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We present results of combined molecular dynamics (MD) and kinetic Monte Carlo (KMC) simulations of damage accumulation in Cu and Fe under conditions relevant to Inertial Fusion Energy (IFE). The MD simulations provide information on the primary damage state for cascades at energies from 100 eV to 50 keV. These results, in the form of a distribution of point defects and clusters, is used as input for a KMC simulation in which the pulsed nature of the IFE irradiation environment is simulated. The MD collision cascades were randomized and introduced into the KMC simulation box at a rate that reproduces the recoil spectrum of 14 MeV neutrons. In the KMC simulation, defects migrate with kinetics dictated by their diffusivities, and interact with each other, with clusters, and with impurities. The system evolves in time and space, and we discuss the manner in which this damage accumulates depending on temperature and pulse frequency. We show that near room temperature there is no effect of the pulse rate for values greater than 10 Hz. However, for rates of 1 Hz the effect is noticeable and reduces the rate of damage accumulation significantly. Although the pulse effects are similar in both materials, the controlling mechanisms are different.

1. INTRODUCTION

Radiation damage production and accumulation in solids can be divided into two stages. In the production stage, the impinging particle gradually gives off its kinetic energy to lattice atoms in the form of energetic recoils. These deposit their energy by generating secondary and higher order recoils that result in a displacement collision cascade. The outcome of this stage, of the time scale of a few to 100 piscoseconds, is a population of point or clustered defects known as the primary state of damage. In the second stage, which can extend over seconds, defects that survive recombination within their nascent cascade migrate over long distances, interacting with the microstructure. These freely migrating defects (FMD) are responsible for the changes in the macroscopic properties of metals under irradiation, such as void swelling, embrittlement, radiation enhanced diffusion, etc. Such changes in mechanical properties are most often detrimental and severely limit the flexibility in materials choice and operating temperature when designing a fusion power plant.

Under most conditions, such as those that would be present in a magnetic fusion energy plant, or when bombarding with fission or spallation neutrons, irradiation takes place at a certain dose rate and temperature, but in a continuous manner. However, in an Inertial Fusion Energy (IFE) reactor, or when using a pulsed neutron source such as that recently proposed by Perkins [1], the irradiation flux is pulsed and the interplay between temperature, flux and pulse frequency controls the kinetics of damage accumulation. For sufficiently low pulse frequency, and at elevated temperature where the defects migrate fast, it may be expected that annealing between pulses may result in a significantly decreased rate of damage accumulation compared to that seen under steady state conditions. On the other hand, very high neutron fluxes in the pulse itself may severely limit recombination therefore leading to extremely fast rates of damage accumulation even at elevated temperatures.

In order to understand these effects and elucidate the role and interplay of intrinsic (material type) and extrinsic (temperature, pulse, flux, fluence) variables on damage accumulation, one needs a theoretical description that can span all relevant length and time scales. In this paper, we describe a hybrid method where we combine molecular dynamics (MD) and kinetic Monte Carlo (KMC) simulations to accomplish that goal. We present results of a study of damage accumulation in Cu and Fe resulting from pulsed irradiation with 14 MeV neutrons at pulse frequencies of 1, 10, and 100 Hz. The simulations are carried out at two temperatures, one just above annealing stage III where vacancies are mobile but vacancy clusters are stable, and the other above stage V where vacancy clusters dissolve. In addition, we present results for continuous irradiation at realistic fluxes for comparison to the pulsed case.

In the next section, we briefly describe the MD and KMC simulation methods. In Section 3 we present results of the MD simulations of damage production in Cu and Fe, and in section 4 we discuss the results of the KMC simulation of damage accumulation. Section 5 presents the conclusions of the paper.

