EFFECT OF RECRYSTALLIZATION ON INTERGRANULAR BUBBLE SWELLING IN IRRADIATED U-MO FUEL

LINYUN LIANG, ZHI-GANG MEI, AND ABDELLATIF M. YACOUT

Nuclear Engineering Division, Argonne National Laboratory
9700 South Cass Avenue, Argonne, IL 60439 - USA

ABSTRACT

Radiation-induced swelling is one of major safety concerns in developing U-Mo metallic fuels for high performance research reactors. The formation of intergranular fission gas bubbles is a main cause of fuel swelling in U-Mo, which takes place simultaneously with fission-induced recrystallization. We present a mesoscale phase-field model to study the evolution of intergranular gas bubbles together with the recrystallization processes in irradiated U-Mo fuel. The developed model can successfully reproduce the microstructural evolution of the intergranular gas bubbles formation on the grain boundaries as well as the grain sub-division processes. The simulated intergranular gas bubble-induced fuel swelling exhibits two stages, i.e., slow kinetics before the recrystallization and rapid kinetics with the recrystallization. It is observed that the grain sub-division expedites the formation and growth of gas bubbles at high fission density because of the increased grain boundaries and decreased diffusion distance. The bubble size, shape, distribution, and density along the grain boundaries were computed and compared with the experimental measurements. The effects of the initial grain size and grain morphology on the gas bubble swelling were predicted. The larger initial grain size and larger grain aspect ratio structures have slower and faster swelling kinetics, respectively. It is suggested that the large initial grain size and small grain aspect ratio can help to reduce the gas bubble swelling rate. We conclude that the developed phase-field model can well capture the main physical phenomena of fission-induced gas bubble swelling in U-Mo fuel. The current model can be further developed as a useful tool to predict and control fission-induced swelling in nuclear materials.

1. Introduction

Development of new fuels with low enriched uranium to replace the use of highly enriched uranium fuels in research nuclear reactors has been extensively researched in order to reduce the nuclear proliferation risks worldwide. U-Mo alloys were selected as a promising candidate because of their excellent irradiation performance. To ensure the safe operation of nuclear fuels, one must be able to predict their behaviors during irradiation. The designed fuel should exhibit mechanical integrity and predictable swelling during irradiation. The swelling is a very important factor that increases the mechanical interaction between the fuel and cladding at high fission densities, which leads to the eventual fuel failure. The formation of gas bubbles are the main contribution to fuel swelling. Fission gases, mainly Xe and Kr, resulting from the fission of uranium atoms tend to be trapped in fabrication porosities and grain boundaries or accumulate on irradiation defects (vacancy clusters, dislocations) because of their low solubilities, resulting in the formation of gas bubbles. At high fission densities, the fuel particle exhibits the recrystallization phenomena, by which the large fuel grains with sizes of several microns are subdivided by small submicron grains, a process is also known as grain subdivision. The recrystallized grain structure with increased grain boundary density and reduced gas atom diffusion distance from grain interior to grain boundaries can accelerate the formation and growth of gas bubbles, which further expedites the fuel swelling. To our knowledge, few experiments have been made of gas bubble behaviors at high fission densities in U-Mo. Therefore, understanding and predicting the fission gas bubbles evolution especially at high fission densities with the effect of recrystallization and fission rate are critical to control the gas-bubble-driven swelling in U-Mo fuels.

Modeling is a very useful technique for simulating and predicting the fuel properties under irradiation conditions. The phase-field model can simulate the fuel microstructure at the mesoscale, which describes the microstructure with spatially continuous and time-dependent phase-field variables and naturally treats the material interfaces in a diffused interface description. Several phase-field models have been developed to study the grain structure effect on the fuel swelling. However, these models assume that the grain structures do not evolve with time, and recrystallization process was not considered [1-4]. In our previous work, [5] we developed a phase-field model to study the fission-induced recrystallization process in U-Mo alloy. The effects of the fission rate, grain size, and grain morphology on the recrystallization kinetics were systematically studied based on the analysis of the recrystallization growth rate using the modified Avrami equation. Although our model can well describe the recrystallization processes, however, it assumes that the intergranular gas bubbles are directly formed without considering their nucleation processes. The effect of recrystallization on the fuel swelling was not studied in detail. In addition, the dislocation density was obtained at steady-state conditions. The accumulation of dislocations because of the irradiation was not considered. Therefore, the objective of this work is to extend our previous phase-field model to incorporate the fuel microstructure information to study the effect of recrystallization on the intergranular gas-bubble-driven fuel swelling kinetics in U-Mo alloys.

Specifically, we extended our previous phase-field model to include the following physical processes: (1) nucleation and growth of intergranular gas bubbles, (2) fission-induced gas atom resolution from intergranular gas bubbles, and (3) irradiation-induced accumulation of dislocations in fuels. These processes can have considerable effects on the intergranular gas bubble evolution and recrystallization. Using the current model, we studied the intergranular gas-bubble-induced swelling kinetics together with the

recrystallization process. The bubble size, shape, distribution, and density along the grain boundaries were computed and compared with experimental measurements. Fuel swelling accelerated by the recrystallization was observed. The effects of the recrystallization rates due to the different initial grain size and grain morphology on the fuel swelling are discussed.

The remaining manuscript is organized as follows. Section 2 presents the model description for the evolution of dislocation density and the intergranular gas bubbles together with the recrystallization. Section 3 presents the computational details of the phase-field simulation and the material properties used for simulations. Detailed simulation results and discussion are given in Section 4. Section 5 provides conclusions and briefly discusses future work.

2. Model description

This section presents the formulations of the mesoscale model by considering the evolution of dislocation density, formation of intergranular gas bubbles, and the recrystallized grain growth processes. The irradiation-induced dislocation density evolution is described by an empirical expression fitted from the experimental data. The formation of intergranular gas bubbles coupled with the recrystallized grain growth is modeled by a multi-phase phase-field model.

