Acta Materialia 109 (2016) 286–291

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Full length article

Phase field modelling of rayleigh instabilities in the solid-state

Chaitanya Joshi*, T.A. Abinandanan, Abhik Choudhury

Department of Materials Engineering, Indian Institute of Science, Bangalore, 560012, India

ARTICLE INFO

Article history:
Received 8 December 2015
Received in revised form
28 February 2016
Accepted 1 March 2016
Available online xxx

Keywords:
Rayleigh instability
Phase-field model
Nanopores
Porous membranes

ABSTRACT

We have used a phase field model to study Rayleigh instability driven evolution of a cylindrical pore. The key feature in the model is its ability to incorporate surface diffusion as the mechanism for mass transport. We first benchmark our model with analytical results for growth rates of sinusoidal perturbations imposed on the surface of a cylindrical pore of radius R at early times. We then use the model to predict breakdown of infinite cylindrical pores; the principal finding from our analysis is that time to failure scales as R^4. We have also studied the break-up of closed and open cylindrical pores of finite length; a series of about five spherical pores get pinched off sequentially at the cylinder ends before the middle parts of the pore break up. Compared to the first closure event in an infinite pore, the first pinch-off event in closed and open pores is faster by about 4 times and 25 times, respectively.

1. Introduction

Interfacial phenomena resulting from instabilities has been a source of interest for physicists and material scientists. Their understanding allows one to predict the principal microstructural length scales in the problem, which is often quite important both for the design of better materials as well as newer alloys. A classic instability is that investigated by Lord Rayleigh [1], on thin cylindrical fluid jets causing them to break up into spherical droplets. In a classical study, Nichols and Mullins developed a formalism for studying morphological changes in solids, including solid cylinders, using surface diffusion [2] and volume diffusion [3]. In a later study, Nichols [4] extended this analysis to the case of instabilities in a cylinder of finite length. Theoretical predictions of Nichols and Mullins have been verified in numerous experiments [5]. A 2001 paper by Mullins [6] presents a review of progress made in the theoretical treatment of capillarity induced surface morphologies.

The theory proposed by Nichols and Mullins has been useful for experimentally measuring surface diffusion coefficients [7]. Rayleigh instability is known to play a role in crack healing in ceramics [8–11]. It has also been exploited in nanowires to produce a long chain of nanospheres [12]. It has been conjectured to be the mechanism for break-up of plate-like structures into cylinders [13].

In this paper, we use the phase field modelling technique to study Rayleigh instabilities. Though well known, it is worth repeating the advantages of a phase field approach: Unlike theories and sharp interface models (which are often linearised to simplify the analysis, and therefore, are applicable only to early times), phase field models allow us to study the process right from the onset of instability to the final break-up of a cylindrical pore. Further, they handle pinch-off (i.e., pore closure) events gracefully, and allow us to study post-pinch-off behaviour (e.g., a second and subsequent pinch off events) as well. Yet another advantage is that the phase field model presented here can be easily extended to study porous, polycrystalline membranes, of the kind investigated by Choudhury et al. [14].

In the next section, we present a recap of the results of Nichols and Mullins for an infinite cylindrical pore. In Section 3, we formulate our phase field model which incorporates surface diffusion as a dominant mechanism for atomic transport (Our model is similar to those in Refs. [15–17]). Section 4 starts with a validation of our model through a critical comparison with analytical results of Nichols and Mullins, followed by a study of infinite and finite cylindrical pores of different radii.

2. Theory

In this section, we summarize the theory of Nichols and Mullins [1,3] for the onset of Rayleigh instability in an infinite cylinder of radius R. The driving force for the growth in amplitude of a sinusoidal perturbation of wavelength λ is the reduction in the surface area; however, this reduction is possible only for perturbations beyond a critical wavelength of $\lambda > 2\pi R$.

* Corresponding author.
E-mail address: chaitanya.4939@gmail.com (C. Joshi).

http://dx.doi.org/10.1016/j.actamat.2016.03.005
1359-6454/

While there may be contributions from viscous flow, evaporation-condensation, or volume diffusion, we focus here on surface diffusion as the dominant mass transport mechanism, as is appropriate for the evolution of a pore surface. In this case, the evolution of an initially cylindrical surface is given by Eq. (1) [3] in cylindrical co-ordinates with r as the radial distance from the z axis, the axis of the cylinder (it is assumed that there is no angular dependence). This is a linearised version of the general equation derived by Nichols and Mullins, and therefore, it is valid only for early stages.

$$\frac{\partial r}{\partial t} = B\nabla^2\left(\frac{1}{r} - \frac{\partial^2 r}{\partial z^2}\right) \tag{1}$$

The parameter B, which is a constant at a given temperature, is given by

$$B = \gamma_s \Omega D_s \delta_s / k_B T \tag{2}$$

where γ_s is the surface energy, Ω is molar volume divided by Avogadro's number, D_s is surface diffusivity, δ_s is surface width, k_B is Boltzmann's constant and T is temperature.