2. METHODS

Simulations of high energy displacement cascades, and of defect energetics and kinetics, were carried out with the MD simulation code, MDCASK [2]. In a MD simulation, the phase space trajectories of an ensemble of atoms are calculated by solving the classical equations of motion. These atoms interact via a prescribed interatomic potential which is used to obtain the forces. The code runs in parallel under MPI on any number of platforms including a Cray T3E at the National Energy Research Supercomputer Center in Berkeley, and at LLNL's 80 node DEC compass cluster and 512 node IBM SP2 computer (ASCI Blue). The Langevin equation of motion [3] is applied to the atoms in the link cells adjacent to the cell boundaries in order to control the temperature of the crystal.

The use of KMC to model defect diffusion during irradiation of materials has been rather sparse in the past, but the technique dates back over thirty years and was discussed extensively by Beeler [4]. The earliest reference to this method is the work of Besco in 1967 [5]. Doran [6] and Doran and Burnett [7] carried out short-term annealing simulations of displacement cascades in fcc and bcc Fe, respectively, using a Beckman 2133 analog computer and a PDP-7 with 8K of memory. More recently, Heinisch et al. [8-10] used a Monte Carlo code named ALSOME to model the migration, agglomeration and dissociation of the defects produced by 25 keV Cu self-irradiation at different temperatures. Within our own group, we have used our KMC code, BIGMAC, to model defect escape from cascades in Fe [11], V [12], Au [13] and Al [14] as well as to compare damage accumulation in Cu and Fe under continuous irradiation conditions [15].

Recently, we have described the BIGMAC code in detail [15,16]. In summary, BIGMAC is a computationally efficient KMC program that tracks the locations of defects, impurities, and clusters as a function of time. The starting point of these simulations is the primary damage state, i.e. the spatially correlated locations of vacancy and interstitials, obtained from MD simulations of displacement cascades. Each defect produced (including the clusters) has an activation energy for diffusion that can be extracted either from MD simulations or in some cases from experiments. The defects are allowed to execute random diffusion jumps (in one, two or three dimensions depending on the nature of the defect) with a probability proportional to their diffusivity. Similarly, dissociation rates from clusters are governed by a dissociation energy that includes the binding energy, E_b, of a particle to the cluster. The BIGMAC program requires input tables of D_0 and E_m for all mobile species, as well as the pre-factors and binding energies E_b for all possible clusters. The input tables can become rather large, but the program is very flexible, as only the input tables need to be changed to study another set of conditions or even another material system.

Input data on defect properties for Fe and Cu used here have been discussed in great detail in a recent publication [15] and will not be detailed here. Suffice it to say that

single self interstitial atoms (SIAs) and vacancies are mobile in both Fe and Cu at all temperatures studied here. In addition, SIA clusters in Fe are mobile in one dimension with an activation energy of 0.06 eV as determined by Soneda and Diaz de la Rubia [17], but get trapped by interstitial impurities with a binding energy of 1.0 eV. In Cu, SIA clusters smaller than 60 are considered mobile in one dimension with an activation energy of 0.1 eV. In Cu, interstitial impurity concentrations are expected to be low and the binding of SIAs to substitutional impurities should be small. Therefore, no trapping of SIA clusters by impurities in Cu was considered.

3. RESULTS

Figure 1 shows the time evolution of the defect distribution during a 30 keV displacement cascade in Cu. Within one picosecond, the cascade core region melts and a large pressure wave is emitted. In addition, a few replacement collision sequences (RCSs) lead to transport of SIAs away from the cascade core. These processes have been discussed in detail recently by Averback and Diaz de la Rubia [18]. Figures 1c and 1d show contour plots of hydrostatic stress at two different times for the same cascade. Clearly, the molten cascade core is under a state of tensile stress corresponding to the transient decrease in density associated with ballistic ejection of SIAs and with local melting. The average temperature in the cascade core at these early times exceeds the melting point of copper (1360K) over a region of approximately 2-5 nm in radius depending on recoil energy.