2.1 Model of dislocation density evolution

During irradiation, the continuous production of point defects such as dislocation loops and dislocation networks leads to the accumulation of dislocations in U-Mo. In contrast, the dislocations can be annealed through thermal diffusion of point defects at high enough temperature. Considering both the production and the thermal annealing effects of the dislocation at the same time during the in-pile irradiation of U-Mo fuel, we can describe the mean dislocation density in U-Mo fuel by the following equation [6],

$$\frac{d\rho_d}{dt} = \left[A \cdot \dot{f} - g(T)\right] \cdot \rho_d, \tag{1}$$

where ρ_d is the mean dislocation density, $\dot{f}$ is the fission rate, A is relates the rate of accumulation of the dislocations, and $g(T)$ is a function of the temperature T in fuel that represents the thermal annealing of the dislocations.

The function $g(T)$ can be fitted by the experimental dislocation density evolution with time. Since the U-Mo fuel is usually operated at the relatively low temperature of 200 ^{o}C, the annealing of the dislocation can be ignored. By solving Eq. (1), the analytic solution of mean dislocation density is

$$\rho_d(t) = \rho_d(0) \cdot exp\left[A\dot{f} \cdot t\right], \tag{2}$$

where $\rho_d(0)$ is the initial dislocation density in the U-Mo fuel. Equation (2) shows that the dislocation density exponentially depends on the fission density.

2.2 Phase-field model for gas bubble evolution coupled with recrystallization

The irradiation introduces Xe gas atoms in fuel materials, which can nucleate, diffuse, and grow to gas bubbles on the grain boundaries. The accumulation of fission-induced dislocations increases the dislocation density, which results in an increase in the elastic

energy. Recrystallization starts when the elastic energy exceed a certain value. Once the recrystallization nuclei are formed, the growth kinetics follows the grain growth law. In the phase-field modeling of the fission-induced gas bubble evolution and recrystallization in the U-Mo alloy, the grain parameter $\eta_i(r,t)$ (*i*=1, 2, …, *p*) is chosen to describe the polycrystalline structure, the phase parameter $\eta_j(r,t)$ (*j*= *p*+1, p+2, …, *q*) is chosen to describe the gas bubble, and c_g is chosen to describe the fission gas Xe concentration. The total free energy of the system can be described by [5]

$$F(c_g,\eta_i) = \int \left[f_{bulk}(c_g,\eta_i) + \sum_{i=1}^{q} \frac{\kappa_\eta}{2} |\nabla \eta_i|^2 + \frac{\kappa_c}{2} |\nabla c_g|^2 + f_{stored}(\eta_i, \varepsilon_{ij}) \right] dV, \quad (3)$$

where f_{bulk} is the bulk free energy density describing the composition and volume fraction of the equilibrium phases; κ_η and κ_c are the gradient energy coefficients for the phase parameter and composition, respectively; *p* represents the total number of grain orientations in a grain structure; *q*-*p* represents the total number of gas bubble; and f_{stored} is the stored elastic energy density due to the introduction of dislocation density.

The bulk free energy used for the multiphase grain growth model is described by

$$f_{bulk}(c_g,\eta_1,\eta_2,\dots,\eta_q) = \sum_{i=1}^{q} \left(-\frac{A}{2}\eta_i^2 + \frac{B}{4}\eta_i^4 \right) + C_p \sum_{i=1}^{p} \sum_{j\neq i}^{p} \eta_i^2 \eta_j^2 + C_q \sum_{i=p+1}^{q} \sum_{j=1}^{p} \eta_i^2 \eta_j^2 + \frac{1}{4} + f_g^m \theta_m + f_g^b \theta_b, \quad (4)$$

where the last term describes the interactions between the fission gas and grain boundaries. Parameters *A*, *B*, C_p, and C_q are positive constants, and f_g^m and f_g^b are the chemical free energies of gas bubbles in the matrix and bubble phase [7].

The stored elastic energy in each grain due to the dislocation is described as

$$f_{stored}(\eta_i) = \frac{1}{2}\eta_i^2 G b^2 \rho_i, \quad (5)$$

where *G* is the shear modulus, *b* is the Burgers vector, and ρ_i is the dislocation density for each grain. In general the dislocation density varies in different grains depending on the grain size. For the U-Mo fuel, the grain size is usually around several micrometers. The difference in dislocation densities in different grains is relatively small due to their large grain sizes. Therefore, the dislocation density can be considered to be identical for different grains.

The spatial and temporal evolutions of grain parameters and Xe concentration follow the Allen-Cahn and Cahn-Hilliard equations [8],

$$\frac{\partial \eta_i}{\partial t} = -L_\eta \frac{\delta F}{\delta \eta_i}, \; i = 1,2,\dots,p, \quad (6a)$$

$$\frac{\partial c_g}{\partial t} = \nabla M(\eta) \nabla \frac{\delta F}{\delta c_g} + \dot{g} - \dot{R}, \quad (6b)$$

where L_η is the kinetic coefficient of grain boundary movement, M is the gas atom mobility, $\dot{g}$ is the fission production of the gas atoms, and $\dot{R}$ is the fission-induced gas atom resolution. The production rate of the Xe gases is described by $\dot{P}_i = \gamma_i Ran$, where γ_i is related to the fission rate and *Ran* is a random number uniformly between 0 and 1.

The fission-induced gas resolution is written as $\dot{R}_{re} = \eta^2 \theta c_g$, where θ is the resolution rate and η^2 (only for gas bubble phase parameters) is used to limit the gas resolution that occurs only inside the gas bubble.

The Xe diffusivity is believed to be higher on the grain boundary than in the bulk. To capture this phenomenon, we use a formula of gas atom mobility as $M(\eta) = M_b\, h(\eta) + M_g\, [1 - h(\eta)]$, where M_b and M_g are the mobilities of the gas atom in the matrix and on the grain boundary, respectively, and $h(\eta)$is an interpolation function having the form $h(\eta) = \eta^3(6\eta^2 - 15\eta + 10)$.

This phase-field model can be used to study the formation and growth of intergranular gas bubbles as well as the recrystallization in the irradiated U-Mo fuels.

