Investigation on Viscoelastic-Creep Behavior of the Phase-Field Crystal Method

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Abstract. The phase-field crystal (PFC) method is a promising computational model with atomistic resolution and diffusive time-scale. In this study, we investigated the viscoelastic-creep behavior exhibited by the PFC model. We considered a one-dimensional crystal subjected to step changes in pressure and studied the time-dependent strain response from the system. The parametric study shows that the PFC model predicts the materials with higher density to exhibit higher stiffness and lower damping capacity while an increase in temperature results in lower stiffness and damping capacity. These predictions agree with experimental observations and show promising capability of the PFC method to model viscoelastic phenomena.

1. Introduction

Due to rapid increase in computing performance and theoretical development, computational methods are becoming powerful tools in facilitating materials advancement. However, many problems in computational materials science involve length and time scales of many orders of magnitude. This large span of length and time scales is a challenge for atomistic models, such as molecular dynamics (MD) and density functional theory (DFT), in that these models are typically applicable in prohibitively-small length and time regimes. On the other hand, continuum models, such as the phase field (PF) models, naturally reside in phenomenologically-relevant length and time scales, but often suffer from lack of atomistic resolution and self-consistency. In the past decade, the phase-field crystal (PFC) method [1] emerged as a robust tool to address these issues. The appeal of the PFC method lies in its atomistic resolution and diffusive time scale, which give the model advantages over the PF method in terms of accuracy and self-consistency and over the MD and DFT in terms of computational expenses. Due to these advantages, the PFC method has been used to predict various materials properties and model several materials phenomena (see Ref. [2] for comprehensive review in this topic).

Despite various applications of the PFC method, especially in deformation-related phenomena, the viscoelastic behavior exhibited by the model has not been extensively studied. Viscoelasticity is a combination of elasticity and viscosity, and is an important characteristic in vibration-damping materials, noise-controlled applications, and microelectromechanical system (MEMS). Therefore, in this work, we investigated the viscoelastic behavior exhibited by the PFC model. In particular, we considered the viscoelastic-creep behavior of a one-dimensional crystalline solid subjected to step changes in pressure, and analyzed the time-dependent strain

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response of the system. From the parametric study, we found that the PFC method predicts higher-density materials to have higher stiffness and damping capacity. Also, an increase in temperature results in lower stiffness and lower damping capacity. These predictions agree with experimental observations and show promising capability of the PFC method to model viscoelastic phenomena. Nevertheless, further investigation is needed in order to better understand the viscoelstic behavior from the PFC model; future improvements include extension to three-dimensional structures, more extensive parametric study, and investigation into other viscoelastic phenomena.

The paper is organized as follows. In section 2, we review the PFC method and the pressure-controlled dynamic equation recently proposed by Em-Udom and Pisutha-Arnond [3]. In section 3, we describe the simulation and analysis procedures. The simulation results and discussion are presented in section 4. Lastly, we provide a summary of our work in section 5.

2. Phase-Field Crystal Method

In the PFC method, the two central ingredients are the energy expression and evolution equations. For the energy expression, we consider the Helmholtz free energy, F, which can be written in terms of dimensionless quantities as [4]

$$F = \int_V f_v d\vec{r} \equiv \int_V \left(\frac{\rho^4}{4} + \frac{\rho}{2} \left[-\epsilon + \left(1 + \nabla^2 \right)^2 \right] \rho \right) d\vec{r}, \tag{1}$$

where f_v is the free energy density, $\rho \equiv \rho(\vec{r})$ is the atomic number density field, ϵ is the model parameter related to temperature, and V is the volume. This free energy is minimized by a constant density profile (representing a liquid phase) and a periodic density profile (representing a crystalline phase). The latter profile can be written in terms of a Fourier expansion of the form

$$\rho(\vec{r}, t) = \bar{\rho}(t) + \sum_j A_j(t) e^{i\vec{G}_j \cdot \vec{r}} + \text{c.c.}, \tag{2}$$

where $\vec{G}_j$ is the reciprocal lattice vector, A_j is the amplitude of the density wave corresponding to $\vec{G}_j$, $\bar{\rho}$ is the average number density, and c.c. denotes the complex conjugate. The second ingredient of the PFC method, which is the evolution equations, governs the time evolution of the state of the system. In this work, we employ two evolution equations. The first is the Cahn-Hilliard type equation that evolves $\rho(\vec{r}, t)$ through dissipative dynamics and with mass conservation [5]:

