Final Report
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Computational Materials Science Network (CMSN) Project:

Multiscale Simulation of Thermo-mechanical Processes in Irradiated Fission-reactor Materials – DE-FG02-07ER46367

Performance Period: 5/1/2007 and ended on 4/30/2010
Total funding: $80,283

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Overview

This project focused on one sub-task of a larger Computational Materials Science Network (CMSN) project with the same title, which was led by Dr. Dieter Wolf of Idaho National Laboratory. This substask focused on the development of phase field approach for microstructure evolution in materials under irradiation. The project supported one doctoral student, Srujan Rokkam, for two semesters per year during the project performance period. This support was augmented by other sources. The student spent substantial summer time at Idaho National Laboratory collaborating with the Dr. Wolf and his post doctoral associates, Paul Millet and Michael Tonks. Mr. Rokkam obtained his Ph.D. degree in the summer of 2011. Dr. El-Azab also spent a few weeks at Idaho National Laboratory as a part of this collaboration.

The phase field model development effort aimed to foster collaboration in the area of mesoscale modeling of irradiation-induced microstructure evolution in reactor materials, with emphasis on bringing the phase-field modeling capabilities into this important field. The model problem chosen for this collaboration among three CMSN team members is the problem of nucleation and growth of voids. Like all other microstructure and micro-chemical evolution processes in irradiated materials, especially alloys, void nucleation and growth is driven by the intense generation of point defects under neutron irradiation, and by the long-range diffusion and interactions of these defects. Below the main accomplishments of this subtask are briefly outlined.

Research Accomplishments

1) Initial development of the phase field model

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We have developed the first phase field model for void nucleation and growth in irradiated materials based on the notion that such a model must include both conserved and non-conserved order parameters. Conserved order parameters represent the point defect species. The non-conserved order parameter represents the material domain portioning between matrix and void phases.

The first version of the model included only vacancies as the main defect necessary to form voids in irradiated materials. Our model was based on two governing equations: a Cahn-Hilliard equation governing the diffusion, reactions and generation of vacancies, and an Allen-Cahn equation for the space and time evolution of the non-conserved order parameter. These equations are written as follows:

$$\frac{\partial c_v(\mathbf{r},t)}{\partial t} = \nabla \cdot M_v \nabla \frac{\delta F}{\delta c_v(\mathbf{r},t)} + S_v + \xi_v(\mathbf{r},t), \text{ and } \quad \frac{\partial \eta(\mathbf{r},t)}{\partial t} = \quad -L \frac{\delta F}{\delta \eta(\mathbf{r},t)} + S_\eta + \zeta(\mathbf{r},t) \qquad (1)$$

In the above, c_v is the vacancy field, η is a non-conserved phase field variable, F is the free energy functional of the system, S_v and S_η are the production rates by collision cascades. The terms ξ_v and ξ_η are thermal fluctuations. The free energy F is given by:

$$F(c_v,\eta) = N\int_\Omega \left[h(\eta)\ \psi^m(c_v) + wg(c_v,\eta) + \kappa_v |\nabla c_v|^2 + \kappa_\eta |\nabla \eta|^2 \right] d\Omega \qquad (2)$$

The function $g(c_v,\eta)$ is the Landau energy, and ψ^m is the free energy of the matrix with defects, which is expressed in the form:

$$\psi^m(c_v) = E_v^f c_v + k_B T\ \left[c_v \ln c_v + (1-c_v)\ln(1-c_v) \right]. \qquad (3)$$

The model outlined above is phenomenological in nature because it is derived from a free energy functional that includes materials parameters that are yet to be determined in terms of fundamental materials properties. In particular, the free energy functional of the system consists of the well known statistical physics expression of the free energy of a non-interacting vacancy system, plus additional energy terms that are inspired by the classical gradient formulations of the free energy of heterogeneous media in the theory of spinodal decomposition of Cahn and Hilliard [1, 2] and the theory of antiphase boundary of Allen and Cahn [3]. Bistability is built into the free energy expression by a Landau-type free energy term [4].

Consideration of the non-conserved order parameter η, which demarcates the matrix and void phases, makes it possible to introduce the surface energy of voids through the gradient term associated with this variable, and thus limits the diffuse interface region to a small width that transitions from a matrix phase with very low concentration of vacancies to a void phase that contains only vacancies.