3. Simulation details and material properties

To study the statistically averaged kinetics and topological features of the ideal fission-induced microstructure evolution, we performed phase-field simulations on a two-dimensional square domain. The phase-field model was implemented in an in-house simulation code, and the semi-implicit FFTW numerical method was employed to solve the coupled Eqs. (6) [46]. Periodic boundary conditions were imposed on the simulation domain. A model size of 25.6 μm×25.6 μm and a grid size Δx = Δy = 0.05 μm were used in the simulations. The average initial grain size is around 3 to10 μm, which is within the measured sizes in the U-Mo dispersion fuels [9]. The temporal evolution of grain structures with different initial grain size was studied in detail. In order to differentiate the different grains in the simulations, a function $\xi(r) = \sum_{i=1}^{p} \eta_i^2\,(r)$ was used, which takes a value of unity within the individual grain and a smaller value along the grain boundary region.

The initial gain structure was generated by using the conventional phase-field grain growth model. The different grain size structure can be obtained by controlling the evolution time. To simulate the nucleation of recrystallized grains, we randomly seeded the newly formed recrystallized grains on the grain boundaries instead of modeling the nucleation process directly. The formed nuclei were assumed to have a circular shape. The corresponding new phase parameter was created and assigned for each recrystallized grain. The dislocation density was set to zero for the newly formed recrystallized grains.

For the nucleation process, the Xe gas bubbles have a higher probability being nucleated on grain boundaries due to the facts that the Xe concentration is higher and its diffusivity is faster on the grain boundaries than in the bulk material. In the simulation, the grain boundary region is discretized into small areas. We assume a small-discretized area along the grain boundary to have a certain probability to generate a nucleus. Once it is generated, the probability of this area to nucleate the nucleus is set to zero. The nucleation probability in a discretized area is calculated as a function of the local nucleation probability [10]. In the simulation the nucleation step is executed at each time step, and the nucleation rate and probability are calculated for each discretized area with the supersaturation. The position of the nucleation can be determined after *P* is computed. The nucleation probability is set to zero if a nucleus already exists. In order to meet the concentration conservation condition, the introduction of nuclei requires a depletion region of the concentration field around the nuclei. In this paper, to avoid the calculation of depletion concentration profiles at each time step, we add the explicit nucleation process only for the non-conserved phase parameter equation (6a).

Table 1 lists the parameters of the U-Mo alloy used for simulating the fuel swelling by the phase-field model. We note that a single value for both the grain boundary energy and mobility was used for all grain boundaries in the present simulations, ignoring their variations. This simplified scenario, called idea grain growth, is used extensively by computer simulation.

Table 1. Material properties of U-Mo alloy used in the simulations.

Quantity	Symbol	Value	Reference
Temperature	T	473 K	
Interstitial diffusivity	D_i^0	3.81×10^{-9} m^2s^{-1}	[11, 12]
Vacancy diffusivity	D_v^0	5.72×10^{-8} m^2s^{-1}	[11, 12]
Atomic volume	Ω	$a^3/2$	[13]
Interstitial migration enthalpy	ε_i	0.27 eV	[12]
Vacancy migration enthalpy	ε_v	1.08 eV	[12]
Interfacial energy	γ	1.64 J/m^2	[14]
Grain boundary energy	σ_{gb}	0.36 J/m^2	[6]
Kinetic coefficient	L_η	1.82×10^{-14} $m^3s^{-1}J^{-1}$	-
Gas atom mobility	M_b	4.55×10^{-31} $m^5s^{-1}J^{-1}$	-
Gas atom mobility on grain boundary	M_g	100 M_b	-
Constant	S_l	1.0	-
Shear modulus	G	36.0 GP	[6]
Burgers vector	bv	3.42×10^{-10} m	[17]
Gradient coefficient	κ_c	2.74×10^{-7} Jm^{-1}	-
Constant	W	1.76×10^{8} Jm^{-3}	-
Initial Xe concentration	C_0	0.12	-

4. Results and discussion

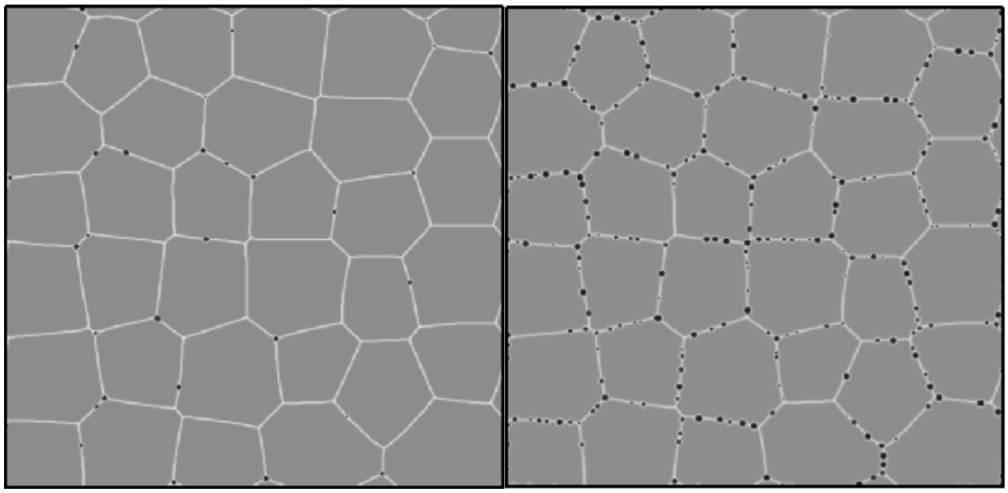
With the fitted dislocation density and the validation of the phase-field model, we simulated the formation of gas bubbles together with the recrystallization process in U-Mo alloys under fission. A fixed fission rate of 5.0×10^{14} $cm^{-3}s^{-1}$ was used in all simulations. We note that the experimental fission rates range from 2.4 to 7.0×10^{14} $cm^{-3}s^{-1}$. However, our previous simulation results showed that the fission rate has a minor effect on the recrystallization kinetics [5]. In this work, we focus on the effects of recrystallization on the gas bubble evolution. Thus, fixing the fission rate may have only a minor effect on the gas bubble evolution by recrystallization. Only a few studies have been made on the effect of fission rate on the swelling kinetics either from experiments or from simulations. It is still not clear how the fission rate affects the swelling kinetics, especially when the fission rate is higher than 7.0×10^{14} $cm^{-3}s^{-1}$. Our current assumption of using a fixed fission rate is probably valid only below this fission density. Detail discussions of the fission rate effect on the fuel swelling at high fission density is beyond the scope of this work. The total number of initial grains in the simulation is 34, and the average grain size in the initial microstructure is about 5.0 μm, which is a classic experimental grain size. During irradiation, the generated fission products Xe atoms diffuse either in bulk materials or on grain boundaries. The solubility of fission gas on the

grain boundary is substantially higher than in the bulk materials. The gas concentration on the grain boundary increases until its solubility limit is reached. Also, the Xe diffusivity is higher in the grain boundary than in the bulk materials. Therefore, Xe atoms prefer to be nucleated on the existing grain boundaries to form the intergranular gas bubbles.