$$\frac{\partial \rho}{\partial t} = L_\mu \nabla^2 \mu \equiv L_\mu \nabla^2 \left(\frac{\delta F}{\delta \rho} \right), \tag{3}$$

where $\mu = \delta F / \delta \rho$ is the chemical potential, L_μ is the mass-diffusion coefficient. The second equation is the pressure-controlled dynamic equation recently proposed in Ref. [3] to simulate the deformation of a system under externally-applied pressure $\bar{P}_{\text{ext}}$:

$$\frac{dV}{dt} = L_P \int_V (P - \bar{P}_{\text{ext}}) d\vec{r}, \tag{4}$$

where $P = -f_v + \mu\rho$ is the internal pressure and L_P is the deformation coefficient.

3. Method

In this section, we describe the method to simulate, analyze and study the viscoelastic-creep behavior in the PFC model. To simulate the viscoelastic-creep behavior, we consider a one-dimensional crystalline phase with the density field $\rho(x, t) = \bar{\rho}(t) + \sum_j A_j(t) \cos(G_j x)$, where x

is the spatial coordinate. The system is initially at equilibrium and this state of the system is characterized by three state variables. First is the mass per unit-cell $N_0 = \bar{\rho}_0 L_0$, where L_0 is the initial unit-cell length and $\bar{\rho}_0 \equiv \bar{\rho}(0)$ is the initial average density. The second state variable is the average pressure $\bar{P}_0 = 1/L_0 \int_{L_0} P dx$, and the third is the temperature which is related to the model parameter ϵ. Then, the system is subjected to step changes in the average pressure quantified by $\Delta \bar{P} = \bar{P}_0 - \bar{P}_{\text{ext}}$. From equation (4), the nonzero $\Delta \bar{P}$ will lead to deformation characterized by the change in the unit-cell length $L(t)$ and the strain $e(t) = (L(t) - L_0)/L_0$. During the deformation, we enforce mass conservation by changing the average density according to $\bar{\rho}(t) = N_0/L(t) = \bar{\rho}_0 L_0/L(t)$, and we also allow the system to attain the equilibrium condition in all deformation states by evolving the density profile using equation (3) until a steady state is reached for each value of $L(t)$.

To analyze the viscoelastic-creep behavior, we fit $e(t)$ from the simulations to the solution of a standard linear solid model [6] which is typically used to describe viscoelastic behavior. In particular, we consider a spring-damper system subjected to uniform stress σ as shown in figure 1. The strain response from this system is governed by $de/dt + \frac{E_1 E_2}{\eta(E_1 + E_2)} e = \frac{E_1}{\eta(E_1 + E_2)} \sigma$, where E_1 and E_2 are the spring constants and η is the damping coefficient [6]. The solution to this ordinary differential equation is

$$ e(t) = e_\infty \left[1 - \exp\left(-\frac{t}{\tau} \right) \right], \quad e_\infty = \frac{\sigma}{E_2}, \quad \tau = \eta \left(\frac{E_1 + E_2}{E_1 E_2} \right), \tag{5} $$

where e_∞ and τ are the strain value at $t = \infty$ and the relaxation time, respectively. When equation (5) is fitted to $e(t)$ from the PFC simulations, the values of e_∞ and τ, which characterize the viscoelastic-creep behavior, can be extracted for further study.

To study the viscoelastic-creep behavior exhibited by the PFC model, we investigate the dependence of e_∞ and τ on the PFC model parameters. In this parametric study, we use $\Delta \bar{P} = \pm 0.05 \bar{P}_0$, $\epsilon = [0.1, 0.3]$, and $|\bar{\rho}_0| = [0.15, 0.5]$. The values of $\Delta \bar{P}$ are selected to yield tensile ($\Delta \bar{P} > 0$) and compressive ($\Delta \bar{P} < 0$) deformation. The ranges of ϵ and $|\bar{\rho}_0|$ are selected from the crystalline region in the phase diagram shown in Ref. [5]. Other fixed simulation and model parameters are $L_\mu = 1$, $L_P = 0.5$, $L_0 = 2\pi\sqrt{2}$, Δx (grid spacing) $= L_0/16$. The time steps for equations (3) and (4) are 0.01 and 0.5, respectively.
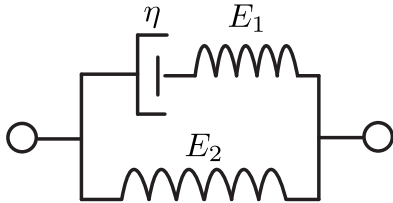

