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A 3D finite strain viscoelastic constitutive model for thermally induced shape memory polymers based on energy decomposition

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ABSTRACT

A 3D finite strain viscoelastic constitutive model for thermally induced shape memory polymers (SMPs) is proposed in this work. The thermodynamic system of SMPs in the shape memory cycle is decomposed into thermal and mechanical parts, and the two parts are described separately. Then the constitutive equation is derived based on the restrictions of the second law of thermodynamics through a decomposing of the thermodynamic free energy. The concept of phase transition is introduced to greatly facilitate the determination of material parameters, which makes the model easier for application. Three sets of experimental data in the literature are used to validate the proposed model. The simulation results have an excellent consistency with experiment results, which demonstrates the effectiveness of the proposed model in predicting the shape memory effect (SME) of SMPs.

1. Introduction

Shape memory polymers (SMPs) are a class of smart materials which have the ability to recover their original shape from a deformed state under specific stimulus, such as temperature, light, humidity or magnetic field (Liu et al., 2017; Ranganatha Swamy et al., 2017; Sessini et al., 2018; Sodhi et al., 2015). The property is referred as shape memory effect (SME). Compared with shape memory alloys and shape memory ceramics, SMPs possess the advantages of large deformation, low energy consumption for shape programming, low cost, low density, potential biocompatibility, biodegradability and excellent manufacturability (Hager et al., 2015; Leng et al., 2009). Due to these properties, SMPs have a great deal of potential applications in aerospace deployment structures, biomedical devices, smart textiles, self-healing composite systems, optical reflectors, morphing skins, automobile actuators, etc. (Lee et al., 2014; Leng et al., 2011, 2015; Li et al., 2012; Lina and Alison, 2014; Marshall et al., 2014).

SMPs with SME caused by a change in temperature are called thermally induced SMPs, which are the most widely used. SME in SMPs is not an intrinsic material property, but a combination of the polymer structure and morphology together with an application process. The realize of SME in thermally induced SMPs needs a shape memory cycle with temperature variation, which generally consists of four steps: loading at high temperature, cooling with the strain maintained, unloading at low temperature, and heating under free constrained or fully constrained. The increasing applications of SMPs have motivated the development of their constitutive models. Generally, the developed constitutive models can be divided into two categories: models based on the viscoelasticity theory and models based on the concept of phase transition (Nguyen, 2013).

Viscoelastic constitutive models have been developed based on the essentially thermodynamic properties of polymers. Tobushi et al. (1997) developed a 1D model based on the combination of rheology elements early. Nguyen et al. (2008) developed a 3D finite

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strain viscoelastic constitutive model which considered the evolution of polymer structure with temperature and its yield behavior. [Srivastava et al. \(2010\)](#) proposed a 3D finite strain viscoelastic constitutive model considering thermo-mechanically coupled effect and conducted a finite element simulation for the shape recovery process of a stent. [Xiao et al. \(2013\)](#) extended the work of [Nguyen et al. \(2008\)](#) by incorporating multiple structure relaxation and stress relaxation processes and developed a model representing the glass transition process of SMPs, which improved the prediction ability of the model. In order to represent the glass transition, there are also other approaches in literature which are based on temperature-dependent viscosities ([Christensen, 1985](#); [Dyre, 2006](#); [Lion et al., 2010, 2017](#); [Schmelzer, 2012](#)). [Li et al. \(2017\)](#) developed a 3D model considering different viscoelastic properties of SMPs at high and low temperatures based on the multiplicative decompositions of the deformation gradient.