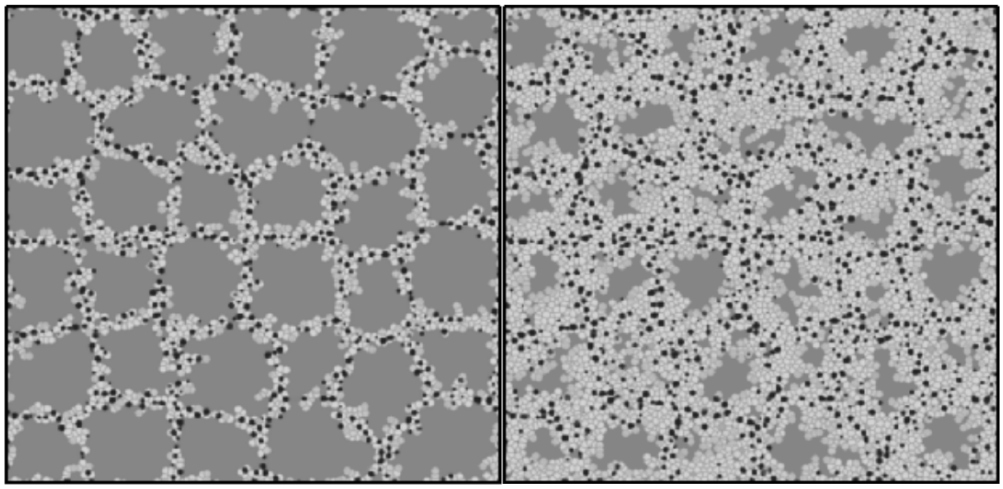
4.1 Gas bubble and grain structures

The simulated microstructural evolution in U-Mo under fission with respect to fission density is shown in Fig. 1. During irradiation, the ongoing production and diffusion of Xe increase the number and size of the gas bubbles. Because of the fission-induced resolution, part of Xe atoms inside the intergranular bubble can be knocked out of the bubble and go back into the grain interior, that reducing the size of the bubbles. For small gas bubbles, they may dissolve eventually. The typical operating temperature of research reactors is about 200 ^{o}C; thus the bubbles are considered immobile. If two bubbles get close to each other, they can coalesce into a bigger bubble. With the increase in fission density, the density of the gas bubbles continuously increase. Two main contributions are responsible for this phenomenon. One is the irradiation-produced nucleation of Xe gas bubbles, and the other is the increased nucleation probability of bubbles (i.e., increased grain boundary area) due to the grain subdivision during recrystallization. The size of the gas bubbles ranges from 0.1 μm to 0.5 μm. The formation of gas bubbles is responsible for the fuel swelling. The increased number and size of fission gas bubbles at high fission densities lead to further increase of fuel swelling.

The increase in the dislocation density results in an increase in the stored elastic energy in the grain structure, which provides a driving force for the nucleation and growth of recrystallized grains. The recrystallized grains prefer to be nucleated on the pre-existing grain boundaries or gas bubble surfaces. These recrystallized grains then grow toward the center of the original grains. The number and size of the original grains decrease with increased fission density. After the new subgrains formed on the grain boundaries, a zero dislocation density was assigned for these grains. Therefore, there is no driving force for forming the recrystallized grains on the newly formed subgrain boundaries, which implies that the recrystallized grains will not form on the grain boundaries between two recrystallized grains. This agrees with the real situation that the elastic energy is released in the recrystallized grain area. Only the initial grain boundaries and the recrystallized front, that is, the grain boundaries between the newly formed grains and original grains, serve as new nucleation sites for recrystallized grains. In the simulation, recrystallization starts when the fission density is larger than 3.0×10^{21} f/cm^3. The full recrystallization of the U-Mo alloy can be achieved at a fission density of 6.2×10^{21} f/cm^3, where the fuel materials are fully covered by recrystallized grains and gas bubbles. The grain size of the fully recrystallized U-Mo alloy fuel ranges from 0.2 μm to 0.5 μm [49]. In terms of the energy reduction during the recrystallization process, the grain boundary energy and stored energy are the main driving forces for grain growth. The grain boundary energy drives the large grains to grow at the expense of small grains with the reduction of grain boundary energy, whereas the stored energy in the deformed grains induces the growth of recrystallized grains. Within the recrystallized area or after the full recrystallization, the grain growth is driven only by the grain boundary energy.



(a) 0.36× 10^{21} f/cm^3 (b) 3.0× 10^{21} f/cm^3



(c) 4.0× 10^{21} f/cm^3 (d) 5.3× 10^{21} f/cm^3

Fig. 1 Evolution of gas bubbles and grain structure as a function of fission density. Black dots: gas bubble; Grey color: original grains; White color: recrystallized grains.

4.2. Gas bubble properties before recrystallization

We first calculated the intergranular gas bubble density on grain boundaries before recrystallization. The grain boundary length is estimated as 0.38 μm^{-1}. Figure 2(a) shows the calculated number of bubbles per unit grain boundary length as well as the related experimental data for comparison. The experimental data are distributed in a large range because they were measured for different samples with different fission rates from 2.3×10^{14} cm^{-3}s^{-1} to 7.0×10^{14} cm^{-3}s^{-1} and temperatures from 66 ^{o}C to 185 ^{o}C. Details about the experimental conditions can be found in [15]. We note that only atomized sample data are plotted in the figures. Overall, the number of gas bubbles continuously increases with the increase in fission density due to the fission. The comparison shows

that the calculated number of bubbles per unit grain boundary length falls within the range of the experimental data.