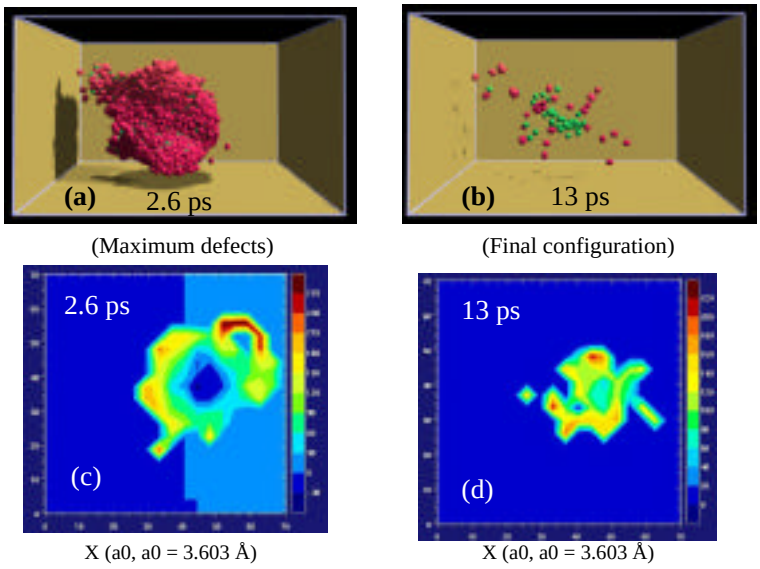

Figure 1. Snapshots of damage evolution of a 30 keV recoil of Cu in Cu as obtained from MD simulations at (a) 2.6 ps and (b) 13ps. Vacancies are in green, interstitials in red. Hydrostatic pressure around the cascade core at 2.6 ps (c) and 13 ps (d).

In Figure 1b, the final defect distribution from a 30 keV cascade in Cu is shown. It consists of monovacancies, vacancy clusters in the cascade core, single SIAs ejected by RCSs, and interstitial clusters. At these recoil energies 50% to 75% of all defect produced are typically found in clusters. It is also important to note that in contrast in Fe very few vacancy clusters are produced in these cascades.

The total number of defects produced in these cascades has been shown to be in very good agreement with experiment [18] and is approximately 1/3 of that predicted by the modified Kinchin-Pease model of Sigmund [19] and Norget, Robinson and Torrens (NRT) [20].

Each individual cascade results in a distribution of defects, some of which may be mobile depending on their properties and irradiation temperature. Because the KMC method takes into account all possible defect reactions at the correct rate depending on temperature, the simulations are ideal for evaluating the effects of pulsed irradiation on damage accumulation. After each pulse, the KMC computation allows the mobile defects to migrate and interact with other defects and traps for a time given by the pulse frequency.

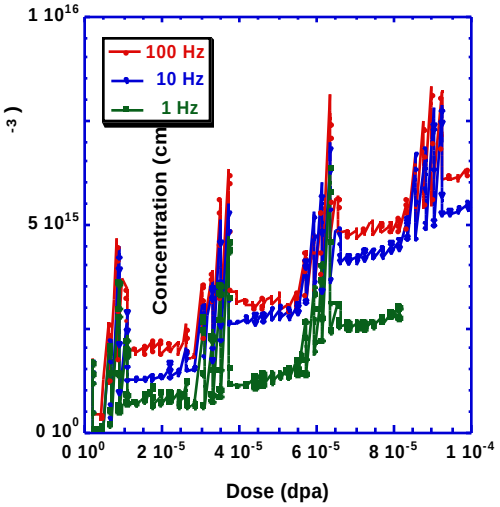

Figure 2. Concentration of Vacancy clusters as a function of dose in Cu for pulse frequencies of 1, 10 and 100 Hz, and for a continuous irradiation at a dose rate of 10^{-4} dpa/s.