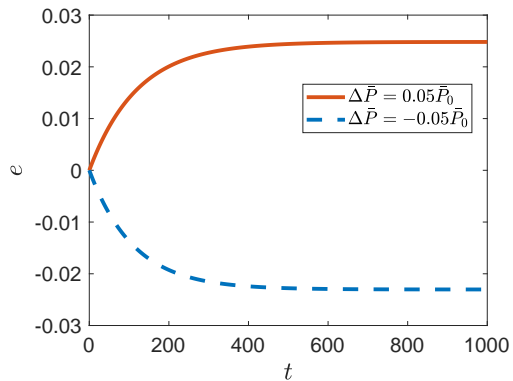**Figure 1.** Schematic illustration of a spring-damper system.

Figure 2. Plot of strain responses from the step changes in the pressure $\Delta \bar{P} = \pm 0.05 \bar{P}_0$. In both simulations, $\epsilon = 0.3$ and $|\bar{\rho}_0| = 0.15$.

4. Results and Discussion

In figure 2, the viscoelastic-creep behavior from the PFC model is shown from the plot of the strain $e(t)$ with $\Delta \bar{P} = \pm 0.05 \bar{P}_0$. Initially, the system is at $e = 0$. After the pressure changes, e increases or decreases exponentially until the convergence value is reached. The positive e indicates tensile deformation which results from the positive $\Delta \bar{P}$; on the other hand, the negative e implies compressive deformation which corresponds to the negative $\Delta \bar{P}$. This time-dependent strain response from a step change in pressure is indicative of the viscoelastic-creep behavior.

In figure 3, the dependence of e_∞ and τ on $|\bar{\rho}_0|$ is shown for the tensile deformation. First, figure 3(a) shows that e_∞ decreases as $|\bar{\rho}_0|$ increases. To interpret the result, we consider the definition of e_∞ from equation (5) which indicates that $e_\infty \propto 1/E_2$. Therefore, the increase in the density, which leads to decreased e_∞, implies an increase in E_2 or the overall stiffness of the system. Second, figure 3(b) shows that τ decreases with increased $|\bar{\rho}_0|$. From the definition of τ in equation (5), the term $(E_1 + E_2)/E_1 E_2$ is not expected to change significantly and, therefore, $\tau \propto \eta$. This proportionality indicates that the increase in the density, which leads to decreased τ, results in lower η or lower damping capacity of the system. Combining the results from figures 3(a) and 3(b), we can see that the PFC model predicts high-density materials to exhibit high stiffness and low damping capacity. This prediction agrees in general with materials such as metals and ceramics [7].
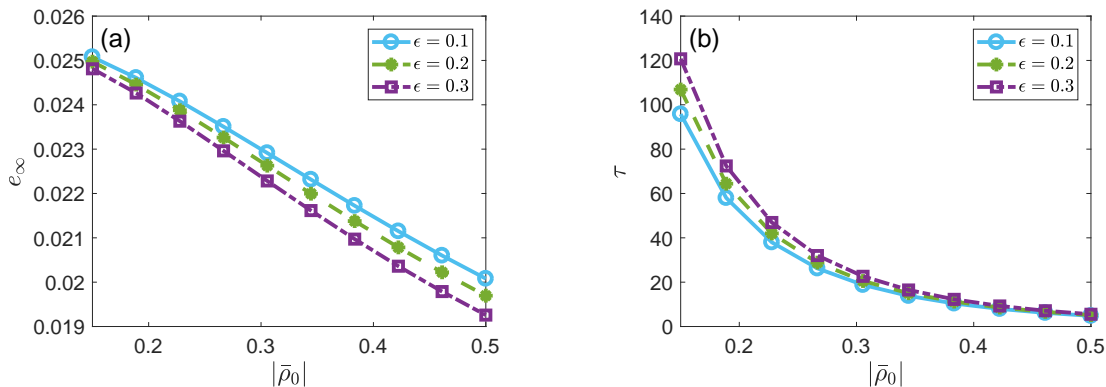

Figure 3. Plots of e_∞ and τ as the functions of $|\bar{\rho}_0|$ for the tensile deformation ($\Delta \bar{P} = 0.05 \bar{P}_0$).