We then calculated the number of gas bubbles per unit fuel cross area before recrystallization and also compared with the relative experimental data as shown in Fig. 2(b). We note that the experimental data for the bubble density are in volume fraction but that the simulated results are in area fraction. However, it has been suggested that the bubble volume fraction is equal to the bubble area fraction measured on the cross section area if the bubbles are homogeneously distributed [16]. Thus, we compared the simulated bubble area fraction directly with the experimental data. The results show a good agreement between them.

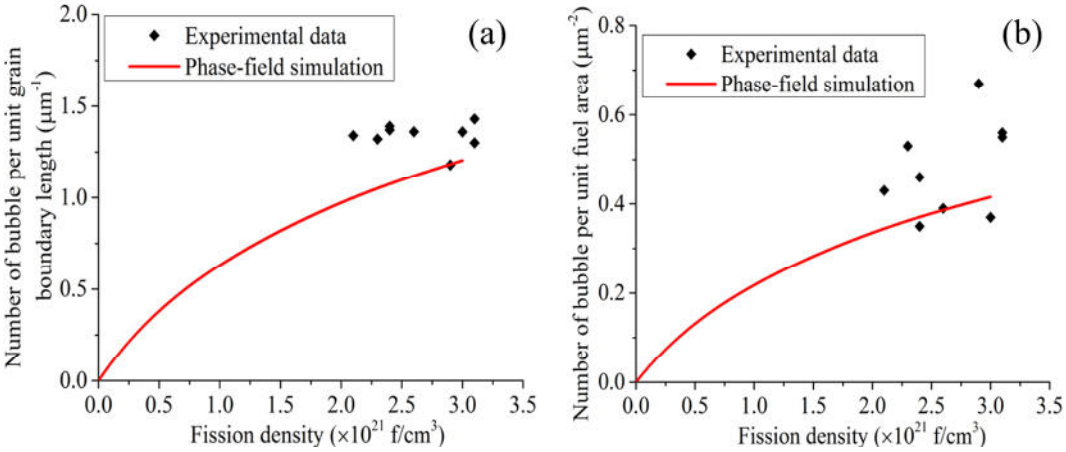


Fig. 2 (a) Comparison between the experimental and calculated bubble density per grain boundary length. (b) Comparison between the experimental number of bubbles per unit fuel volume and calculated number of bubbles per unit fuel area.

4.3 Gas-bubble-driven fuel swelling and recrystallization kinetics

To study the effect of recrystallization on the kinetics of gas bubbles evolution under fission, we calculated the fuel swelling as well as the recrystallized volume fraction as a function of fission density. The intergranular gas-bubble-induced fuel swelling was calculated by using the formula $\Delta V/V_0$, where ΔV is the increased volume due to the formation of gas bubbles, and V_0 is the original volume of the fuel. The calculated fuel swelling as well as the experimental correlation are shown in Fig. 3(a) [17]. The experimental correlation derived by Kim et al, [17] based on the experimental data describes the fission gas-bubble-induced fuel swelling. The fuel swelling empirical expression is a function of fission density and is a step function for describing two different swelling kinetics: before recrystallization and with recrystallization. Before recrystallization, the gas-bubble-induced fuel swelling is linearly proportional to the fission density, which is small compared with the fuel swelling with recrystallization, that the fuel swelling is a quadratic polynomial function of fission density. In our simulation, we also observed that the fuel swelling shows two different stages before and after the fission density of 3.0×10^{21} f/cm^3. The formation of gas bubbles can be divided into two mechanisms based on the two stages, before recrystallization and with recrystallization. Before recrystallization, the original grains grow slowly at low operating temperature.

The grain boundary density remains almost the same at this stage. The nucleation and growth of gas bubble are relatively slow and not affected by the recrystallization. At high fission densities with recrystallization, the formation of gas bubbles is significantly accelerated. This acceleration is attributed to the increase in the grain boundary density with forming more recrystallized grains during recrystallization, which provides more nucleation sites for the formation of gas bubbles. In addition, the large grains divided by small recrystallized grains further decreases the Xe diffusion distance from the grain interiors to the grain boundary areas. The growth rate of the corresponding gas bubbles increases. Therefore, the recrystallization expedites the fuel swelling at high fission densities.

The recrystallization volume fraction was also calculated by dividing the total volume of the recrystallized grains and newly formed gas bubbles by the initial sample volume, an approach similar to the one used by Kim et al, [9]. The results are shown in Fig. 3(b) together with the related experimental data. The good agreement between simulation and experimental results implies that the current model can well capture both the intergranular gas bubble evolution and recrystallization.

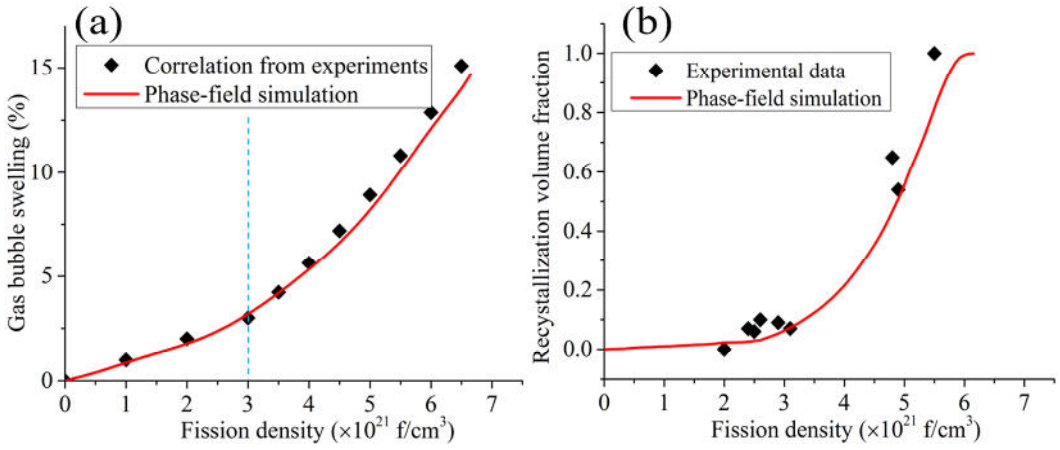


Fig. 3. (a) Comparison between experimental and calculated bubble-driven fuel swelling. (b) Comparison between experimental and calculated recrystallization volume fraction.

