Computational Materials Science 145 (2018) 86–95

Contents lists available at ScienceDirect

Computational Materials Science

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

Three-dimensional phase-field simulations of intragranular gas bubble evolution in irradiated U-Mo fuel

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ARTICLE INFO

Article history:
Received 27 September 2017
Received in revised form 28 December 2017
Accepted 29 December 2017

Keywords:
Phase-field
Intragranular gas bubbles
Fuel swelling

ABSTRACT

The evolution of fission gas bubbles in irradiated materials plays a critical role in the microstructural processes that leads to dimensional changes of U-Mo alloy fuels, e.g., fuel swelling. Although the intergranular bubbles-induced fuel swelling has been long-discussed for U-Mo fuel, there are very few computational studies of the formation of intragranular gas bubbles and its impact on fuel swelling. To this end, we develop a three-dimensional phase-field model to investigate the evolution of intragranular gas bubbles in U-Mo fuel. Fission induced defect formation and annihilation processes, such as vacancy-interstitial recombination, fission gas atom resolution, and interactions with dislocations and grain boundaries are incorporated in the model. Simulations show that the intragranular gas bubbles can be stabilized to certain sizes due to the balance between the generation and annihilation of defects. The intragranular gas bubbles induced fuel swelling is predicted to be comparable to experimental measurements. The effects of the irradiation and fuel fabrication conditions (i.e., fission rate, fuel grain size, and mechanical work induced deformation) on the bubble evolution and the resultant swelling are investigated. The current simulations provide a better understanding of intragranular gas bubble-induced swelling and a solid foundation for the future study of the nucleation and growth of intergranular gas bubbles and recrystallization in U-Mo fuel.

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1. Introduction

Understanding and predicting fission-induced microstructural changes in materials are of great importance to developing new fuels for both nuclear power and research reactors. U-Mo alloys are under investigation as a candidate fuel for future high power research reactors [1,2], because of their stable swelling behavior at moderate fission densities [3,4]. During the fission reactions of uranium-235, fission gas atoms, such as Xe, are generated at a rate of about 0.25 atom per fission as a result of decay of primary fission products. These fission gas atoms are almost insoluble in metallic fuels, and tend to accumulate in voids to form gas bubbles. The formation of fission gas bubbles can lead to serious dimensional changes of nuclear fuels, e.g., fuel swelling, which can affect the fuel performance and the long-term safety of reactors. Therefore, understanding the mechanism of the nucleation and growth of fission gas bubbles and their impact on fuel swelling plays a critical role in the qualification of U-Mo fuel for high performance research reactors.

Various types of fission gas bubbles have been observed in irradiated U-Mo alloy fuels. These gas bubbles can be categorized into three types based on their morphology and location: (1) intragranular bubbles – those inside fuel grains, usually of nanometer scale and evenly distributed; (2) intergranular bubbles – those on grain boundaries, usually of micrometer scale; and (3) periphery bubbles – those in the interaction layer of the fuel and Al matrix, usually large and unevenly distributed [5]. The final morphology of gas bubbles is affected by several factors, such as types of fuel, fuel fabrication conditions, and operation conditions of reactors. Although fission gas bubbles have different morphologies, the initial stage of bubble formation is considered to be similar, starting as isolated fission gas atoms inside fuel grains. Therefore, understanding the mechanism of the nucleation and growth of intragranular bubbles is critical to investigating the evolutions of other types of gas bubbles.

During fission events, incident irradiations produce isolated gas atoms and Frenkel pairs. The generated vacancies and self-interstitial atoms (SIAs) in equal numbers can annihilate either through their recombination to form perfect lattices or by interactions with defect sinks such as dislocations, grain boundaries, or precipitates. The efficiency of vacancy/SIA absorption by different sinks varies vastly because of the different sink strengths. The

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https://doi.org/10.1016/j.commatsci.2017.12.061
0927-0256/Published by Elsevier B.V.

relaxation volumes of SIAs in bcc alloys are typically much larger than those of vacancies, resulting in a higher rate of SIAs to interact with edge dislocations than vacancies [6]. This property is the origin of dislocation bias, and is the driving force for the formation of vacancy clusters. The fission gas atoms can be easily trapped by vacancy clusters to form intragranular gas bubbles inside fuel grains. Subsequently, the growth of intragranular gas bubbles are closely related to the effective diffusivities of fission gas atoms in fuel grains. Generally, intragranular gas bubbles confined inside fuel grains do not grow to an appreciable size. The phenomenon is attributed to the fission-induced gas atom re-solution process [7], in which small intragranular gas bubbles are partially or completely destroyed by close-passing fission fragments and some of the gas atoms in the bubbles are resolved into the fuel matrix. Therefore, the formation, growth, and dissolution of gas bubbles in U-Mo fuel are closely related to the diffusion of point defects and their reactions with defect aggregates.

Computer simulation as a cost-effective method can provide important information for the development of new materials. Various computational approaches have been used to study the behavior of gas bubbles in irradiated materials. Among those, phase-field method is a mesoscale approach that has been successfully applied to study the nucleation and growth of voids [8–11] and their migration [12–14], interstitial kinetics [15,16], crack propagation [17], gas bubble evolution [18–24], and recrystallization [25] in nuclear materials. In particular, Millett et al. [9,21] proposed a physics-based phase-field model to simulate the evolution of voids and gas bubbles in irradiated materials. Their model can capture the processes of point defect generation and recombination, and bubble nucleation and growth in the presence of grain boundaries. However, their simulations were performed in two dimensions, and several important defects reactions were ignored, such as fission-induced gas atom resolution, vacancy and SIA interaction with dislocations, and diffusion of gas atoms to grain boundaries. These defect reactions are important to the formation of the stable nanometer size gas bubble structures [7,21]. Although significant progress has been made, few phase-field models can reproduce the stable intragranular gas bubble structures under irradiation. One exception is a recent work by Hu et al. [26], in which a two-dimensional phase-field model was developed to study the formation of gas bubble superlattice in U-Mo. However, it should be pointed out that the generation of fission gas was artificially turned off in order to maintain a saturated Xe concentration in their model.

