MULTISCALE MODELING OF DEFECT GENERATION AND EVOLUTION IN METALLIC ALLOYS FOR FUSION POWER—Jörg Rottler, David J. Srolovitz, and Roberto Car (Princeton Institute for the Science and Technology of Materials, Princeton University)

OBJECTIVE

This work aims at creating a computational framework that is capable of describing the microstructural evolution of metallic alloys under irradiation on length scales reaching from the atomistic to the continuum. We focus explicitly on the annealing kinetics of elemental point defects (self-interstitials and vacancies) generated in high-energy collision cascades. To study the defect dynamics, we employ molecular dynamics and kinetic Monte Carlo simulations in combination with continuum rate equations. We begin by first considering pure bcc metals and then extend our study to the effects of alloying.

SUMMARY

We perform an analysis of the time evolution of self-interstitial atom and vacancy (point defect) populations in pure bcc metals under constant irradiation flux conditions. Mean-field rate equations are developed in parallel to a kinetic Monte Carlo (kMC) model. When only considering the elementary processes of defect production, defect migration, recombination and absorption at sinks, the kMC model and rate equations are shown to be equivalent and the time evolution of the point defect populations is analyzed using simple scaling arguments. We show that the typically large mismatch of the rates of interstitial and vacancy migration in bcc metals can lead to a vacancy population that grows as the square root of time. The vacancy cluster size distribution under both irreversible and reversible attachment can be described by a simple exponential function. We also consider the effect of highly mobile interstitial clusters and apply the model with parameters appropriate for vanadium and iron.

PROGRESS AND STATUS

We implemented a general kinetic Monte Carlo (kMC) model that represents Frenkel pair production, point defect diffusion, defect recombination and absorption at sinks in bcc metals. Of particular importance in our study is the mixed 1D/3D nature of self-interstitial diffusion in bcc metals [1]. We study the consequences of this 1D/3D random walk for the evolution of the point defect distributions with explicit kMC and arrive at excellent agreement with analytical scaling predictions. The elemental model is then solved with kMC and shown to be equivalent to a set of classic rate equations. Figure 1 shows the time evolution of interstitial and vacancy populations n_i and n_v for several different values of the production rate. Up to four distinct scaling regimes appear that can be easily described through the corresponding rate equations. Two of these are steady state regimes in which defect production is balanced by a combination of loss processes. Most interesting, however, is a non-steady state regime in which the vacancy density does not increase linearly with time but as the square root of time.

We then include vacancy clustering into our model and compute the time-dependent cluster size distribution in a point defect "gas". We find a simple, general exponential expression (see Fig. 2) that is a consequence of the continuous diffusive arrival of vacancies attaching to clusters and interstitials recombining with them. Other aspects of this study include the effect of highly mobile self-interstitial clusters, which is shown to anneal quickly at early times. In the final part of the study, we apply the model to vanadium and alpha-iron in which the diffusivities were determined using molecular dynamics simulations. Interestingly, our model predicts that the point defect kinetics in these two metals are very similar, despite large differences in the specific values of the energy barriers for interstitial and vacancy migration between these metals. While explicit kMC simulations can accurately capture the early dynamical regimes in the sub-dpa region, it is not feasible to extend these simulations to the macroscopic time scales of interest. Hence, properly

parametrized rate equations must be used. This work has been summarized in a research article currently submitted for publication [2].
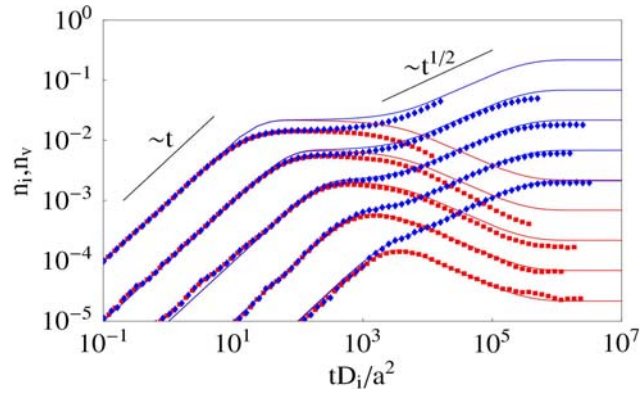