4.4 Grain size effects on the fuel swelling

The initial grain size of the fuel can greatly affect the recrystallization kinetics in U-Mo fuels during fission [5]. The initial large grain size in the microstructure can notably reduce the rate of recrystallization and therefore delay the full recrystallization to a higher fission density. As a result of the reduced recrystallization rate, the grain boundary density and the Xe atom diffusion distance from the grain interior to the grain boundary decrease in the initial large grain size structure. Therefore, the formation of the gas bubbles on the grain boundaries decreases, which influences the swelling kinetics of fuel materials eventually. Unfortunately, few experimental studies have been reported on the effect of recrystallization on the swelling kinetics due to different grain size for U-Mo alloy fuels.

To study the initial grain size effect, we simulated two additional grain structures with initial average grain sizes of 3.0 μm and 9.6 μm. The microstructure of U-Mo with

different initial grain size at a fission densities of 4.1×10^{21} cm^{-3} are given in Fig. 4. The full recrystallization in the microstructure with the smallest grain size is achieved fastest. The large grain boundary area in the small grain size structure provides more nucleation sites for the recrystallized grains, that increasing the recrystallization rate. In terms of the energy, the front of the recrystallized grains is driven by both the grain boundary energy and the stored energy between the recrystallized grains and initial grains, whereas the grains behind the recrystallized fronts have a smaller driving force that is attributed to the grain boundary energy. With a relatively large recrystallized front area, the recrystallized grains in the initial small grain size structure grow faster. On the other hand, the small grain size area can reduce the time of propagation for the recrystallization front, and the short grain boundary length can decrease the possibility of the formation of recrystallized nuclei. In general, a fine-grained microstructure recrystallizes more rapidly than a coarse-grained microstructure does, which is consistent with our previous conclusion [5].

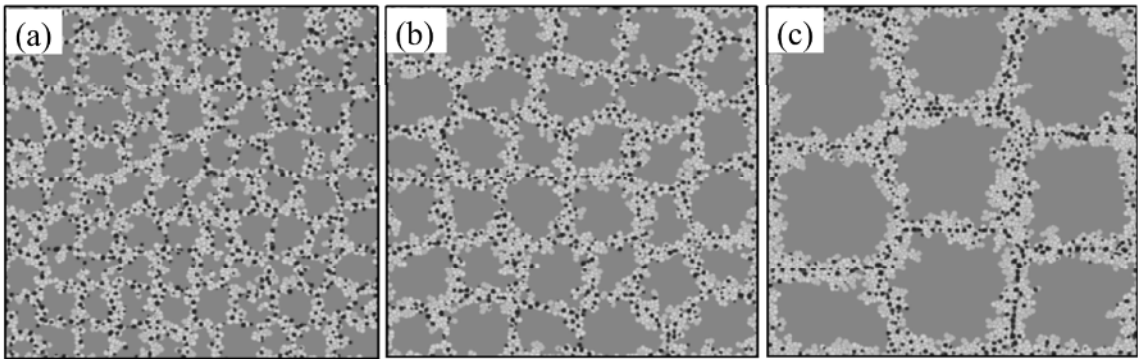


Fig. 4 Simulated grain microstructures of U-Mo alloy with different initial grain sizes (a) 3.0 μm; (b) 5.0 μm; (c) 9.6 μm at fission density of 4.1×10^{21} cm^{-3}.

To study the effect of the recrystallization rate on the fuel swelling by varying the initial grain size, we compared the fuel swelling for three initial grain size structures. As shown in Fig. 5(a), the case with the smallest initial grain size has the largest fuel swelling. Our simulation results qualitatively agree with the experimental observations, where the annealed samples with a relative large grain size give a relatively small gas bubble volume fraction in experiments [15]. We can also categorize the swelling kinetics into two stages, before recrystallization and with recrystallization. The effect of initial grain size on the fuel swelling is small before the recrystallization. In the small grain size structure, the large fuel swelling can be attributed to the increased bubble densities due to the increased grain boundaries and the reduced time needed for Xe gas atom diffusion from the grain interior to grain boundaries. With recrystallization, the fuel swelling is accelerated, and this effect becomes more prominent in initial small grain size structures. The increased fuel swelling in the initial small size grain structure is due to the creation of more grain boundary area during the recrystallization, which increases the probability of the formation of gas bubbles. The calculation of gas bubble density as a function of fission density confirms that the number of formed gas bubbles is relatively large in the small grain size structure, as shown in Fig. 5(b). The fast recrystallization

rate in the small grain size structure induces the large increase in the fuel swelling. Thus, the increase in grain size by annealing for the fuel particle can effectively decrease both the recrystallization rate and the fuel swelling.

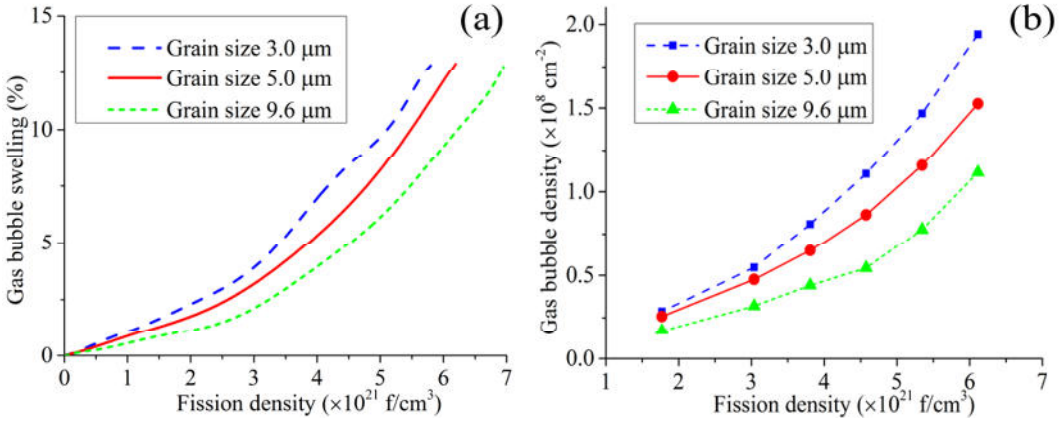


Fig. 5 Calculated fission gas-bubble-driven fuel swelling (a) and gas bubble density (b) for different grain sizes in the U-Mo alloy.