In this work, we develop a three-dimensional phase field model to study the gas bubble evolution in bcc U-Mo alloy with 7 wt.% Mo (U-7Mo) based on Millett et al.'s two-dimensional model. The large size difference between intragranular bubbles (size around 1.0–5.0 nm) and intergranular bubbles (size around 0.1–0.3 μm) makes simulating them simultaneously difficult because of the limitation of computing resources. In this work we focus on intragranular gas bubbles in U-Mo alloy fuel. The current model incorporates most of the key defect processes, including the defect production by fission, gas atoms re-resolution, grain boundary sinks for gas atoms, vacancy-SIA recombination, and dislocation sinks for the vacancy and SIA. We develop a new free energy for gas bubble phases based on the van der Waals equation of state (vdW EOS). By including the critical defect annihilation and gas-atom resolution processes, we demonstrate that the stable intragranular gas bubble structure observed in experiments can be reproduced. The predicted fission gas bubble size distribution and bubble-induced swelling of U-Mo are in agreement with experimental measurements. We also investigate the influence of fabrication and irradiation conditions on the growth kinetics of intragranular gas bubbles and the resultant fuel swelling in U-Mo.

2. Phase-field methodology

To study the evolution of fission gas bubble in U-Mo fuel, we chose three parameters as composition fields in the phase-field model, namely, the concentrations of fission gas Xe $c_X(r,t)$, vacancy $c_V(r,t)$, and SIA $c_I(r,t)$, which represent atoms or mole fractions at position r and time t. The phase parameter $\eta(r,t)$ is chosen to represent the gas bubble phase with $\eta = 1$ and the matrix phase with $\eta = 0$. η changes continuously from 0 to 1 across the interface between the gas bubble and matrix. The total free energy of the system can be described by

$$F(c_X, c_V, c_I, \eta, \varepsilon_{ij}) = \int \Big[f_{chem}(c_X, c_V, c_I, \eta, T) + \frac{\kappa_X}{2}|\nabla c_X|^2 + \frac{\kappa_V}{2}|\nabla c_V|^2 + \frac{\kappa_I}{2}|\nabla c_I|^2 + \frac{\kappa_\eta}{2}|\nabla \eta|^2 + f_{elas}(c_X, c_V, c_I, \eta, \varepsilon_{ij}) \Big] dV, \tag{1}$$

where $f_{chem}$is the chemical free energy density describing the composition and volume fraction of the equilibrium phases,κ_X, κ_V, κ_I, and κ_η are the gradient energy coefficients for Xe, vacancy, and SIA concentrations, and the phase parameter, respectively, f_{elas} is the elastic energy density, ε_{ij} is the strain, and T is the temperature. The first term in the volume integral represents the local contribution to the free energy from short-range chemical interactions. Four second derivative terms in the volume integral represent the interfacial properties. The last term in the volume integral represents the long-range elastic interactions.

The chemical free energy density describes the thermodynamic properties of the system by

$$f_{chem}(c_X, c_V, c_I, \eta, T) = h(\eta) f_b(c_X, c_V, c_I, T) + [1 - h(\eta)] f_m(c_X, c_V, c_I, T) + w g(\eta), \tag{2}$$

where f_b and f_m are the free energy densities of the gas bubble and matrix, respectively, $h(\eta)$ is an interpolation function with the form $\eta^3(6\eta^2 - 15\eta + 10)$, $g(\eta)$ is a double-well function with the form $\eta^2(1-\eta)^2$ to promote the stable phases, and w is the potential barrier height.

In deriving the free energy of the matrix, the condition $c_X + c_V + c_I + c_M = 1.0$ (c_M is the concentration of perfect lattice site) is always satisfied since we assume that the defects can occupy only the lattice sites. Thus, by adopting the ideal solution model, we can express the free energy of the matrix as

$$f_m(c_X, c_V, c_I, T) = E_X^f c_X + E_V^f c_V + E_I^f c_I + k_B T (c_X \ln c_X + c_V \ln c_V + c_I \ln c_I + (1 - c_X - c_V - c_I) \ln(1 - c_X - c_V - c_I)), \tag{3}$$

where E_X^f, E_V^f, and E_I^f are the formation energies of Xe, vacancy, and SIA in the matrix phase, respectively, and k_B is the Boltzmann's constant.

For the free energy of gas bubbles, the definition of the Xe atom concentration differs from that in the matrix. In bubbles, Xe atoms can occupy either lattice or non-lattice sites. The Xe concentration in the bubble is defined as $c_X^b = N_X/V_b$, where N_X is the number of Xe atoms in the bubble and V_b is the volume of the bubble. Based on this definition, the Xe EOS can be used to determine the free energy of gas bubble contributed by Xe accumulation. A detailed derivation of the Xe bubble free energy is given in the appendix. The free energy of the Xe bubble based on the vdW EOS is given by

$$f_X^b(c_X^b) = c_X^b \mu_0(p_0) + c_X^b k_B T \ln c_X^b + c_X^b k_B T \ln \frac{k_B T}{p_0} - c_X^b k_B T \ln(1 - B' c_X^b k_B T) + c_X^b k_B T B' \left(\frac{c_X^b k_B T}{1 - B' c_X^b k_B T} - p_0 \right), \tag{4}$$