Fig. 1. Self-interstitial (red squares) and vacancy (blue diamonds) densities as a function of time for several different ratios of the interstitial hopping rate to the production rate (increasing from top to bottom). The solid lines show the result of direct numerical integration of continuum rate equations and the symbols correspond to the results of the kMC simulations in a periodic simulation cell.
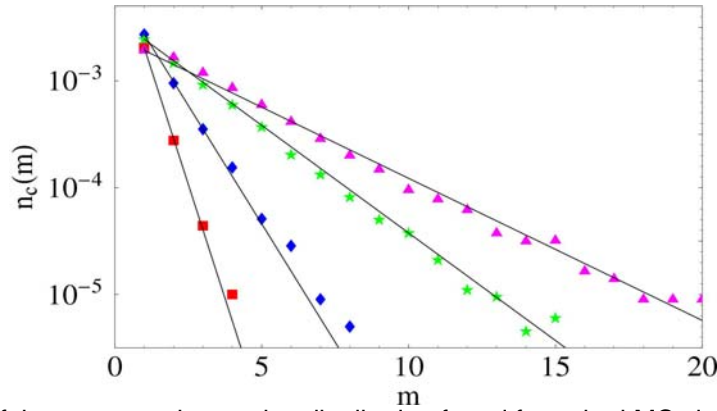

Fig. 2. Plot of the vacancy cluster size distribution found from the kMC simulations at four different times (time increasing from red to purple). The latest distribution corresponds to steady state. The straight lines show a simple analytic prediction $n_v(t)^m/(\text{const } n_i(t))^{(m-1)}$, where m is the number of vacancies in the cluster and the constant depends on the relevant ratios of production and hopping rates.

Based on our kMC simulation for pure metals, we have begun studies of models for diffusion in alloys. Impurity atoms must lead to a local modification of the defect hopping rates. We initially considered two different types of models [3] suitable for an implementation into lattice kMC:

- a "trap-model" (see Fig. 3), in which the hopping rates only depend on the initial position of the point defect, and
- a "barrier-model" (see Fig. 4) in which the hopping rate depends on the direction of defect motion (the link between two sites) and therefore on the initial and final defect position.
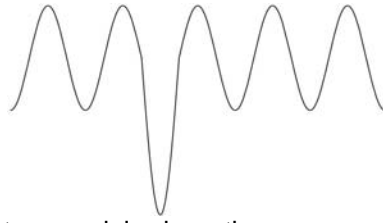
Fig 3. Energy landscape in a trap model, where the escape rate is increased at the impurity site.
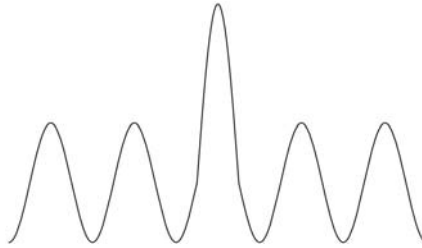
Fig 4. Energy landscape in the barrier model, where the hopping rates depend on the direction of motion near an impurity.

The most important difference between these two models is that the "barrier-model" induces correlations in the random walk, while the "trap-model" does not. For the trap model, one can therefore easily compute a total effective hopping rate by averaging over the total number of inverse hopping rates occurring in the system. For the "barrier-model," one can obtain a mean hopping frequency by directly averaging over all hopping rates, but the effective diffusivity is modified by a suitable correlation factor. The value of the correlation factor depends on the details of lattice geometry and hopping frequencies, and its computation represents the major challenge to describing diffusion in alloys.

Typical theories of vacancy assisted diffusion in alloys focus on models of the barrier type, and methods to compute the correlations are well established [4]. The simplest model, for instance, considers only two frequencies, one for vacancy-solvent exchange and one for vacancy-solute exchange regardless of the local environment. More complicated models such as the "five-frequency-model" are often considered for fcc metals. Somewhat less explored than vacancy diffusion is self-interstitial diffusion, because it is usually only relevant in radiation environments. Here the presence of solute atoms may not only modify the (usually very low) migration barriers, but may also affect the rotation frequencies. There is, therefore, a greater need to identify suitable models that capture the most relevant frequencies.

Work in progress therefore includes the implementation of some of the standard models for vacancy and self-interstitial diffusion in alloys into our kMC framework for bcc metals. As in the pure metals, we begin with the simplest models and then include more processes. Unlike in conventional diffusion studies, the focus here is on the transport of the defects themselves rather than on the self- and solute diffusivities. Also, the presence of defect production and annihilation may induce interesting dynamical effects on the alloy structure such as demixing or precipitation. Our objectives are, therefore, twofold:

- determine the effective diffusivities and the resulting modifications of the annealing kinetics of point defects in impurity rich environments
- study the time dependent effects of defect production, recombination and absorption on alloy composition and structure.

Once the general methodology is established, we will use EAM-type potentials to compute static exchange frequencies for specific ferritic alloys of interest, e.g., FeCr. Molecular dynamics of single self-interstitial diffusion will also be employed, as appropriate.

References

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