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A finite deformation thermomechanical constitutive model for triple shape polymeric composites based on dual thermal transitions



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ABSTRACT

Shape memory polymers (SMPs) have gained strong research interests recently due to their mechanical action that exploits their capability to fix temporary shapes and recover their permanent shape in response to an environmental stimulus such as heat, electricity, irradiation, moisture or magnetic field, among others. Along with interests in conventional "dual-shape" SMPs that can recover from one temporary shape to the permanent shape, multi-shape SMPs that can fix more than one temporary shapes and recover sequentially from one temporary shape to another and eventually to the permanent shape, have started to attract increasing attention. Two approaches have been used to achieve multi-shape shape memory effects (m-SMEs). The first approach uses polymers with a wide thermal transition temperature whilst the second method employs multiple thermal transition temperatures, most notably, uses two distinct thermal transition temperatures to obtain triple-shape memory effects (t-SMEs). Recently, one of the authors' group reported a triple-shape polymeric composite (TSPC), which is composed of an amorphous SMP matrix (epoxy), providing the system the rubber-glass transition to fix one temporary shape, and an interpenetrating crystallizable fiber network (PCL) providing the system the melt-crystal transition to fix the other temporary shape. A one-dimensional (1D) material model developed by the authors revealed the underlying shape memory mechanism of shape memory behaviors due to dual thermal transitions. In this paper, a three-dimension (3D) finite deformation thermomechanical constitutive model is presented to enable the simulations of t-SME under more complicated deformation conditions. Simple experiments, such as uniaxial tensions, thermal expansions and stress relaxation tests were carried out to identify parameters used in the model. Using an implemented user material subroutine (UMAT), the constitutive model successfully reproduced different types of shape memory behaviors exhibited in experiments designed for shape memory behaviors. Stress distribution analyses were performed to analyze the stress distribution during those different shape memory behaviors. The model was also able to simulate complicated applications, such as a twisted sheet and a folded stick, to demonstrate t-SME.

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1. Introduction

Shape memory polymers (SMPs) are a class of smart materials capable of fixing their temporary shape and recovering to their permanent shape in response to an environmental stimulus such as heat (Lendlein and Kelch, 2002, 2005; Liu et al., 2007; Mather et al., 2009; Xie, 2011), light (Jiang et al., 2006; Koerner et al., 2004; Lendlein et al., 2005; Li et al., 2003; Long et al., 2009, 2010b, 2011; Scott et al., 2005, 2006), moisture (Huang et al., 2005,

2005), magnetic field (Mohr et al., 2006), among others. SMPs have promising applications such as microsystem actuation components, active surface patterns, biomedical devices, aerospace deployable structures, and morphing structures. (Davis et al., 2011; Lendlein and Kelch, 2002, 2005; Liu et al., 2004, 2006; Ryu et al., 2012; Tobushi et al., 1996a; Wang et al., 2011; Yakacki et al., 2007).

For most previously developed thermally activated SMPs, a typical shape memory (SM) cycle involves two shapes: one is the permanent shape and the other one is the temporary shape (or programmed shape). This kind of SMPs is often referred to as "dual-shape" SMPs. SMPs can also be "multi-shape". There are two approaches to achieve multi-shape memory behavior. In the

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first approach, the SMP has a very wide temperature range of thermal transition. Recently, Xie (2010) reported that a thermo-plastic SMP, perfluorosulphonic acid ionomer (PFSA), which has a very broad thermal transition temperature range from 55 to 130 °C, showed multi-shape SM effect if the temperature was increased in a staggered manner during free recovery. The second approach is to use multiple thermal transitions, most notably, to use two distinct transitions to obtain the triple-shape memory effects (t-SMEs). In the t-SME, the SMP is capable of fixing two temporary shapes and recovering sequentially from one temporary shape to the other, and ultimately to the permanent shape (Bellin et al., 2006, 2007; Luo and Mather, 2010; Xie et al., 2009). Several methods of achieving the t-SME were reported. For example, Bellin et al. (2006, 2007) used polymer networks consisting of two microscopic segments with two separated transitions. Xie et al. (2009) developed a macroscopic bilaver crosslinked polymer structures with two well separated phase transitions to achieve the t-SME.

