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Analytical model of the effect of misfit dislocation character on the bubble-to-void transition in metals

Enrique Martínez [a, *], D. Schwen [b], J. Hetherly [a], A. Caro [a]

[a] Material Science and Technology Division, MST-8, Los Alamos National Laboratory, Los Alamos, 87545 NM, USA
[b] Fuels Modeling and Simulation, Idaho National Laboratory, Idaho Falls, ID 83415, USA

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ABSTRACT

This paper addresses the role of misfit dislocations in the nucleation and growth of nanoscale He bubbles at interfaces. In a recent work, we studied the nanoscale effects on the capillarity equation and on equilibrium conditions. We proposed an expression for surface energy and for the equation of state, EOS, for He in bubbles, which have a size dependence that captures the role of the interface forces, which become relevant at the nanoscale. Here we determine the EOS for several twist grain boundaries in Fe and Cu and incorporate these results into the rate equation that determines the bubble-to-void transition, focusing on the influence of interface dislocations on the evaporation rate of vacancies. We find a significant effect of the magnitude of the Burgers vector of the dislocations on the critical radius for the transition. These results give a quantitative way to characterize grain boundaries in their ability to capture He and alter the onset of swelling.

1. Introduction

Helium appears in fusion and fission nuclear materials as a result of nuclear reactions. Its presence is usually a nuisance as it affects mechanical properties in dramatic ways. Chemically inert, He has an exceedingly low solubility in all metals and a strong tendency to precipitate heterogeneously at defects such as dislocations, grain boundaries, or interfaces, as is the case in ODS steels [1]. Both experiments and modeling have recently shown that the atomic structure of the interface affects the He bubble distribution at the interface and the void denuded zone around the boundary [2–6]. He bubbles and their influence on swelling have received significant attention from the nuclear materials science community for years. Historically, the 80's was a decade when most of the analytic treatment of this problem, in terms of rate or diffusion equations, was developed. For a comprehensive treatment of the problem see Ref. 7. The time evolution of a bubble in a material under irradiation is determined by the balance between defects (interstitials, vacancies, and He) arriving and escaping the evolving bubble. Since each one of these defects have their own properties (source, annihilation, recombination, and trapping at sinks terms) the resulting rate equations usually do not have a truly predictive power, but help at giving the type of effects that may occur for a reasonable choice of the numerous parameters entering the model.

One of the main conclusion of these theories was the existence of a critical radius for the bubble-to-void transition, that is, a critical size for a given He content, from which bubbles grow indefinitely by capturing vacancies and becoming voids. Another key discovery was the existence of a second radius, smaller than the critical, called stable radius, where bubbles remain at stationary size under irradiation. These two radii together determine a bimodal bubble size distribution, a fact verified experimentally.

The equation that displays these effects is [7–12],

$$\frac{dR}{dt} = \frac{\Omega}{R}\left[Z_v^c D_v C_v - Z_i^c D_i C_i - Z_v^c D_v C_v^c\right] \quad (1)$$

where R is the cavity radius, Ω is the volume of an atom, Z's are capture cross sections or sink strengths, proportional to the area of the cavity, D's are diffusion coefficients, C's are concentrations, v stands for vacancies, i for interstitials, and c for cavity. The first term on the r.h.s represents the contribution to growth rate via vacancy capture, the second, with negative sign, represents interstitials capture; both depend on the concentration of defects in bulk far from the cavity, determined by the irradiation condition. The third term represents the evaporation of vacancies and depends on the

* Corresponding author.
E-mail address: enriquem@lanl.gov (E. Martínez).

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concentration of vacancies at the surface of the cavity. This quantity is obtained assuming that in the matrix, but very close to the bubble surface, the vacancy concentration in the crystal is at equilibrium with the bubble. The equilibrium condition translates into the equality of the chemical potentials inside and outside the bubble. The chemical potential for vacancies in the lattice close to the cavity, μ_v^L at concentration C_v^c, and the chemical potential for vacancies at a bubble with radius R, gas pressure P, and surface energy γ, μ_v^b, are given by Ref. 11,

$$\mu_v^L = kT \ln\left(\frac{C_v^c}{C_v^e}\right) \tag{2}$$

$$\mu_v^b = \Omega\left(\frac{2\gamma}{R} - P\right) \tag{3}$$

Equating them, we obtain:

$$C_v^c = C_v^e \exp\left[\frac{\Omega}{kT}\left(\frac{2\gamma}{R} - P\right)\right] \tag{4}$$

with

$$C_v^e = \exp\left[\frac{-\varepsilon_v^f}{kT}\right] \tag{5}$$

where k is the Boltzmann constant, and T the temperature. In the absence of irradiation, Eq. (3) tells that if the gas pressure P equals the surface tension $2\gamma/R$ then the bubble is in equilibrium with the crystal in the sense that there is no net vacancy flux, since their concentration at the surface of the bubble equals that in bulk.

