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# A phase-field model for systems with coupled large deformation and mass transport

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ABSTRACT

Phase-field model is a powerful tool for studying the microstructure evolution of materials. This paper seeks to introduce phase-field modeling to the field of soft materials, especially for studying polymeric gels. A general framework for the field theory of coupled large deformation and mass transport is established, and two specific models of diffuse interface are proposed. The ideal liquid-like interface has a deformation-independent energy and gives rise to a constant surface tension, and a non-ideal interface would result in a strain-dependent surface stress. Either model gives a stress field consistent with the effect of interface line force. The field theory is implemented into a finite-element code, and several numerical examples are calculated with representative material models in which deformation is weakly or strongly coupled with mass transport. The numerical models demonstrate the versatility of the phase-field methodology, and reveal some interesting phenomena due to the coupling. For example, the composition of a separated phase is significantly affected by the kinematic constraint, and varies during coarsening.


## 1. Introduction

Phase field is a versatile tool for modeling phase transition and morphological and microstructure evolution in condensed matter, and has undergone continuous development for various applications (Boettinger et al., 2002; Chen, 2002; Emmerich, 2008). In this paper, we seek to advance the theory to incorporate concurrent finite deformation and mass transportation. Besides the classic problem of diffusional solid-state transition in alloys (Cahn, 1961; Onuki, 1989a; Onuki and Furukawa, 2001), a typical problem requiring such considerations is the volumetric phase separation of polymeric gels (Shibayama and Tanaka, 1993; Matsuo and Tanaka, 1988; Shibayama and Nagai, 1999; Hu et al., 2001). A swollen gel consists of a crosslinked polymer network and a solvent. Under certain conditions, a homogeneous gel may separate into shrunk and swollen phases characterized by the drastic differences in both solvent concentration and the stretch of polymer network (Tanaka, 1979; Hochberg and Tanaka, 1979). During the phase separation of a gel, the growth of one phase is enabled by absorbing swelling liquid from its neighbors, as well as spatially replacing other phases (Shibayama and Tanaka, 1993). Understanding the physics of this model system will provide further insights towards the role of elasticity in pattern formation in soft materials during more complicate processes, such as gelation (Bansil et al., 1992; Matsuo et al., 1993; Hong and Chou, 2000), transient gel formation during viscoelastic phase separation (Tanaka, 2000), phase dynamics in polymer-stabilized liquid crystals (Lapena et al., 1999), vesicle dynamics (Du et al., 2004; Biben et al., 2005), and even the growth of biological tissues (Lappa, 2004). Unfortunately, most established theoretical analysis are limited to the small-strain region

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(Cahn, 1961; Tanaka et al., 1985; Onuki, 1988, 1989a, 1989b; Onuki and Furukawa 2001; Garcke, 2003) or homogeneous deformation (Hirotsu and Onuki 1989; Cai and Suo 2011), whereas the inhomogeneous stress plays an dominant role in the morphology (Nishimori and Onuki, 1990; Onuki and Puri, 1999; Onuki and Furukawa, 2001) and the kinetics of pattern growth (Tanaka, 2000), and the typical strain involved is several hundred percent, for soft materials in particular. A few efforts have been made to consider finite deformation and its coupling with swelling, and interesting structures similar to experimental observation have been revealed (Onuki and Puri, 1999; Uchida, 2002; Zhou et al., 2010). The effect of introducing both kinematic and physical nonlinearity is yet to be discussed, and the detailed phase separation processes in these systems are far from being fully understood.

