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Computer simulation of primary damage creation in displacement cascades in copper. I. Defect creation and cluster statistics

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ABSTRACT

Atomic-scale computer simulation has been used to investigate the primary damage created by displacement cascades in copper over a wide range of temperature (100 K $\leqslant T \leqslant$ 900 K) and primary knock-on atom energy (5 keV $\leqslant E_{PKA} \leqslant$ 25 keV). A technique was introduced to improve computational efficiency and at least 20 cascades for each (E_{PKA},T) pair were simulated in order to provide statistical reliability of the results. The total of almost 450 simulated cascades is the largest yet reported for this metal. The mean number of surviving point defects per cascade is only 15–20% of the NRT model value. It decreases with increasing T at fixed E_{PKA} and is proportional to $(E_{PKA})^{1.1}$ at fixed T. A high proportion (60–80%) of self-interstitial atoms (SIAs) form clusters during the cascade process. The proportion of clustered vacancies is smaller and sensitive to T, falling from 30% to 60% for $T \leqslant$ 600 K to less than 20% when T = 900 K. The structure of clusters has been examined in detail. Vacancies cluster predominantly in stacking-fault-tetrahedron-type configurations. SIAs tend to form either glissile dislocation loops with Burgers vector $\boldsymbol{b}$ = 1/2<1 1 0> or sessile faulted Frank loops with $\boldsymbol{b}$ = 1/3<1 1 1>. Despite the fact that cascades at a given E_{PKA} and T exhibit a wide range of defect numbers and clustered fractions, there appears to be a correlation in the formation of vacancy clusters and SIA clusters in the same cascade. The size and spatial aspects of this are analysed in detail in part II [unpublished], where the stability of clusters when another cascade overlaps them is also investigated.

1. Introduction

Materials selected for structural components in the core of nuclear power reactors are exposed to a flux of fast neutrons and high temperature. They have to withstand radiation damage processes that occur over wide ranges of time and length, i.e., from defect production at the atomic-level ($\sim 10^{-15}$ s, 10^{-10} m) to microstructural evolution over the mesoscale ($\sim 10^{6}$ s, 10^{-3} m), without exhibiting a significant degradation of service properties. In order to achieve progress in predictive modelling of property changes across these ranges, atomic-scale computer simulation occupies a special place, for it is able to provide much of the input to models at the higher levels. Large strides in simulating damage production and defect properties have been made over the past decade as a result of the increasing power of computational facilities and in this paper we present results obtained recently for pure copper. Although not actually employed for core structure applications, copper is the archetypal face-centred-cubic (FCC) metal as far as radiation damage is concerned, for it has been studied extensively in order to avoid complications of chemical effects that can occur in austenitic stainless steel, for example, and to exploit its suitability for detailed microscopic examination.

We consider displacement cascades, which are the primary source of radiation damage during fast-neutron irradiation of metals. They are formed by the recoil of primary knock-on atoms (PKAs) with a kinetic energy of more than ~1 keV. The cascade process is characterised by lengths and times of the order of nm and ps, respectively, and cannot be investigated directly by either experimental or analytical techniques. Only atomic-scale computer simulation by the method of molecular dynamics (MD) is suitable for studying the creation of point defects – for reviews see [1–5] and references cited therein – and their stability, mobility and interaction. The latter features are important because point defects cluster directly in the cascade process and the properties of the clusters have important consequences for microstructure evolution and property changes under cascade damage conditions. For example, an imbalance in the proportion of clustered defects of one species versus another, e.g. self-interstitial atoms (SIAs) versus vacancies, creates a complementary

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imbalance in the proportion of single defects, which diffuse three-dimensionally and are responsible for solute transport, swelling by void formation and creep, etc. Furthermore, SIA and vacancy clusters formed in close proximity create a weaker stress field than either would individually, and they also facilitate mutual recombination of defects as a result of cascade overlap in high-dose conditions.

A significant fraction of MD studies of cascades was applied to copper (see e.g. [5–15]), treating it as a model FCC metal with low stacking fault energy for fundamental study of radiation damage. However, most studies have treated only a few (typically ≈5) cascades at a given energy and irradiation temperature, and so firm statistics on cluster type and size distribution, particularly as needed for computational models of damage evolution, are lacking. Also, and possibly for the same reason, there appears to be an insufficiency of cluster types observed in earlier cascade simulations, so that it is not easy to compare results of modelling with experimental data. For example, low- and high-temperature experiments on copper have shown efficient production of stacking fault tetrahedra (SFTs) [16–18], but, with the exception of preliminary reports of our work [13–15], SFTs have not been observed systematically in cascade simulation. This is rather surprising because MD simulations of the evolution of a hot, vacancy-rich zone have demonstrated effective formation of SFTs in this environment [19–22]. Thus, we have undertaken an in-depth study of cascades in copper, but to convert MD modelling into a tool that produces not only qualitative information but also reliable quantitative data, it must be verified by a comparison with experimental results. This is not a trivial task because such a comparison can be made only with the predictions of a theoretical model that uses the MD data and the model itself may contain approximations and uncertainties. However, in some cases such comparison can be made with the results of specially designed experiments (e.g. [23–25]) with a reasonable accuracy. This places a requirement on the MD results that they be made realistic by carrying out a large number of cascade simulations and then undertaking detailed analysis of defect structure and properties, and statistical analysis of the results.

