8.1 OBJECT KINETIC MONTE CARLO SIMULATIONS OF MICROSTRUCTURE EVOLUTION

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OBJECTIVE

The objective is to report the development of the flexible object kinetic Monte Carlo (OKMC) simulation code ***KSOME*** (kinetic simulation of microstructure evolution) which can be used to simulate microstructure evolution of complex systems under irradiation. In this report we briefly describe the capabilities of *KSOME* and present preliminary results for short term annealing of single cascades in tungsten at various primary-knock-on atom (PKA) energies and temperatures.

SUMMARY

We have developed a flexible lattice-based object KMC code, *KSOME*, to simulate the evolution of point defects in metals created by particle irradiation. In principle, *KSOME* deals with the migration, emission, creation, transformation and recombination of all types of intrinsic point defects and their complexes. In addition, the interaction of these point defects with sinks such as dislocations, grain boundaries and free surfaces is also treated. *KSOME* is designed such that all the necessary information regarding the properties of defects such as type, size, location, orientation etc., and their diffusion-reaction processes, along with simulation parameters regarding the size, temperature and properties of the simulation cell, are provided via text-based input files. This gives *KSOME* the capability to simulate complex systems involving complex processes that have not been possible with previously developed OKMC codes.

PROGRESS AND STATUS

Description of KSOME

KSOME is a flexible and computationally efficient OKMC code written using object-oriented methodologies in C++ to simulate long-time scale evolution of microstructure under irradiation. Patterned after the original FORTRAN code ALSOME [1], *KSOME* is significantly more sophisticated, faster and flexible. Accordingly, in *KSOME* objects of interest are vacancies, self-interstitial atoms (SIA), interstitial impurities and their respective clusters, produced during irradiation. Depending on various properties of these defects, the events these objects can perform are diffusion, dissolution (emission of defects from a cluster) and transformation of defect conformation and diffusion types. The probabilities for these events are calculated in terms of Arrhenius frequencies for thermally activated events, $G_i = n_i \exp(E_i/k_B T)$, where n_i is the attempt frequency (prefactor) for an event (process) I, E_i is the corresponding activation energy, k_B is Boltzmann's constant and T is the absolute temperature. The interaction between objects is treated as a non-activated or spontaneous event. *KSOME* is based on a rejection-free kMC algorithm also known as the BKL method [1]. The simulation proceeds by the random selection of an event from all possible events, weighted by its rate. The simulation clock is incremented by the time drawn from an exponential distribution. The efficiency of our simulations is improved by using a binary tree algorithm [2, 3] combined with lists.

KSOME keeps track of the locations of lattice defects, impurities and their clusters as a function of simulated elapsed time. Defects and impurities are treated as objects characterized by their

type, size, shape, position, and orientation. These defects (more precisely their centers of mass) are each associated with a specific lattice site, and they migrate by hopping from one lattice site to another in specific crystallographic directions corresponding to the lattice type. If a defect is within the interaction range of another defect, that interaction is performed, and the lists of defects are updated. The present version of *KSOME* can deal with cubic, fcc and bcc lattice types. In addition, *KSOME* allows for user-defined non-crystallographic diffusion directions to be included in the input script file. The present version of *KSOME* has no limit on the size of the simulation cell or the number of type of objects that can be included into the simulation. However, the memory consumption and speed of *KSOME* depends on the number of defects in the simulation cell as well as the size of the diffusion-reaction event databases, which are read at the start of a simulation.

Since *KSOME* simulates microstructure evolution under irradiation, the starting point for these simulations is the primary damage state, i.e., *(x,y,z)* locations of all defects along with their properties such as type, size and other available properties, obtained from molecular dynamic (MD) simulations of displacement cascades. With *KSOME*, apart from standard properties such as defect type and size we can assign additional properties to a defect if such information is available in the form of parameters. For example, one can differentiate between glissile and sessile SIA clusters or between a vacancy loop and a spherical void. Information about a primary damage state is provided to *KSOME* via a text-based input file; as such, defect information from other simulation tools can also be given as input for these simulations.

Depending on their properties, defects and defect clusters can diffuse randomly in 1-, 2- or 3-dimensions depending on their nature. If an object is a cluster it can also emit one or more defects. *KSOME* allows emission of multiple defects of the same type or different types if the cluster is a “complex”. For example, a vacancy, a SIA or an interstitial impurity atom (IIA) can emit single, or multiple entities of the same object while V+IIA or SIA+IIA can emit basic entities or complexes. We note that emission events can also include loop punching or trap mutation events as well. In addition to a migration and an emission event, another possible event is a transformation event. For example a transformation event includes an activated event like rotation or change of direction of 1D diffusing SIA clusters, or a vacancy loop transforming into a spherical void, or transformation from a glissile to a sessile loop. All thermally activated events a defect or an object can perform are user-defined in an activated-event database.