The instantaneous amplitude $\in(t)$, of a sinusoidal perturbation of wavelength λ and an initial amplitude of $\in_0$ is given by

$$\varepsilon(t) = \varepsilon_0 \exp\left(\frac{t}{\tau}\right) \tag{3}$$

and the rate of growth of amplitude $\frac{1}{\tau}$ has the following dependence on the wave number ($k = 2\pi/\lambda$):

$$\frac{1}{\tau} = \frac{(kR)^2 - (kR)^4}{R^4/B} \tag{4}$$

The following key results flow from the preceding equations. There exists a maximum in the growth rate for a perturbation with a wavelength $\lambda_{max} = 22\pi R$ which is a function of R alone [3]. Physically, the origin of the terms in Eq. (4) may be understood as follows: in a cylinder with a sinusoidal perturbation of amplitude $\in$, there arise gradients in two principal curvatures, $\kappa_1 = (1/(r+\in))$ and $\kappa_2 = (\partial^2 \in/\partial z^2)$. The variation of κ_1 causes a chemical potential gradient that drives atoms from the depressions (larger κ_1) to bulges (lower κ_1) along the interface, while that in κ_2 drives atoms in the reverse direction. The former grows the perturbation amplitude $\in$, while the latter shrinks it. From dimensional arguments, it can be shown that growth rates in $\in$ due to these two effects vary as $(k/R)^2$ and $-k^4$, respectively, and causing the emergence of a dominant length scale $(2\pi/k_{max})=\lambda_{max}$.

As we show in our results in the following sections, not only does a perturbation with λ_{max} dominate the onset of instability, it also becomes the principal length scale even at late stages when the pore is about to close.

3. Phase field model and solution techniques

We formulate our phase-field model in terms of non-conserved order parameters $\eta_s(r,t),\eta_v(r,t)$ which act as indicator functions denoting the presence and absence of the solid or vapour phase respectively; i.e., when η_s=1 and η_v=0, it denotes the solid-phase, and conversely when η_s=0 and η_v=1, it denotes the vapour phase. At the surface, $0<(\eta_s,\eta_v)<1$. Along with these non-conserved order parameters, we also use the density field $\rho(r,t)$ as a conserved order parameter which takes its equilibrium value of unity in the bulk solid; in the vapour phase, it has a value of zero.

Using the order parameters, we construct a free energy functional as follows:

$$F = N_v \int_V \left[f_{sv} + \sum_{i=s,v} \kappa_i (\nabla \eta_i)^2 \right] dV \tag{5}$$

$$f_{sv} = \rho^2 f_s + (1-\rho)^2 f_v + W_{sv}\eta_s^2\eta_v^2 \tag{6}$$

$$f_s = 0.25 + \frac{\eta_s^4}{4} - \frac{\eta_s^2}{2} + \eta_v^2 \tag{7}$$

$$f_v = 0.25 + \frac{\eta_v^4}{4} - \frac{\eta_v^2}{2} + \eta_s^2\ , \tag{8}$$

where κ_i is gradient energy coefficient and W_{sv} sets the height of the barrier between the energy of the solid and that of the vapour phase. This functional gives a free energy surface where the solid phase with a density of unity exists in equilibrium with vapour phase with zero density.

The evolution of the density field variable $\rho(r,t)$ is governed by the Cahn–Hilliard equation [18].

$$\frac{\partial \rho}{\partial t} = \nabla.\left[M\nabla\left(\frac{\delta(F/N_v)}{\delta\rho}\right)\right] \tag{9}$$

where atomic mobility M is decomposed into its bulk (M_{bulk}) and surface (M_s) components as follows:

$$M = M_{bulk} + 16.0(M_s\phi_s) \tag{10}$$

M is related to diffusivity, and φ_s is a function of order parameters $\eta_i(r,t)$ which has non-zero values only at the surface and vanishes at all other locations. We have chosen the following form for φ_s:

$$\phi_s = \eta_s^2\eta_v^2 \tag{11}$$

The non-conserved order parameters, η_s and η_v, evolve according to the Allen–Cahn equation [19].

$$\frac{\partial \eta_i}{\partial t} = -L_s\left[\frac{\delta(F/N_v)}{\delta\eta_i}\right] \tag{12}$$

where L_s is a kinetic parameter related to surface mobility. The model offers flexibility in choosing different M_s and L_s values to study their effect on microstructure evolution.

A note about the way our model is constructed is in order. We use two order parameters for the evolution of a single physical interface in the problem. Our model differs from other phase field models that use multiple order parameters with a sum constraint [20]. Also, since there is only a single interface, a single order parameter may have sufficed; however, we are motivated to choose two order parameters so that it can be extended easily to study instabilities in porous, polycrystalline materials; the results of this study will be published elsewhere.

The second point is about the incorporation of surface diffusion. In this particular formulation we follow a scalar variant (Eq. (10)), similar to several other formulations (see Refs. [15–17,21–23]). We are aware of the deficiencies due to such a formulation [24]; however, in the present study we will be investigating interface scales which are very small (atomic dimensions), thus reducing the errors originating from the simplistic construction of the interface mobilities. Therefore, our results remain unaffected as a function of the interface widths in this range of application. For modelling systems with much larger interface widths quantitatively, we will