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Unconditionally stable, second-order accurate schemes for solid state phase transformations driven by mechano-chemical spinodal decomposition

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Highlights

- We consider a coupled system of the Cahn–Hilliard model and gradient elasticity.
- We develop an accurate time-integration scheme for this phase-field model.
- The key idea is the use of exact Taylor expansions of non-convex energy densities.
- We demonstrate accuracy of the scheme in our numerical examples in three dimensions.

Abstract

We consider solid state phase transformations that are caused by free energy densities with domains of non-convexity in strain-composition space; we refer to the non-convex domains as mechano-chemical spinodals. The non-convexity with respect to composition and strain causes segregation into phases with different crystal structures. We work on an existing model that couples the classical Cahn–Hilliard model with Toupin's theory of gradient elasticity at finite strains. Both systems are represented by fourth-order, nonlinear, partial differential equations. The goal of this work is to develop unconditionally stable, second-order accurate time-integration schemes, motivated by the need to carry out large scale computations of dynamically evolving microstructures in three dimensions. We also introduce *reduced* formulations naturally derived from these proposed schemes for faster computations that are still second-order accurate. Although our method is developed and analyzed here for a specific class of mechano-chemical problems, one can readily apply the same method to develop unconditionally stable, second-order accurate schemes for any problems for which free energy density functions are multivariate polynomials of solution components and component gradients. Apart from an analysis and construction of methods, we present a suite of numerical results that demonstrate the schemes in action.

Keywords: Unconditional stability; Mechano-chemistry; Cahn–Hilliard; Strain gradient elasticity; Spinodal decomposition; Phase-field

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1. Introduction

Many multicomponent solids undergo phase-transformations in which a diffusional redistribution of their different components is coupled with a structural change of the crystallographic unit cell. One example is the phase-transformation of yttria-stabilized zirconia $Zr_{1-x}Y_xO_{2-x/2}$ from cubic at high-Y composition to tetragonal at low-Y if quenched to a low temperature. Another is observed in the spinel structures of Li-ion electrodes when cubic $LiMnO_4$ transforms into tetragonal $Li_2Mn_2O_4$ upon discharging to low voltages. Pure ZrO_2 and TaS_2 are other materials that may be susceptible to such mechano-chemical phase transformations.

The underlying phenomenology can be described by a free energy density function, which is non-convex in its mechanical (strain) and chemical (composition) arguments. Cahn and Hilliard [1] famously introduced the chemical spinodal as the domain in composition space where the free energy density is non-convex, and varies smoothly between local minima that correspond to distinct phases. This notion has recently been extended to the mechano-chemical spinodal, defined as the domain in strain-composition space where the Hessian of the (sufficiently smooth) free energy density function has non-positive eigenvalues [2]; see Fig. 1. If the state of a material point lies within the mechano-chemical spinodal, and specifically if the free energy density is non-convex with respect to composition, the solid will undergo diffusional segregation. Here we concern ourselves with cases in which the two resulting phases have cubic and tetragonal crystal structures, respectively. The cubic structure corresponds to a minimum of the free energy density function in strain-composition space. Furthermore, we adopt the undistorted cubic structure as the reference state for strain. Then, the tetragonal lattice is naturally obtained by the strain relative to the cubic structure. The symmetries of the cubic lattice are split into three identical sub-groups, each of which corresponds to a tetragonal lattice oriented along one of the cubic crystal axes. These three tetragonal variants, however, correspond to different strains relative to the reference cubic lattice. The free energy density function admits three additional minima, each corresponding to the strain that transforms the reference cubic lattice into one of the tetragonal variants. However, the compositions at these additional minima are identical. As the states of material points traverse such a multi-welled free energy density surface in strain-composition space the solid develops a microstructure. The mechano-chemical spinodal is regarded as a domain of instability since small fluctuations in strain and composition tend to grow as the state evolves towards one of the wells in strain-composition space. Stress softening and "uphill" diffusion result, respectively.

A non-convex free energy density function can lead to microstructure as explained above. However, a mathematical model restricted to the above phenomenology leads to ill-posed partial differential equations (PDEs) for elasticity and transport characterized by spurious mesh dependence. The volume fractions of the cubic and tetragonal phases would be set by initial and boundary conditions on the transport problem, and the microstructural pattern of tetragonal variants would depend on mechanical boundary conditions. However, in the absence of intrinsic length scales in the mathematical model, the extent and thickness of interfaces between phases and variants would be determined by the mesh. There is a well-understood physical aspect to this argument, also: The model promotes free energy minimizing microstructures, but incurs no penalty for the strain and composition gradients as these fields fluctuate between one phase or variant and the next. Arbitrarily fine energy minimizing microstructures are therefore admissible—an essentially unphysical result. Mathematical well-posedness and physical realism are restored by extending the free energy density function to include a dependence on gradient fields of strain and composition. The corresponding free energy coefficients introduce intrinsic length scales, and the gradient energies distinguish between microstructures of differing fineness, penalizing those that vary rapidly.

Gradient free energies in classical settings lead to the Cahn–Hilliard equation for mass transport [1], and variants of strain gradient elasticity that represent size effects. Because the transformation strains between the cubic/tetragonal phases and between the tetragonal variants are of finite magnitude in many material systems, we are led to Toupin's theory of nonlinear (finite strain) gradient elasticity [3]. The Cahn–Hilliard equation and Toupin's strain gradient elasticity present fourth-order spatial derivatives in primal strong form, with corresponding weak forms carrying second-order derivatives. Carrying out large scale computations of these dynamically evolving microstructures in three dimensions is challenging; efficient and accurate time-integration algorithms are demanded. Rudraraju and co-workers used the Backward Euler algorithm [2] to solve the same problem that we consider here. However, in that work the authors concerned themselves with introducing the notion of the mechano-chemical spinodal, and exploring the associated physics; not with a development/an analysis of accurate schemes, which is the goal of the present communication.