The present model has been solved using explicit finite difference scheme and applied to a set of 2D test problems. All test cases indicate that the void surface (or void-matrix interface) can act as a source or sink of vacancies, and that voids grow (or shrink) depending on the background vacancy concentration. In particular, the test cases for single voids have successfully reproduced the Gibbs-Thomson effect; see Figure 1. A second important verification case has been the interaction between large and small voids that are closely separated. In this case, the model also successfully reproduces the Ostwald ripening effect; that small voids dissolve by giving vacancies back to the matrix, which are then absorbed by the larger void; see Figure 2. A third test case that was successfully simulated by this model was the nucleation in the vicinity of a free surface. This test case was modeled by investigating the nucleation process close to the surface of a large void, and the model revealed the expected trend that the presence of a free surface leads to the formation of a denuded or void-free zone due to massive absorption of vacancies by the free surface. Yet, the most important test of the present model has been the reproduction of the dynamics of homogeneous nucleation and growth of voids in a system that is initially void-free and subject to stochastic vacancy generation; see Figure 3. In particular, it has been found that the model reproduces the distinct stages of (I) incubation, (II) transient and steady state void nucleation, and (III) nucleation saturation and void growth, including ripening and coalescence of voids. The void fraction and void density were fit to formulae of classical models in both the nucleation and growth regimes, in spite of important differences between the current problem conditions and those considered in the related classical models.

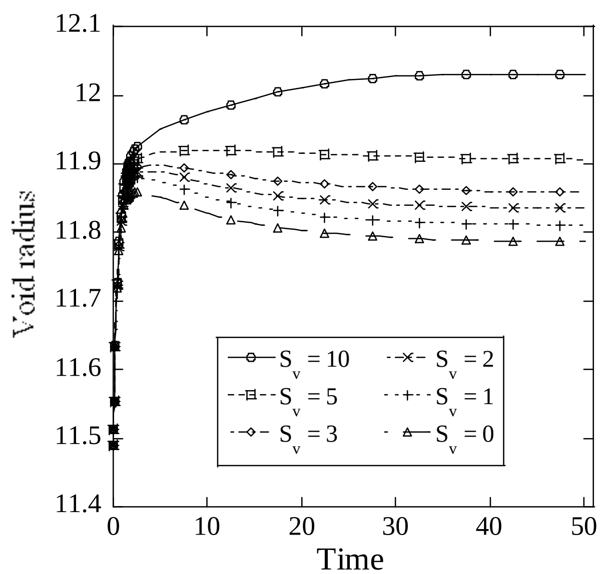

Figure 1. Void radius as a function of time for different initial vacancy supersaturation levels.

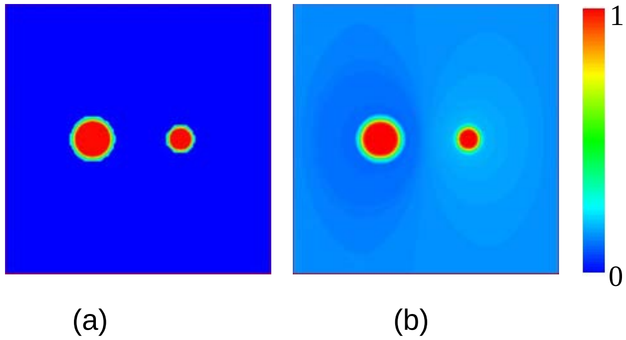

Figure 2. Evolution of concentration field around two interacting voids: (a) initial field and (b) a snapshot of the system at a later time. The concentration field is depressed around the larger void due to vacancy absorption, and it is higher than the remote background values closer to the small void due to vacancy emission.

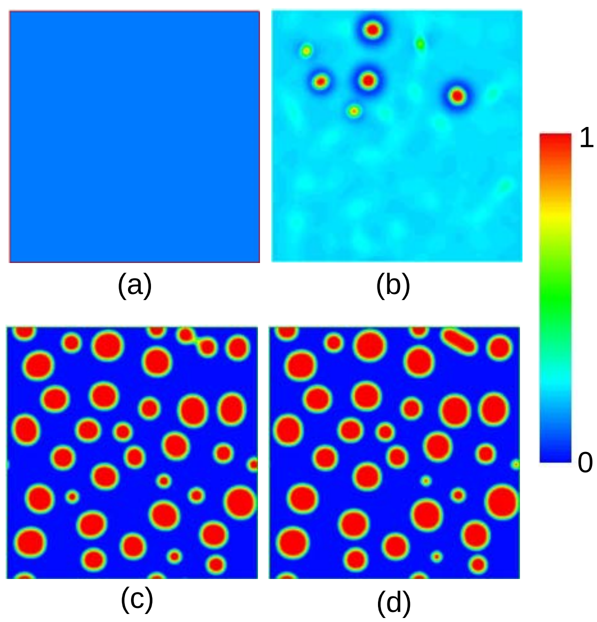

Figure 3. Display of the vacancy field illustrating the concurrent void nucleation and growth in a supersaturated system under the effect of stochastic vacancy generation: (a) initial system, (b), (c) and (d) evolution snapshots.