4.5 Grain aspect ratio effects on the fuel swelling

The grain aspect ratio is another important factor that has a large impact on the recrystallization kinetics based our previous studies [5]. In general, the grain structure with a large aspect ratio of the initial grains exhibits fast recrystallization. The fast recrystallization kinetics can further expedite the swelling kinetics. Therefore, it is of critical importance to understand the effect of grain morphology on the swelling of U-Mo.

We simulated three grain structures with different grain aspect ratios in order to study the effect of recrystallization on the gas-bubble-driven fuel swelling. The initial elongated grain structures are generated by a conventional grain growth phase-field model, and the grain boundaries are evolved to have diffused interfaces. The average grain size along one direction (e.g., *x*-direction) is larger than the other direction (e.g., *y*-direction). The grain aspect ratio is set as 1:1, 1:3, and 1:8 (e.g., *x*-direction/*y*-direction). The average original grain size remains the same 5.0 μm. Figure 6 shows the simulated recrystallized grain structure of U-Mo with three different aspect ratios at a fission density of 4.1×10^{21} cm^{-3}. As expected, the grain structure with the largest aspect ratio of the initial grains shows the highest recrystallization volume fraction. Full recrystallization is achieved fastest for the largest aspect ratio of 1:8. The reason can be attributed to the increased grain boundary area in the grain structure with large grain aspect ratio, and therefore the nucleation density of recrystallization grains increases. Moreover, the short diffusion distance perpendicular to the elongated direction largely reduces the time for the propagation of recrystallization front. Therefore, a larger grain aspect ratio structure can lead to a higher recrystallization rate [5].

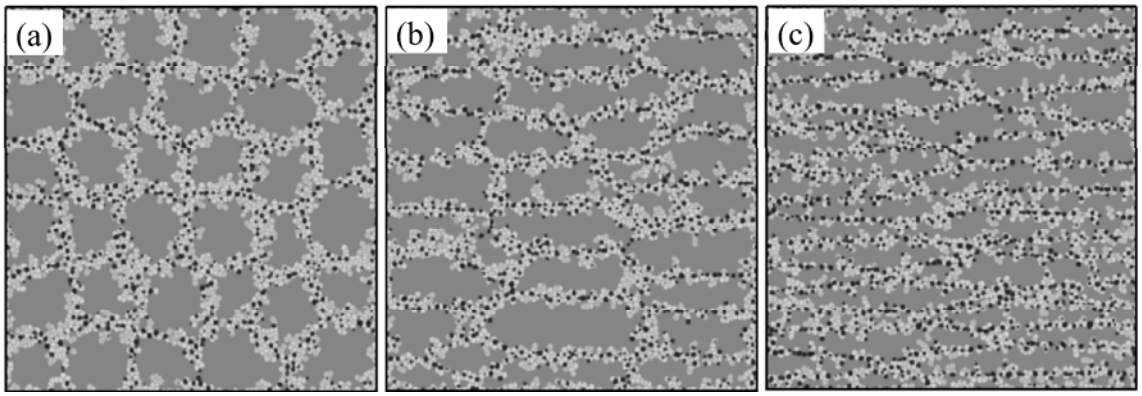


Fig. 6 Simulated grain microstructures of U-Mo alloy with different initial grain sizes (a) 3.0 μm; (b) 5.0 μm; (c) 9.6 μm at fission density of 4.1×10^{21} cm^{-3}.

We then studied the effect of recrystallization rate by varying grain aspect ratios on the fuel swelling of U-Mo alloys as a function of fission density. As shown in Fig. 7(a), the grain structure with the largest aspect ratio of the initial grains shows the highest fuel swelling. The number of gas bubbles is the largest for the largest aspect ratio of grain structures as shown in Fig. 7(b). The reason for forming more bubbles can also be attributed to the increased grain boundary area and reduced Xe atom diffusion distance in the grain structure with larger grain aspect ratio as discussed for the grain size effect. Compared with the grain size effect, however, the effect of the grain aspect ratio on the swelling rate is more prominent. This may be due to the significantly reduced diffusion distance perpendicular to the elongated direction for the high grain aspect ratio case. The fast diffusion of Xe gas atoms from grain interior to the grain boundaries leads to a large fuel swelling. Comparison between the effect of grain size (Fig. 11 [b]) and grain aspect ratio (Fig. 13 [b]) on the bubble density also indicates that the number of gas bubbles for the grain aspect ratio cases of 1:3 and 1:8 is larger than the one for the cases of largest grain size. Thus, the effect of grain aspect ratio on the fuel swelling is more prominent than the effect of grain size.

