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A multi-scale approach to model the curing process in magneto-sensitive polymeric materials

Mokarram Hossain [a], George Chatzigeorgiou [b], Fodil Meraghni [b], Paul Steinmann [a,*]

[a] Chair of Applied Mechanics, University of Erlangen-Nuremberg, Egerlandstr. 5, 91058 Erlangen, Germany
[b] LEM3-UMR 7239 CNRS, Arts et Métiers ParisTech Metz-Lorraine, 4 Rue Augustin Fresnel 57078 Metz, France

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ABSTRACT

We propose a magneto-mechanically coupled multi-scale model for simulating the curing process of polymers. In the case of magneto-sensitive polymers, micron-size ferromagnetic particles are mixed with a liquid polymeric matrix in the uncured stage. The polymer curing process is a complex process that transforms a fluid to a solid with time. To transfer the constituent parameter information from the micro-scale to the macro-scale for a composite magneto-mechanically coupled polymeric material, an extended Mori–Tanaka semi-analytic homogenization procedure is utilized. The stiffness gaining phenomenon as in the case of a curing process is realized by time-dependent material parameters appearing within the composite piezomagnetic material tensors. Moreover, to compute the volume reduction during curing, a magnetic induction dependent shrinkage model is proposed. Several numerical examples show that the model proposed herein can capture major observable phenomena in the curing process of polymers under magneto-mechanically coupled infinitesimal deformations.

1. Introduction and outline

Recently magnetorheological elastomers (MREs) became a new class of smart materials. The basic mechanism in MREs is that under an external magnetic excitation, their mechanical properties can be altered. They are relatively a new group in the realm of so-called smart materials. Due to the magnetically controllable stiffness and damping behavior, they are attractive candidates for various technical applications, e.g. suspension bushing, brakes, clutches, smart springs in dynamic vibration absorber to civil engineering devices such as building vibration isolators (Boczkowska and Awietjan, 2009; Böse et al., 2012; Jolly et al., 1996; Varga et al., 2006; Danas et al., 2012; Kaleta et al., 2011; Zhou, 2003; Chen et al., 2007).

In the curing process of polymers, a viscoelastic fluid transforms into a viscoelastic solid due to a series of chemical reactions. Such reactions result in polymer chains cross-linking to each other and formation of chemical bonds allow the chains to come closer. The packing of chains due to cross-linking will yield a decrease in specific volume which is denoted as the volume or curing shrinkage. For an illustrative review on the constitutive modeling of the curing process of polymers, our previous works, cf. Hossain et al. (2009a,b) can be considered. In developing a cure-dependent small strain constitutive model for a thermosetting polymer, Kiasat (2000) assumed that the formation of new cross-links during curing does not affect the current stress state caused by previously applied strains, i.e. new cross-links form unstrained and stress-free. Several researches agree with this assumption (Hojjati et al., 2004; Gillen, 1988). Lion and co-workers (Liebl et al., 2012; Lion and Höfer, 2007; Johlitz, 2012; Johlitz and Lion, 2013) proposed a phenomenologically-inspired viscoelastic curing model. In order to model the evolution of material parameters during curing, they introduce the so-called intrinsic time concept which is then related to the degree of cure, a key parameter to quantify the state and the completeness of a curing process. For more models on the curing process, see Mahnken (2013) and Heinrich et al. (2013) et al.

It is well established nowadays that composite material's overall behavior depends strongly on the properties of the material constituents and the microscopic geometry, i.e. the volume fraction, shape and orientation of constituents. Homogenization methods, as pioneered by Hill (1963) and Hill and Rice (1972), allow to study the overall mechanical behavior of composites with general and periodic microstructures (Hashin and Shtrikman, 1963; Bensoussan et al., 1978; Mura, 1987; Murat and Tartar, 1995;

* Corresponding author. Tel.: +49 09131 8528501; fax: +49 09131 8528503.
E-mail addresses: mokarram.hossain@ltm.uni-erlangen.de (M. Hossain), georges.chatzigeorgiou@ensam.eu (G. Chatzigeorgiou), fodil.meraghni@ensam.eu (F. Meraghni), paul.steinmann@ltm.uni-erlangen.de (P. Steinmann).