Recently, based on the fabrication of shape memory elastomer composites (SMECs) (Luo and Mather, 2009), Luo and Mather (2010) introduced a new and broadly applicable method for designing and fabricating triple-shape polymeric composites (TSPCs) with well controlled properties. In the TSPC system, an amorphous SMP (epoxy with $T_g \approx 20-40$ °C, T_g , the glass transition temperature) works as a matrix providing overall elasticity and fixes one temporary shape using the glass transition; a crystallizable polymer (PCL with $T_m \sim 50$ °C, T_m , the melting temperature) interpenetrating the epoxy matrix is used as fiber network and fixes the other temporary shape using the melt-crystal transition. One advantage of this approach is its fabrication flexibility, since one can tune the functional component separately to optimize material properties (Luo and Mather, 2010). A triple-shape memory cycle of TSPC requires eight thermomechanical loading steps (Fig. 1). In Step 1 (S1), the material is deformed from L_0 to L_1 at a high temperature T_H , higher than the two thermal transition temperatures ($T_{Trans I}$ and $T_{Trans II}$). In Step 2 (S2), the temperature is cooled down to T_{L1} ($T_{Trans II} < T_{L1} < T_{Trans I}$), while maintaining the load. In Step 3 (S3), the external load is suddenly removed and the material fixes the first temporary shape (temporary shape I) at T_{l1} . In Step 4 (S4), the sample is deformed again at T_{l1} . (In practice, the loading at S4 is not necessary to have the same direction with the loading at S1.) In Step 5 (S5), the temperature is decreased to T_{12} ($T_{12} < T_{Transll}$) while keeping the external load applied in S4. In Step 6 (S6), after a sudden removal of the external load, the second temporary shape (temporary shape II) is fixed at T_{12} . In Step 7 (S7), once the temperature is elevated to T_{L1} , the material recovers into its first temporary shape. In Step 8 (S8), the permanent shape is recovered by heating back to T_H .

Along with the fast development of SMPs, constitutive models also have been developed. In amorphous SMPs, where the SM effect is due to the glass transition, modeling approaches include the early model by Tobushi et al. (1996b), the continuum finite deformation thermoviscoelastic model by Nguyen et al. (2008), the finite three dimension phase based model by Qi et al. (2008), the thermo-mechanically coupled theories for large deformations of amorphous polymers by Ames et al. (2009), Anand et al. (2009), Srivastava et al. (2010a,b), the finite strain 3D thermoviscoelastic constitutive model by Diani et al. (2006), the modified standard linear solid model with Kohlrausch-Williams-Watts (KWW) stretched exponential function by Hermiller et al. (2011), and the recent three dimensional (3D) finite deformation constitutive model with a multi-branch modeling approach to represent nonequilibrium process during the glass transition by Westbrook et al. (2011a). In semicrystalline SMPs, Barot and Rao developed a constitutive model for crystallizable shape memory polymers using the notion of multiple natural configurations (Barot and Rao, 2006). Westbrook et al. successfully applied the phase-based modeling approach to the one-way and two-way SM effects in semicrystalline (Westbrook et al., 2010b). Recently, Ge et al. developed a 3D thermomechanical constitutive model for SMECs, which consists of an elastomeric matrix and crystallizable fiber networks (Ge et al., 2012). In that model, the SMEC is developed by treating matrix and fiber network as a homogenized system of multiple phases, and the fiber networks are taken to be an aggregate of melt and crystalline regions. It also gives an evolution rule for crystallization and melting from existing theories (Ge et al., 2012).

The authors have recently reported a 1D thermomechanical model to explain the underlying shape memory mechanism of t-SMP (Ge et al., 2013). In this paper, we formulate a 3D finite deformation thermomechanical constitutive model for the TSPCs. The model combines the multi-branch modeling approach for viscoelasticity of amorphous SMPs (the matrix), and the constitutive model with different deformed crystalline phases for the shape memory behavior of the crystallizable SMP (the fiber network) to describe the t-SME. For the matrix, the time-temperature superposition principle is used to describe glass transition; for the fiber network, the assumption that newly formed crystalline phases of the fiber network are initially in stress-free state is used to track the kinematics of evolving phases. The rest of the paper is arranged in the following manner: In Section 2, the material is introduced briefly and experimental results including DMA, thermomechanical



Fig. 1. Schematic of a temperature (*T*)-loading (*P*)-length (*L*) plot showing the eight-step thermomechanical cycle to achieve t-SME.

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