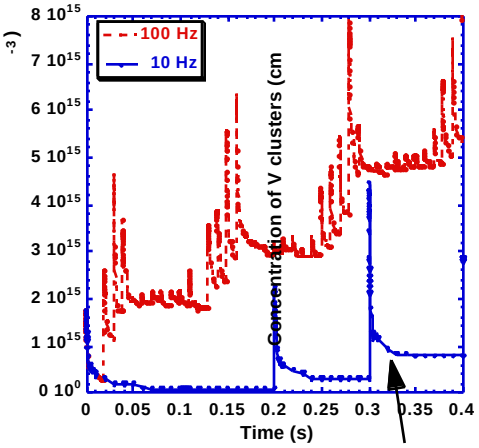

Figure 3. Concentration of V clusters in Cu as a function of irradiation time for 10 and 100 Hz.

The simulated dose rate corresponds to 10^{15} n/cm^2/s integrated over time. The dose rate in the pulse is of the order of 1.4 dpa/s in Fe, with a pulse length of a 1μs. At the pulse rate of 4 Hz, it produces a damage rate of 150 dpa/year. The overall conditions are intended to simulate those that would be encountered if a material were to be irradiated with the laser driven 14 MeV neutron source recently proposed by Perkins et. al. [1]. The recoil spectrum used was the calculated for the first wall of the magnetic fusion energy (MFE) reactor Starfire [21] with the SPECOMP/SPECTER code [22]. The probability function associated with each energy group and the energy groups of the spectrum were modified in two ways:

a) Electronic energy losses were subtracted from the primary knock-on atom (PKA) energy to include only damage energy. Robinson's approximation to Lindhard's theory was employed for this purpose [23].

b) The energy groups of the recoil spectrum were changed to the PKA energies available in our MD data base of recoils. The total energy deposited per neutron-iron collision was kept constant.

We should point out that the highest energy simulated by MD is 50keV recoils of Fe in Fe, while the spectrum includes recoils of energies higher than 300keV (after subtracting the electronic stopping). It is known that above 20keV cascades break up into subcascades of lower energy. The energy and spatial distribution of subcascades for recoils higher than 50keV was estimated using TRIM [24].

Each microsecond pulse was simulated with 6 cascades picked at random with probability of occurrence weighed by the modified recoil spectrum. That input rate in a KMC box of 300nm edge corresponds to 1.4 dpa/s in Fe and 2.2 dpa/s in Cu.

Figure 2 shows the evolution of the vacancy cluster population in Cu as a function of dose for 1, 10, and 100 Hz at a temperature of 340 K. Under these conditions, the rate of damage accumulation in Cu is not influenced by pulse frequency for frequencies above 10 Hz. However, the rate of damage accumulation decreases significantly at 1 Hz. To understand this, it is instructive to look at figure 3 where the time dependence of defect accumulation is plotted. Figure 3 reveals that immediately after the pulse, defect migration is so rapid that most of the damage anneals on a time scale of about 0.03 s. Since in Cu there are very few free vacancies and self-interstitial atoms (SIA) immediately after the cascade, most of this annealing takes place as a result of rapid recombination between

mobile SIA clusters and immobile vacancy clusters. In this recombination stage, mobile SIA clusters also coalesce and produce large immobile ones. At this temperature, only at 1 Hz are small vacancy clusters able to break and recombine with sessile interstitial clusters and agglomerate with other vacancy clusters, thus reducing the global cluster density. The time scale implies that at 1 Hz significant annealing can take place between pulses at this temperature, but not at 10 Hz or above.