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Kouznetsova et al., 2001; Miehe and Koch, 2002). Homogenization of composites with linear as well as nonlinear constituents has been studied extensively (Suquet, 1987; Guedes and Kikuchi, 1990; Terada and Kikuchi, 1995; Aboudi et al., 2003; Ponte Castañeda, 1996; Smit et al., 1998; Feyel and Chaboche, 2000; Michel et al., 1999; Terada and Kikuchi, 2001; Asada and Ohno, 2007; Miehe, 2002; Yvonnet J et al., 2009) and thorough reviews of different multi-scale approaches are available in the literature (Pindera et al., 2009; Kanouté et al., 2009; Charalambakis, 2010; Geers et al., 2010). Such averaging approaches have also been considered for the magneto-mechanical response of magnetorheological elastomers (Borcea and Bruno, 2001; Yin et al., 2002; Wang et al., 2003; Yin et al., 2006; Ponte Castañeda and Galipeau, 2011; Galipeau and Ponte Castañeda, 2013; Javili et al., 2014; Chatzigeorgiou et al., 2014). In this paper, a modified version of the Mori–Tanaka method is utilized. The differences between the classical mean-field theories (Eshelby dilute approach, Mori–Tanaka and self-consistent) have been extensively discussed in several books (Qu and Cherkaoui, 2006; Mura, 1987). These three averaging homogenization techniques are based on the Eshelby's equivalent inclusion theory (Mura, 1987). In brief: it is commonly established that the Eshelby dilute approach provides reliable estimations only for small particles volume fractions, whereas Mori–Tanaka and self-consistent are acceptable also for moderate particle volume fractions. However, self-consistent is an iterative method even in elasticity, and it is generally and extensively used to describe polycrystalline material structures. Mori–Tanaka does not require iterations in the elasticity problem and is more efficient in the case of matrix-particle type of composites, where the particles can have arbitrary orientation.

Several experimental works demonstrate the formation of isotropic and anisotropic magneto-sensitive polymeric composites during the curing process with or without the application of a magnetic induction (Jolly et al., 1996; Varga et al., 2006; Danas et al., 2012; Kaleta et al., 2011; Zhou, 2003; Chen et al., 2007). However, there is a lack of constitutive modeling that can capture the curing process in the presence of a magneto-mechanically coupled load. Several multi-scale approaches are developed for magneto-sensitive polymers with coupled loads where the constitutive material parameters appearing in a model are non-evolving (Schröder and Keip, 2012; Javili et al., 2014). Moreover, a multi-scale approach is proposed during the curing process in which parameters are considered to be evolving with time (Klinge et al., 2012,). However, in the latter works, only a mechanical load is considered during the curing process. To the authors best knowledge, there is currently no multi-scale approach that can capture homogenized behavior as well as can predict the stiffness gaining process in the presence of a magneto-mechanically coupled load. The simulation of microheterogeneous polymers, especially particle-filled magneto-sensitive composites during the curing process, can be studied using various homogenization techniques. The numerical homogenization approach is especially suitable for the simulation of heterogeneous materials with a highly oscillatory microstructure, cf. Klinge et al. (2012,).

The main framework of the proposed model is within the hypoelastic concept of our previously proposed purely mechanical curing model (Hossain et al., 2009a) which is recently extended to the case of particle-filled magneto-sensitive polymers at finite strains (Hossain et al., 2015a). The earlier works that appeared in Hossain et al. (2009a, 2014), on the one hand, are only for phenomenologically-motivated curing modeling under a purely mechanical load. This is for the case of unfilled polymers. On the other hand, the recent works published in Hossain et al. (2015a,b) consider the magneto-mechanically coupled load. Thereby, the main contribution of Hossain et al. (2015a,b) is a phenomenologically-motivated modeling framework for the curing modeling for magnetizable particle-filled polymers. The constitutive model proposed in the current manuscript transfers the constituent parameter information from the micro-scale to the macro-scale for a magneto-mechanically coupled composite at each time step of the curing evolution. The effective parameters of magnetic particle-filled polymers are coming from composite micromechanics rather than taking a continuum parameter set as in the previous two contributions. As a first step in the multi-scale modeling during the curing process, we herein propose a micromechanical approach for small magneto-mechanically coupled deformations where a time-dependence of the mechanical parameters appearing in the constitutive relation is considered. The curing phenomenon is a highly temperature-sensitive and exothermic reaction process. However, for simplicity, we develop the cure-dependent magneto-mechanical coupled model for the case of isothermal processes.