For the present work, therefore, we have simulated more cascades (∼450) for a wider range of conditions of irradiation temperature, T = 100–900 K and PKA energy, E_{PKA}, = 5–25 keV, and carried out more rigorous structural and statistical analysis than hitherto. The outcome is presented in the following manner. The computational methods used for cascade modelling and analysis are summarised in Section 2. The results for the number of defects created under the different irradiation conditions are presented in Section 3. A significant proportion of both vacancies and SIAs form clusters in the cascade process and statistics for vacancies and interstitials that are not single point defects are presented in Section 4. Typical cluster configurations that arise are described in Section 5. The nature of these is such that only those containing more than a certain number of defects behave as extended defects with at least some characteristics of dislocations, and these are analysed as a function of the irradiation conditions in Section 6. The results are discussed and conclusions drawn in Section 7.

We have computed the size distribution of the population of defect clusters in the MD modelling and compared the vacancy data with experiments on neutron-irradiated copper [24]. These results are to be reported in another paper, referred to here as part II [26]. The simulations indicate a correlation in the size and spatial arrangement of vacancy and SIA clusters formed in the cascade process, and this is also assessed in part II. Finally, we have investigated stability of typical SIA and vacancy clusters in copper against overlap by later displacement cascades, a situation that is common in neutron-irradiated copper when SFTs produced with high number density (up to $\sim 10^{24}$ m^{-3}) may overlap with new cascades. This part of the study is also presented in part II.

2. Simulation technique

2.1. MD method

An equilibrium, short-range, many-body interatomic potential [27] was used for the simulations. With this potential, the point defect formation energy for a vacancy and <100> dumbbell SIA is 1.19 eV and 3.62 eV, respectively. The vacancy migration energy is 0.69 eV. The melting point is ≈1250 K [22,28] and the energy of the intrinsic stacking fault on a {111} plane is estimated as 20–36 mJ m^{-2} [22,27–29]. Although newer interatomic potentials for copper exist [30] they reproduce nearly the same behaviour. The close similarity of the potentials is obvious from the comparison of calculated lattice properties, defect energies and values of planar fault energies, see [27] and [30] for further particulars. However, in contrast to the embedded atom type potentials [30] with non-analytical embedded function the Finnis–Sinclair type one [27] is computationally cheaper because of simple analytical form of the embedded function. The pair part of the potential used was modified for small interatomic separation by fitting to the Universal Screened Coulomb repulsion function for Cu–Cu interactions [31].

Four ambient crystal temperatures were considered, namely 100, 300, 600 and 900 K. A low temperature (such as 100 K) was used for most previous computer simulation studies, whereas the higher temperature range is more consistent with conditions met by metals in practical situations. The model crystals were equilibrated for about 50 ps prior to initiation of the PKA. The MD simulation box had cubic shape with {100} faces. It was maintained at constant volume, with lattice parameter a_0 set to the zero-pressure value for the chosen temperature (see Table 2 of Ref. [15]). Periodic boundary conditions with no energy/temperature damping were employed. The number of atoms in the box was chosen in accordance with the values of E_{PKA} and T, and is given in Table 1. The MD box size was larger than those used previously for a similar E_{PKA} and T condition, thereby leading to a smaller increase in lattice temperature once the PKA energy was dissipated. (The temperature increase did not exceed 80 K in any of the simulations.)

From 20 to 50 cascades for each PKA energy and ambient temperature were simulated in order to ensure statistical reliability of the results and obtain as many point defect clusters of various configurations, shapes and sizes as possible. The actual number of cascades is recorded in Table 1. PKAs were introduced along high-index crystallographic directions such as <123>, <234>, <345>, <135> and <112> in order to avoid recoil-atom channelling events, which are quite possible especially at low T and high E_{PKA}. (To study channelling by MD it would be necessary to use boxes with unreasonably large numbers of atoms. However, a channelled atom experiences a low rate of energy loss until it is defocused and so channelling would not have a significant effect on the data reported here. This would not be the case at higher energy where sub-cascade formation becomes dominant.) PKAs were created at different sites in the box at different times so that (quasi-)random spatial and temporal PKA distributions were simulated. The centre of the simulation box was repeatedly shifted to the centre of gravity of kinetic energy of the constituent atoms. Furthermore, the likelihood of a cascade extending beyond the box boundary and re-entering via periodic conditions was minimised. This is reflected in the small number of simulations abandoned in this research: only seven events in more than 450 simulations had to be stopped due to adverse channelling. It also lessened the selectivity effect in which there is a tendency for