The phase transition models, on the other hand, describe SMPs as a mixture of two phases: rubbery phase for high temperature and glassy phase for low temperature. SME is realized through the transformation between the two phases. [Liu et al. \(2006\)](#) developed the first phase transition-based model within the small strain regime and the SME was modeled through the evolution of an internal state variable named storage strain. Based on the work of [Liu et al. \(2006\)](#), [Baghani et al. \(2012\)](#) proposed a 3D phase transition-based constitutive model considering the time-dependent material behavior with the introduction of viscous strains. Then, [Baghani et al. \(2014\)](#) extended the model into the large strain regime. [Qi et al. \(2008\)](#) developed a 3D finite strain phase transition-based model, which considered the yield behavior and the post-yield strain softening. Considering the transition of thermodynamic energy between the rubbery phase and glassy phase, [Reese et al. \(2010\)](#) proposed a 3D finite strain model and simulated the shape recovery response of stent structures. Based on the work of [Reese et al. \(2010\)](#), [Boatti et al. \(2016\)](#) developed a 3D finite strain phase transition-based model incorporating multiple internal state variables, which can describe the imperfect shape fixity and incomplete shape recovery phenomena in SMPs. By decomposing the strains of the rubbery phase and glassy phase into multiple parts and considering the viscoplastic and time-dependent material properties, [Park et al. \(2016\)](#) proposed a 3D finite strain phase transition-based constitutive model.

Although the above-mentioned models have achieved great success, their inadequacies cannot be ignored. Usually, the viscoelastic constitutive models realize SME through reasonably describing the variation of thermodynamic properties of SMPs with temperature and time. But they usually contain many parameters because of the complicated material behaviors in the shape memory cycle, which makes the practical application of the models difficult. On the other hand, the phase transition-based constitutive models are based on the phenomenon of drastic variation of thermodynamic properties of SMPs in the shape memory cycle and always contain parameters that have no physical significance, which makes the models can not reflect the physical nature of SME. Under such circumstances, a reasonable model with clear physical meaning which can represent the comprehensive behaviors of SMPs well and easy material parameter determination is needed. Here, a novel 3D viscoelastic constitutive model within the framework of finite strain is proposed. The model is based on the decomposition of thermodynamic energy in the shape memory cycle and the introduction of the phase transition concept, which avoid the need to consider complex structure relaxation and stress relaxation mechanisms at the same time in most viscoelastic models and the definition of virtual deformation modes in most phase-transition models. This greatly facilitates the material parameter determination process.

The paper is organized as follows. In Section 2, a 3D finite strain viscoelastic constitutive model is developed based on the consideration of thermodynamic energy in the shape memory cycle and the introduction of phase transition concept. The specific form of the proposed model in uniaxial loading case is given in Section 3. In Section 4, verification of the model is conducted using three testing examples. Finally, some conclusions are given in Section 5.

2. Model development

2.1. General consideration of the constitutive model

SMPs are a special class of polymers, so they have the essential viscoelastic material behaviors possessed by polymers. In the classic shape memory cycle for thermally induced SMPs, the viscoelastic material behaviors vary with temperature. At high temperature, SMPs approach to hyperelastic materials. At low temperature, SMPs approach to viscoplastic materials. The mechanism underpinning the variation of material behaviors is the drastic change of molecular chain mobility with temperature. And the macroscopic measure of molecular chain mobility is material viscosity, which can be represented by the rheological dashpot element. So, a standard linear viscoelastic model described by a spring element in parallel with a Maxwell element which consists of a spring

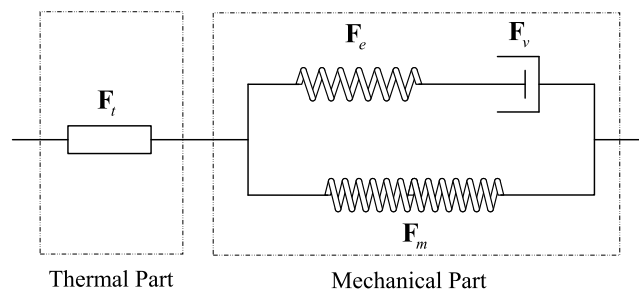


Fig. 1. Rheological representation of the proposed viscoelastic model.

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