When two defects come within a given interaction distance, their interaction is modeled as a non-thermally activated event called a “reaction event”. The most common reaction events are the annihilation of a defect after encountering a defect of opposite nature (e.g., a SIA encountering a vacancy), and aggregation by adding a point defect to a cluster. All reaction events are considered diffusion limited, and there is no barrier for a reaction between two defects. Reaction events are considered as spontaneous events and therefore do not contribute to the progression of time. Since the defects are described as point objects, these events are identified on the basis of geometrical considerations (overlap of reaction volumes), and they take place spontaneously whenever two defects come closer than a reaction distance, which is equal to the sum of the capture radii associated with each of the two objects. The capture radius depends on the size and properties of a defect. At each KMC step all possible reaction events in the system are performed before the next KMC step is performed. Possible reaction events and capture radii are user-defined, and they are read from two separate input files at the beginning of a simulation. A reaction event database has a set of rules on how two defects should interact; while the capture radius database stores the sum of capture radii depending on the properties of interacting defects. In *KSOME* defects are considered as spherical objects, and their interactions with other defects are isotropic.

The fact that the possible diffusion-reaction events and capture radii are read from a text-based file at the beginning of a simulation and are not hardwired into the code allows a user to define any type of system. For example, for a *SIA-V-H-He* system it allows inclusion of intrinsic defects (vacancies (V) and self-interstitials (SIA)), He-V, H-V, H-He-V, He-SIA, H-SIA, H-He-SIA and H-He complexes as well as their interactions via reaction events. One new feature of *KSOME* that may not exist in previous or existing codes is its ability to recognize if no information about an active defect is available in the activated-event or in reaction or capture radius databases. For example, at any point during a simulation if no information on the activated event is in the database for a defect, the simulation stops after saving the simulation data (defect information). It is important to note that the user provides the information about activated events as an input; the code itself cannot predict them. It only informs the user that the required information does not exist in the database. *KSOME* is also structured such that additional modification to extend its capability and testing can be done relatively easily.

The basic *KSOME* code has been completed and it has been tested using data for 1MeV Kr^+ ion irradiation at 80 °C on a 36 nm molybdenum film 4. Fig. [1]1 compares the results from *KSOME* with cluster dynamics calculations of the areal defect density as a function of ion dose.

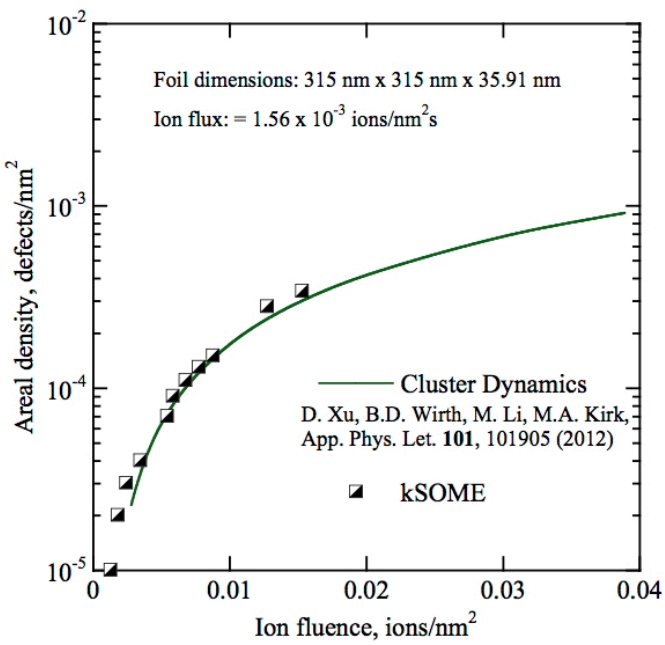

Figure 1. Comparison of cluster dynamics and *KSOME* calculations of the areal defect density dose dependence in a 36 nm molybdenum foil irradiated with 1 MeV Kr ions.

Short term annealing simulations of single cascades in Tungsten

Input data to KMC

A simulation was performed on a cubic block of tungsten atoms of size 162 nm^3 with each axis parallel to a <100> direction of the crystal. Each atom hops to a nearest neighbor site at a/2(111). Initial defect distributions of SIA and vacancy defects obtained from MD cascade

damage simulations were placed in the center of the box. Absorbing boundary conditions were adopted in all three directions.

