insensitive fracture behavior of elastomers are spontaneously characterized.
4. With the accumulation of material damage, the disentanglement of polymer chains loosens the topology constraints on the reptation of the confined polymer chain gradually. Thus, it is reasonable to formulate the material viscosity of polymeric materials to be deformation-dependent and damage-dependent.

5. The history-dependent constitutive behaviors of hydrogels are numerically characterized with the proposed iterative integration algorithm with guaranteed convergence.

7.2 Future work

This thesis has conducted a comprehensive numerical study on investigating the constitutive behaviors and some common failure modes of polymeric materials. Despite those contributions, there are still some limitations of the thesis. First of all, only the electromechanical coupled deformation and failure behaviors are investigated by the proposed FE framework. Some other multiphysics coupled behaviors such as temperature-, solvent concentration-, pH-, and light-dependent failure behaviors need to be further investigated. Meanwhile, since Chapter 6 mainly focused on the micro-damage evolution of the sacrificial bonds in the self-healing hydrogels, the damage mechanism of the primary bonds as well as the global fracture behaviors still await to be investigated. Here are some suggestions for the future work:

1. The numerical simulation paves the way for better design of polymeric material-based devices. It can be utilized for topology optimization of polymeric materials to improve the desired performance of smart and soft material-based devices, or for the development of metamaterials.

2. This thesis mainly concentrates on the numerical simulation on some quasi-static behaviors of polymeric materials. The numerical framework can also be extended to include mass matrix and damping matrix to characterize the dynamic deformation behaviors such as wave propagation in the polymeric medium.
3. As fatigue also plays a significant role in affecting the performance of polymer structures when subjected to cyclic loading, it is also essential to conduct theoretical and numerical simulation on investigating and predicting their fatigue life.

4. Due to the complexity of microstructures of self-healing double-network polymers, the mechanisms of fracture behaviors also need further elucidation. Future theoretical and numerical study may be pursued based on continuum mechanics or statistical mechanics.
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