Section 2 discusses the main mathematical foundation that leads to a hypoelastic type constitutive relation for the polymer curing process in the presence of a magneto-mechanically coupled load. In Section 3, a magnetic load dependent curing shrinkage model is proposed where the duration and magnitude of the load is taken into account while the evolution of relevant material parameters is expressed in Section 4. The main mathematical framework to transfer the micro-scale information to the macro-scale is described in Section 5 while the corresponding numerical discretization procedures are elaborated in Section 6. Several numerical examples by pure mechanical, pure magnetic as well as magneto-mechanically coupled loads are presented in Section 7 along with a few illustrative examples for the shrinkage-induced stress generation for a magnetic load dependent curing process.

2. Modeling curing in nonlinear magneto-elasticity

In polymer curing processes, successive chemical reactions yield a cross-linked structure from an initial solution of monomers. This phase transition is analogous to the addition of more and more springs to the already-formed network. According to the literature (Kiasat, 2000; Gillen, 1988), the formation of a new cross-link is unstrained and stress-free. It means that a curing material does not change its state of stress as resulted from previous deformations – even though its material properties continue to evolve. Keeping the above mentioned physical fact in mind, a magneto-elastic coupled energy potential for the case of isothermal curing processes is proposed in the form of a convolution integral as

$$\begin{aligned}\Phi(\boldsymbol{\varepsilon},\mathbb{b},t) = \frac{1}{2}\int_0^t \big[\boldsymbol{\mathcal{A}}'(\tau) : [\boldsymbol{\varepsilon}(t)-\boldsymbol{\varepsilon}(\tau)]\big] \\ : [\boldsymbol{\varepsilon}(t)-\boldsymbol{\varepsilon}(\tau)]d\tau + \frac{1}{2}\int_0^t [\boldsymbol{\mathcal{K}}'(\tau)\cdot[\mathbb{b}(t)-\mathbb{b}(\tau)]] \\ \cdot [\mathbb{b}(t)-\mathbb{b}(\tau)]d\tau + \int_0^t [\boldsymbol{\mathcal{C}}'(\tau)\cdot[\mathbb{b}(t)-\mathbb{b}(\tau)]] \\ : [\boldsymbol{\varepsilon}(t)-\boldsymbol{\varepsilon}(\tau)]d\tau \end{aligned} \tag{1}$$

where $\boldsymbol{\mathcal{A}}'(\tau) = d\boldsymbol{\mathcal{A}}(\tau)/d\tau$, $\boldsymbol{\mathcal{K}}'(\tau) = d\boldsymbol{\mathcal{K}}(\tau)/d\tau$ and $\boldsymbol{\mathcal{C}}'(\tau) = d\boldsymbol{\mathcal{C}}(\tau)/d\tau$. In Eq. (1) $\boldsymbol{\varepsilon}$ is the infinitesimal strain, $\mathbb{b}$ is the magnetic induction vector and t is the curing time. A finite strain version of the potential function proposed in Eq. (1) and its corresponding derivations are given in Hossain et al. (2015a). Note that $\boldsymbol{\mathcal{A}}$ is the fourth order mechanical stiffness tensor, $\boldsymbol{\mathcal{K}}$ is the second order magnetic permeability tensor, and $\boldsymbol{\mathcal{C}}$ is the third order coupled magnetomechanical tensor, where the material parameters appearing within the tensors are generally time-dependent. Specific forms of these tensors are given in Section 7. The second law of thermodynamics in the form

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