Phase-field models are distinguished by two principle characteristics: a continuous phase field to differentiate domains of dissimilar microstructures (Landau and Khalatikow, 1965), and a diffuse interface across which physical properties smoothly transition from one phase to another (Van der Waals, 1894). Depending on the nature of the problem, the phase field could be an auxiliary variable (Collins and Levine, 1985; Karma et al., 2001), or a physical parameter (Cahn and Hilliard, 1958; Landau and Khalatikow, 1965; Cahn and Allen, 1977). In either case, the diffuse interface is associated with an excess free energy, which is often written as a function of the spatial gradient of the phase field. However, in a large-deformation context, it is unclear whether the gradient should be taken with respect to the deformed configuration or the reference configuration. It has recently been shown that by using the gradient in the deformed configuration, the interface energy function could recover the behavior of surface tension (Levitas and Samani, 2011). As will be shown in the current paper, such an interface model represents a liquid-like interface, on which the molecules are capable of rearranging themselves. The corresponding interface energy per unit current area is independent of the state of deformation. More generally, the interface energy may vary with deformation. For example, for a solid-like interface that preserves its atomic structure during deformation, the surface tension may be a function of strain, and is often referred to as the surface stress (Shuttleworth, 1950; Gurtin et al., 1998). Although the effect of surface tension/stress on deformation or stress field could often be neglected on macroscopic structures, it is of importance to nanomaterials (Levitas and Samani, 2011) in which the interfacial layer occupies a significant portion of the total volume, and at a much larger length scale to soft materials which deform under very low forces. In this work, we aim at formulating a general framework for the phase-field model of coupled deformation and mass transport, and developing interface models which describe the behaviors of liquid-like and solid-like interfaces.

Thermodynamic theories of coupled mass transport and elastic deformation date back at least to Gibbs (1878), who formulated the equilibrium theory of a solid that absorbs liquid. The more general non-equilibrium theory that describes the transport processes is known as poroelasticity (Biot, 1941; Rice and Cleary, 1976). Specifically for polymeric gels, existing models include nonlinear poroelasticity (e.g. Baek and Srinivasa, 2004; Hong et al., 2008; Duda et al., 2010; Chester and Anand, 2010), and multi-phasic models (e.g. Doi, 2009; Rajagopal, 2003). However, with these continuum field theories alone, it is difficult to deal with problems like phase separation, in which a boundary between domains could spontaneously emerge, migrate, and vanish. In this paper, we extend the capability of nonlinear poroelasticity by combining it with a phase-field model. In Section 2, by introducing the gradient energy terms, the basic non-equilibrium thermodynamic theory is modified to incorporation the interface contribution. Two interface models and their coupling with elastic deformation is introduced in Section 3. The theory is then specialized with a detailed material model and tested with finite-element calculations in Section 4. It is shown both analytically and numerically that the liquid-like interface model gives a thermodynamically consistent contribution to the stress field. The coupling behavior of elastic deformation and mass-transport-enabled phase separation is illustrated through several numerical examples. The effect of strong coupling and elastic misfit between separated phases is demonstrated through a numerical model by assuming the molecular incompressibility. The effect of strain-dependent interface stress is exemplified through a model with stress-induced interface anisotropy.

## 2. Non-equilibrium thermodynamics

The microstructure evolution of a multicomponent material system usually involves mass transportation and structure reconfiguration. To track the deformation, we introduce the reference by assuming that at least some material particles do not change relative positions and the aggregate remains to be a continuum during the evolution process. An example of the background continuum frame is the polymer network of a permanently crosslinked gel. Imagine attaching to these material particles a set of markers, with coordinates $\mathbf{X}$ in the reference state. We will associate the properties of a material particle to the local marker, by writing the physical fields as functions of $\mathbf{X}$ and time $t$. The coordinates of a material particle at time $t$, for example, is written as $\mathbf{x}(\mathbf{X},t)$. The field of deformation gradient,

$$F_{iK}(\mathbf{X},\mathbf{t}) = \frac{\partial x_i(\mathbf{X},\mathrm{t})}{\partial X_K}, \tag{1}$$

measures the deformation of the continuum part of the material.

Just as all phase-field models in a small-deformation context, we differentiate dissimilar phases using a continuous phase field, and write it also as a function of the reference coordinates and time, $C(\mathbf{X},t)$. The phase field may be conservative, such as the concentration of a species, or non-conservative, such the internal variable characterizing the state of damage. While the general framework is applicable to both cases, in the current paper, we will illustrate it through the conservative case, in