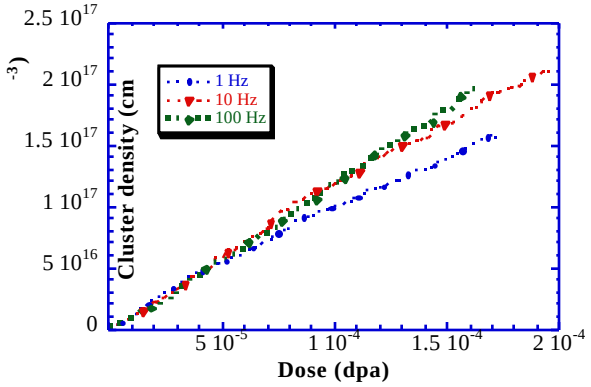

Figure 4. Vacancy cluster density as a function of dose for 1, 10 and 100 Hz

In Fe, the situation is somewhat different. Because bcc metals tend to contain large interstitial impurity concentrations, we carry out our simulation in the presence of 5 appm impurities. These impurities are treated as traps for SIA clusters (glissile dislocation loops) with a binding energy of 1.0 eV that arises from an elastic interaction. These impurities quickly immobilize the SIA clusters, thus limiting recombination even a 1 Hz. Figure 4 shows the damage accumulation in the form of vacancy clusters in Fe as a function of dose.

Figure 5 shows the damage production during the pulse and its recovery between pulses. A detailed analysis of this curve provides explains the damage accumulation trends in figure 4. The initial descent in figure 5 (labeled as 1 in figure 5) is a consequence of vacancy recombination with fast mobile SIA clusters. Once the SIA clusters have either disappeared or been trapped by impurities, a slower process takes place. Free vacancies diffuse and either shrink interstitial clusters or agglomerate in vacancy clusters (The slope labeled as 2). This second stage takes place after 0.5 s annealing, therefore the most pronounced difference in vacancy clustering is for a pulse rate of 1 Hz (Figure 1). In short, the higher the pulse rate, the smaller the vacancy clusters and the higher the number of free vacancies. The pulse annealing in copper is slightly different. The cluster density drop in stage 1 is sharper due to the lack of impurities and the slope in stage 2 is small, due to the small number of free vacancies.

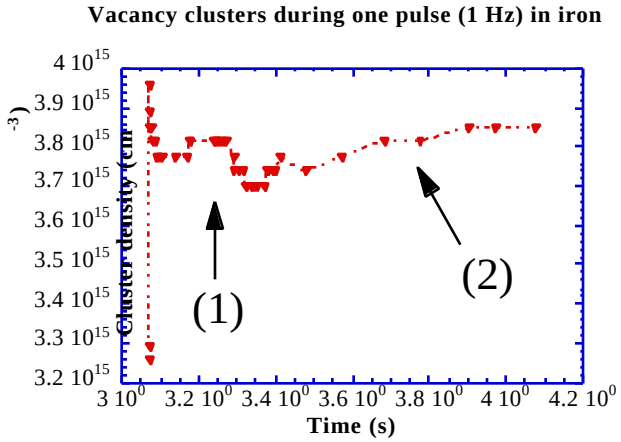

Figure 5. Vacancy cluster concentration in Fe as a function of annealing time for 1 Hz. At short times (1) recombination with glissile interstitial clusters takes place. At ~ 0.4 s after the pulse all glissile interstitials are trapped and vacancy clusters grow (2) by V-V recombination.

The effect of temperature on vacancy clustering in iron is similar to that of pulse rate. At high temperature (Stage V), the rapid migration of vacancies makes them cluster at a faster rate, so their overall number of clusters is smaller but their size is greater.

SUMMARY

We present a study of pulsed irradiation under conditions relevant for IFE in bcc and fcc metals. We analyze the influence of pulse rate and temperature in the context of defect production and clustering. In both cases we observe that the influence of pulse

rate is clearly noticeable at 1 Hz, the lowest pulse rate studied, but not a higher rates. Damage recovery occurs almost instantaneously after the pulse due to fast SIA cluster migration. This recombination stage anneals out most of the damage in copper but it is rather incomplete in iron due to the presence of traps. The second annealing stage is governed by vacancies. An increase in vacancy clustering is observed in iron while the slope is flat in copper. We ascribe this difference to the high population of free vacancies in iron. It is noteworthy that even at the low temperatures studied, the greatest differences in damage accumulation are between 1 and 10 Hz, which is the rate bracket where future IFE devices will operate. Therefore, choosing the right pulse rate might have beneficial consequences for the first wall of these reactors.

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