In figure 4, the dependence of e_∞ and τ on ϵ is shown for the tensile deformation. First, figure 4(a) shows that as ϵ decreases, e_∞ increases. We note that ϵ is related to the degree of undercooling from the solidification temperature [1]; therefore, a higher (lower) ϵ value corresponds to lower (higher) temperature. Then, the result from figure 4(a) indicates that increasing temperature (lowering ϵ) leads to higher e_∞ or lower stiffness of the system. Second, figure 4(b) shows that τ decreases with decreasing ϵ; however τ is less affected by ϵ for higher values of $|\bar{\rho}_0|$. This result indicates that, at lower density, increasing temperature (lowering ϵ) leads to lower damping capacity while at higher density, the damping capacity is less affected by temperature. Combining the results from figures 4(a) and 4(b), we find that the PFC model predicts, for lower-density materials, that higher temperature leads to lower stiffness and lower damping capacity. The decrease in materials stiffness with increased temperature is typically observed experimentally. Also, the decrease in damping capacity (faster strain response) at higher temperature agrees with what is generally observed in creep phenomena; in other words, the relaxation time in the creep phenomena generally increases with temperature [6]. Despite the agreement between the predictions from the PFC model and experimental observations, further developments need to be implemented in order to better understand the viscoelastic behavior

from the PFC model; these improvements include extension to three-dimensional structures, more extensive parametric study, and investigation into other viscoelastic phenomena.
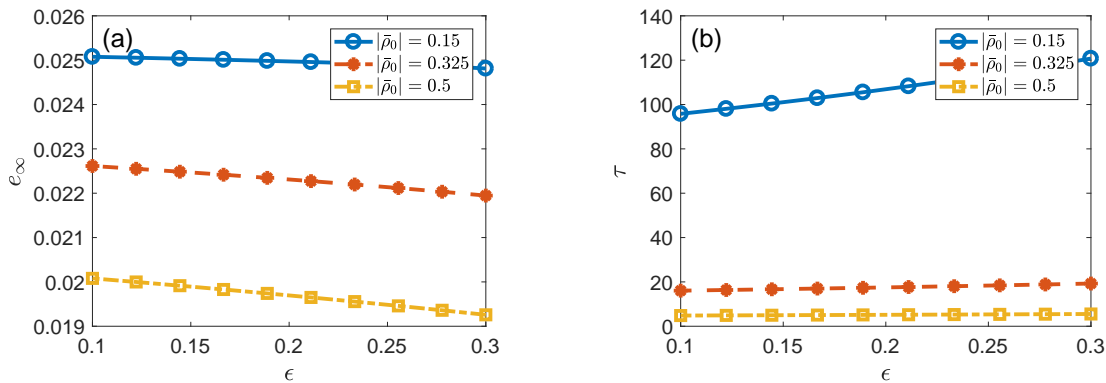

Figure 4. Plots of e_∞ and τ as the functions of ϵ for the tensile deformation ($\Delta \bar{P} = 0.05 \bar{P}_0$).

5. Summary

In this work, we explored the viscoelastic creep behavior exhibited by the PFC model. We considered a one-dimensional crystalline solid subjected to step changes in pressure and the resulting time-dependent strain response was analyzed. From the parametric study, we found that the PFC model predicts higher-density materials to exhibit higher stiffness but lower damping capacity; this prediction agrees in general with materials such as metals and ceramics. Also, increasing temperature leads to lower material stiffness and damping capacity. This prediction agrees with experimental observations where materials typically become less stiff at high temperature and the relaxation time (proportional to the damping capacity) in creep phenomena generally decreases with temperature. The agreement between the PFC predictions and experimental observations shows promising capability of the PFC method to model viscoelastic phenomena. Nevertheless, further investigation is needed in order to better understand the viscoelstic behavior from the PFC model; future improvements include extension to three-dimensional structures, more extensive parametric study, and investigation into other viscoelastic phenomena.

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