2) Generalization of the phase field model

The above model has also been generalized to polycrystalline solids [5] and to include both vacancies and interstitials [6,7]. In the later version, the model includes: (a) point defect generation by cascade processes and long-range diffusion of these defects in the crystal, (b) the mutual annihilation of point defects as well as the annihilation of point defects at other sinks, such as dislocations and grain boundaries, (c) the interaction of point defects and growing voids with the applied stress field, (d) the thermally- and cascade-induced fluctuations of defect densities, and (e) the evolution of voids by absorption of both vacancies and interstitials. Key aspects of the model and important results can be found in our publocations [5-7].

3) Significance of the research

The prevailing framework for understanding the effects of irradiation on materials microstructure involves rate equations that consider defect production, microstructure nucleation and microstructure evolution under the effects of temperature and stress. Theoretical and computational modeling effort in this field had proceeded in the past by assuming that these processes represent more or less separate stages of the overall problem. In reality, though, the three processes are coupled. The current framework tackles this very important issue and, as such, it has opened a new way for modeling radiation effects in materials. The community is now pushing harder in this direction.

Other Contributions by the PI

Dr. El-Azab, the PI, played a leading role under this community-based CMSN project. He lead the coordination of this subtask among the entire project team and that included:

- A major role in project meeting organization and coordinating of the talks related to this task area.
- Exchange visits with the lead PI and his post doctoral associates at Idaho National Laboratory.
- Rallying the community to generalize the results of this research throughout the field of radiation effects in materials.

Personnel Supported

As mentioned earlier, one doctoral student, Srujan Rokkam, was supposted under this project for two semesters per year for three years.

References

1. Cahn J W and Hilliard J E 1958 Free energy of a nonuniform system. I. Interfacial free energy J. Chem. Phys. 28 258-267

2. Cahn J W 1961 On spinodal decomposition Acta Metall. 9 795-801
3. Allen S M and Cahn J W 1979 A microscopic theory for antiphase boundary motion and its application to antiphase domain coarsening Acta Metall. 27 1085 - 1095
4. Rokkam S, El-Azab A, Millet P, and Wolf D 2009 Phase field modeling of void nucleation and growth in iorradiated metals, Modell. Simul. Mater. Sci. Engng. 17 paper # 064002: 1-18
5. Millett P, Rokkam S, El-Azab A, Tonks M and Wolf D, Void nucleation and growth in irradiated polycrystalline metals: a phase field formulation, Modell. Simul. Mater. Sci. Engng. 17 paper # pp 064003: 1-12
6. Millett P, El-Azab A, Rokkam S, Tonks M and Wolf D 2010 Phase field simulation of irradiated metals. Part I: Void kinetics. Computational Mater. Sci. 50 (2011) 949-959.
7. Rokkam S, Millett P, Wolf D and El-Azab A, Phase field simulation of void growth in irradiated materials, in: Proceedings of Fourth International Conference on Multiscale Materials Modeling, October 27-31, 2008, pp. 405-408

Publications that Resulted from this Research

1. Srujan Rokkam, Anter El-Azab, Paul Millett, Dieter Wolf, Phase Field Modeling of Void Nucleation and Growth in Irradiated Metals, Modelling and Simulation in Materials Science and Engineering, vol. 17 (2009) pp 064002: 1-18
2. Paul Millett, Srujan Rokkam, Anter El-Azab, Michael Tonks, Dieter Wolf, Void Nucleation and Growth in Irradiated Polycrystalline Metals: A Phase Field Formulation, Modelling and Simulation in Materials Science and Engineering, vol. 17 (2009), pp 064003: 1-12
3. Paul Millett, Anter El-Azab, Srujan Rokkam, Michael Tonks, Dieter Wolf (2010) Phase Field Simulation of Irradiated Metals. Part I: Void Kinetics. Computational Materials Science, 50 (2011) 949-959.
4. S.K. Rokkam, P.C. Millett, D. Wolf and A. El-Azab, Phase field simulation of void growth in irradiated materials, in: Proceedings of Fourth International Conference on Multiscale Materials Modeling, October 27-31, 2008, pp. 405-408

About five conference presentations were given.