The competition between out and incoming fluxes of defects opens the possibility to changes in sign in the bubble growth rate. For a judicious choice of parameters, and the simplifying assumptions of stationary vacancy and interstitial concentrations in bulk (which depend on the irradiation conditions) and of equilibrium concentration of vacancies at the surface of the cavity, i. e. C_v^c given by Eq. (3), a possible solution to this equation is represented in Fig. 1, where the intersections of the curve with the x-axis determine the stable, R_s, and critical, R_c, radii. The arrows at the bottom of the figure indicate the stability of the points: departures from R_s bring the bubble back to R_s, while deviations from R_c bring the bubble to either infinite growth or to the stable radius.

For constant values of the first two terms in Eq. (1), the growth rate is determined by the equilibrium vacancy concentration at the surface of the cavity; for a value larger than the stationary concentration of vacancies in bulk, the bubble will evaporate vacancies, while for values less than bulk, it will absorb vacancies. Again, for a detailed derivation of these equations see Ref. 7.

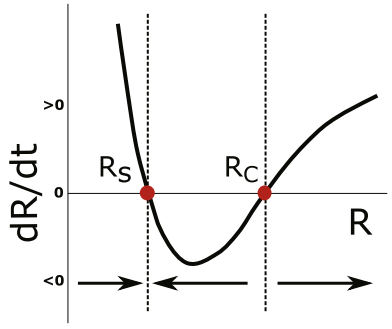

Fig. 1. Schematic view of the radial growth rate of a He bubble versus its radius in a situation of constant He content and irradiation produced flux of vacancies and interstitials. R_s and R_c are the stable and critical radii respectively.

This paper studies the validity of this approach to heterogeneous bubbles that evolve at an interface characterized in terms of misfit dislocations.

2. Background

We have heretofore presented the traditional formulation of the problem, developed in the 80's [9,13]. Eqs. (3) and (4) are the result of a continuum linear elastic approximation, namely, the properties of the gas are represented by P, and the properties of the metal matrix, where the bubble is located, by γ, and the two terms give independent contribution to the energy involved in adding a vacancy to the cavity, neglecting any gas–surface interaction. This paper analyzes how P and γ change for a given He density when the bubble grows at an interface that can be characterized in terms of misfit dislocations.

Recently, we have shown [14,15] that the capillarity equation in its original form can not be used for fluid bubbles embedded in solids because it suffers from the same limitations discussed by Tolman [16] for the case of a membrane separating two fluids. The fact that the interface has a finite width leads to diverse phenomena, such as the generation of a structure within the bubble that makes pressure an ill defined quantity, or the fact that the equilibrium condition does not happen for $P = 2\gamma/R$ but for an amount of He atoms that differs by more than a factor of two from it. We also determined that in the case of bubbles in solids, P and the density ρ can no longer be defined as global quantities related by an equation of state (EOS) for the fluid, but they become functions of position since the bubble develops a core–shell structure originated in the atomic scale nature of the interface. The range of the metal-gas interaction defines the width of the interface, that for the He–Fe system studied is ~0.4 nm, implying that bubbles around 1 nm in diameter are almost entirely affected by this interface effect, creating a radial pressure profile that is not constant in the fluid, nor zero in the metal, as the theory of the Eshelby inclusion would predict [17].

By relating the average pressure to an average density we proposed a new EOS for the pressure in the bubble that depends on the bubble radius and could be used in models of equilibrium or kinetic processes. The purpose of this paper is to explore in detail these nanoscale effects for bubbles nucleated at different environments characterized by a dislocation structure, such as twist grain boundaries (TGB) in bcc Fe and fcc Cu. Ultimately, the question we want to answer is: what is the effect of the microstructure in the bubble-to-void transition? Specifically we aim at finding the dependence of the critical radius, a relevant parameter that determines the onset of swelling, in terms of the material and GB properties.

3. Results

3.1. Study of the bubble pressure at interfaces

The fact that the He-metal interaction makes He atoms inside a bubble feel different forces depending on where in the bubble the atoms sit (close to the surface of the bubble, a strong metal repulsion creates forces that lower He density) has consequences when the bubble locates at a boundary composed of misfit dislocations: these density variations are sensitive to the presence of dislocations and translate into changes in the EOS of He. We derive these new EOSs and apply them to calculate the critical radius for the bubble-to-void transition.

To find the pressure dependence on bubble radius and location (i. e. bulk or dislocation cores in twist boundaries) that could be used in Eq. (4) to predict equilibrium or rate processes, we perform