The values of activation energies and pre-factors for diffusion and binding energies of defects used in our annealing simulations are taken from the *ab initio* calculations of Becquart *et al*. [5]. In the present model SIAs of all sizes were constrained to diffuse in 1-D along one of the <111> directions, and only single SIAs were allowed to rotate from one <111> direction to another with an activation barrier of 0.38 eV. Since MD does not provide information on the orientation of SIA diffusion, directions were assigned randomly to SIAs at the start of a simulation. All interstitial clusters are considered to be glissile (mobile), and their migration/diffusion rates decrease with increasing size. For a single vacancy, the activation barrier for diffusion is taken as 1.66 eV, and vacancy clusters larger than a dimer were assumed to be inactive i.e., they neither diffuse nor emit -- although they interact with other defects if they are within the range of interaction. The vacancy (SIA) dissociation rate is given by $G_d = n_d \exp((E_m + E_d)/k_BT)$, where E_d is the binding energy of a vacancy (SIA) to a vacancy (SIA) cluster, and E_m is the migration energy of a single vacancy (SIA).

RESULTS

Short-term annealing simulations using KSOME have been used to follow the evolution of cascade damage at an atomic scale beyond MD time scales. For PKA energies ranging from 1keV to 100keV, cascade annealing simulation results were averaged over a set of 20 cascades for larger PKAs (40-100 keV) and 15 cascades for smaller PKAs, generated using MD. Annealing simulations were carried out at temperatures of 300, 1025, and 2050 K for 10 ns. Figure [2] shows total number of defects (SIA + vacancies) in the simulation box initially and after 10 ns of simulation time.

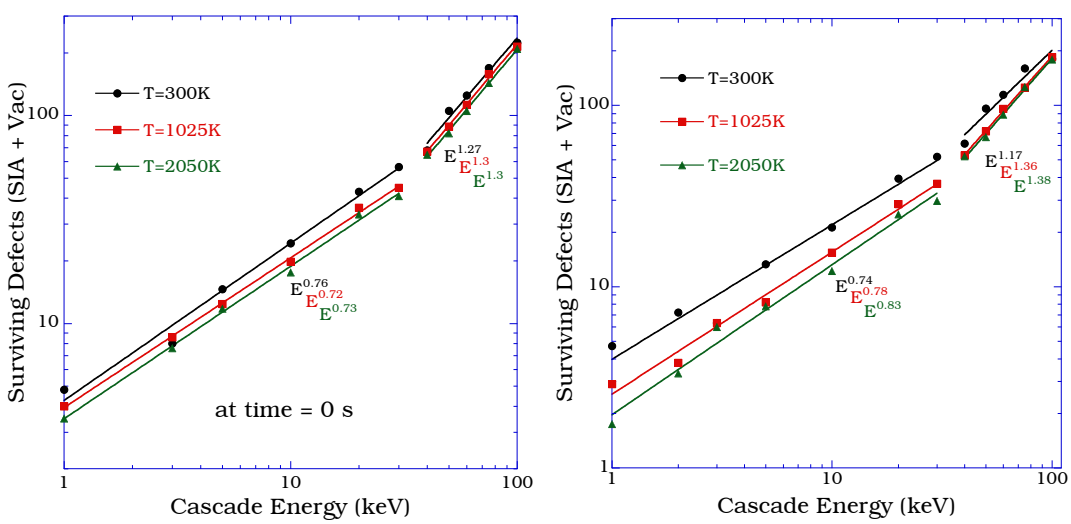

Figure 2. Surviving defects in tungsten before (left) and after (right) annealing for 10 ns.

It can be seen from Fig. [2] that the exponents did not change significantly after 10 ns of annealing. The characteristic slopes shown in Figure [2] are presumably related to different cascade morphologies as a function of PKA energy. The diffusion mechanism of the SIAs has a strong effect on the free interstitial fraction of stable defects. While none of the vacancies left the box due to 1D diffusion, most SIAs escape the cascade region, and they are either near the boundaries of the simulation box or have escaped the simulation box. Any loss in the number of vacancies after 10ns is entirely due to recombination. We find (not shown here) that the amount of recombination after 10ns of annealing depends on the temperature at which the cascade was generated and the PKA energy.

FUTURE WORK

Current knowledge of the diffusion mechanisms of SIA clusters in tungsten comes from MD, and the SIA migration and its activation energy remain a debated question, the sensitivity and effects of which can be studied further with parameter variations in *KSOME* simulations. Moreover due to the uncertainty in the diffusion mechanism of mid-size interstitial clusters, additional simulations will be carried out to illustrate the effects of rotation.

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