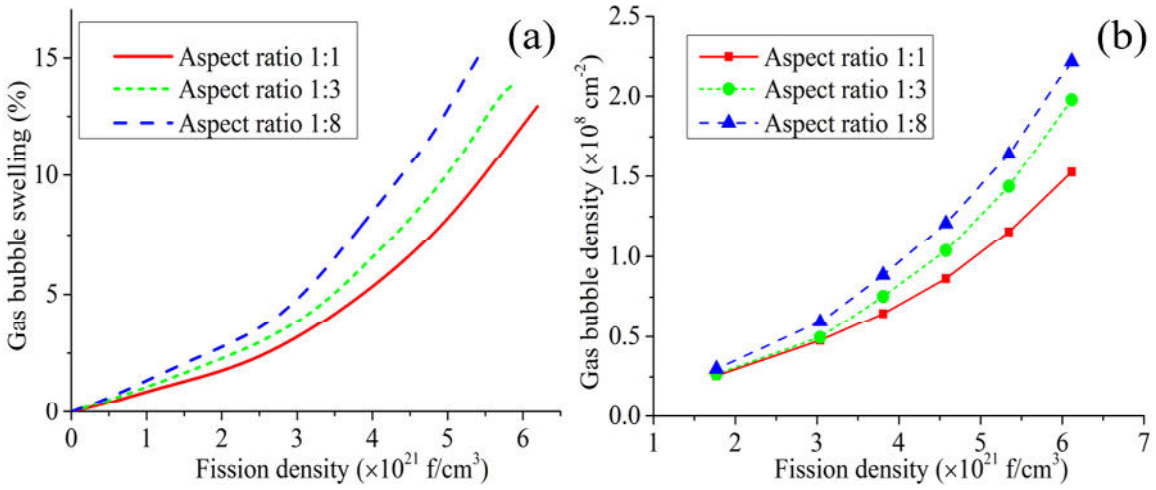


Fig. 7 Calculated fission gas-bubble-driven fuel swelling (a) and gas bubble density (b) for different grain aspect ratios in the U-Mo alloy.

Conclusions

We developed a phase-field model and used it to study the effect of recrystallization on the intergranular gas-bubble-driven fuel swelling in U-Mo alloy. The simulated fuel microstructure can well reproduce the microstructural evolution of intergranular gas bubbles together with the grain subdivision processes. The predicted fission density dependence of the recrystallized volume fraction agrees well with the experimental data. The simulated intergranular gas-bubble-driven fuel swelling exhibits two stages: a slow swelling kinetic and a rapid swelling kinetic. The rapid fuel swelling at high fission densities is attributed to the increased grain boundaries and the reduced gas diffusion distance due to the recrystallization processes. The grain morphology in the initial microstructure is found to have a significant impact on the fuel swelling kinetics. Increased initial grain size in the microstructure can reduce the recrystallization rate and therefore decrease the fuel swelling, whereas a high grain aspect ratio of initial microstructure can notably increase the recrystallization rate and therefore increase the fuel swelling. We point out that the effect of the grain aspect ratio on the fuel swelling of the U-Mo alloy is more prominent than that of grain size. Therefore, it is desired to increase the grain size and reduce the grain aspect ratio in the U-Mo dispersion and monolithic fuels in order to suppress the fuel swelling. The fission rate could have a considerable effect on the fuel swelling especially at high fission densities after full recrystallization. We plan to incorporate the fission rate effect on the recrystallization and gas-bubble-driven fuel swelling in our future work.

Reference

[1] S. Hu, A.M. Casella, C.A. Lavender, D.J. Senor, D.E. Burkes. Assessment of effective thermal conductivity in U-Mo metallic fuels with distributed gas bubbles, Journal of Nuclear Materials 462 (2015) 64-76.

[2] P.C. Millett, A. El-Azab, S. Rokkam, M. Tonks, D. Wolf. Phase-field simulation of irradiated metals Part I: Void kinetics, Computational Materials Science 50 (2011) 949-959.

[3] P.C. Millett, A. El-Azab, D. Wolf. Phase-field simulation of irradiated metals Part II: Gas bubble kinetics, Computational Materials Science 50 (2011) 960-970.
[4] S.Y. Hu, D. Burkes, C.A. Lavender, V. Joshi. Effect of grain morphology on gas bubble swelling in UMo fuels - A 3D microstructure dependent Booth model, Journal of Nuclear Materials 480 (2016) 323-331.
[5] L. Liang, Z.G. Mei, Y.S. Kim, B. Ye, G. Hofman, M. Anitescu, A.M. Yacout. Mesoscale model for fission-induced recrystallization in U-7Mo alloy, Computational Materials Science 124 (2016) 228-237.
[6] H.X. Xiao, C.S. Long, H.S. Chen. Model for evolution of grain size in the rim region of high burnup UO2 fuel, Journal of Nuclear Materials 471 (2016) 74-79.
[7] K. Chang, N. Moelans. Phase-field simulations of the interaction between a grain boundary and an evolving second-phase particle, Philosophical Magazine Letters 95 (2015) 202-210.
[8] I. Steinbach. Phase-Field Model for Microstructure Evolution at the Mesoscopic Scale, Annual Review of Materials Research, Vol 43 43 (2013) 89-107.
[9] Y.S. Kim, G.L. Hofman, J.S. Cheon. Recrystallization and fission-gas-bubble swelling of U–Mo fuel, Journal of Nuclear Materials 436 (2013) 14-22.
[10] J.P. Simmons, C. Shen, Y. Wang. Phase field modeling of simultaneous nucleation and growth by explicitly incorporating nucleation events, Scripta Materialia 43 (2000) 935-942.
[11] J. Rest, G.L. Hofman. An alternative explanation for evidence that xenon depletion, pore formation, and grain subdivision begin at different local burnups, Journal of Nuclear Materials 277 (2000) 231-238.
[12] J. Rest. Derivation of analytical expressions for the network dislocation density, change in lattice parameter, and for the recrystallized grain size in nuclear fuels, Journal of Nuclear Materials 349 (2006) 150-159.
[13] J. Rest. A model for the influence of microstructure, precipitate pinning and fission gas behavior on irradiation-induced recrystallization of nuclear fuels, Journal of Nuclear Materials 326 (2004) 175-184.
[14] L. Liang, Z.-G. Mei, Y.S. Kim, T. Wiencek, G. Hofman, M. Anitescu, A.M. Yacout. Intragranular Gas bubble kinetics in an irradiated U-Mo fuel using a multistate simulation approach. 2015.
[15] Y.S. Kim, G.L. Hofman, J. Rest. Characterization of Intergranular Fission Gas Bubbles in U-MO Fuel. Argonne National Laboratory: Argonne National Laboratory, 2008.
[16] R.T. DeHof, F.N. Rhines. Quantitative microscopy, McGraw-Hill, New York, 1968.
[17] Y.S. Kim, G.L. Hofman. Fission product induced swelling of U-Mo alloy fuel, Journal of Nuclear Materials 419 (2011) 291-301.