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Vacancy defects in Fe: Comparison between simulation and experiment

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ABSTRACT

The evolution of radiation damage under heavy-ion irradiation in thin foils of pure bcc Fe has been investigated by simulation and experiment. Simulations showed that vacancy loops are about as mobile as interstitial loops, and can be lost to the surface of a foil. Consistent with this, *in situ* real-time dynamic observations of the damage evolution showed that loops, many of which are believed to be of vacancy nature, were mobile and were often lost during irradiation. Atomistic simulations of vacancy defects in Fe showed that spherical voids, rather than vacancy loops, represent the lowest energy configurations for clusters of vacancies of any size. The simulations also indicated that the stability of loops strongly varies depending on their size. Closed loops above a critical diameter (~2 nm) are highly metastable due to the difficulty of their transformation into voids. The greater stability of voids explains why the loop yield in Fe and other ferritic materials is very low.

1. Introduction

Ferritic–martensitic steels are candidate materials for structural applications in fusion power plants and for other advanced nuclear applications, where they will suffer from radiation damage caused by fast neutrons. The accumulation of this radiation damage may adversely affect the structural integrity of the materials, leading to a range of deleterious effects, including, for example, low-temperature hardening [1]. There is therefore an urgent need to understand the formation and behaviour in ferritic materials of vacancy and interstitial clusters, which constitute the accumulated damage produced by irradiation-induced displacement cascades.

In order to address this need, we have carried out studies of the relative stability and mobility of vacancy and interstitial clusters in Fe, which was chosen as the simplest model ferritic material. Simulations were performed using Molecular Dynamics (MD) and Molecular Statics on lattices of pure Fe with periodic boundary conditions. The interactions between atoms in the simulations were described by the recently developed 'magnetic' many-body magnetic interatomic potential [2]. In parallel we have performed *in situ* transmission electron microscope (TEM) experiments to study the evolution of radiation damage under heavy-ion irradiation in thin foils of pure Fe, where the incident ions mimic the primary knock-on atoms produced in neutron irradiation. These experiments were carried out using the Argonne IVEM-Tandem Facility which consists of a Hitachi H-9000NAR transmission electron microscope interfaced to a 2 MV tandem ion accelerator and a 0.65 MV ion implanter, only the latter of which was used in the present experiments [3]. Specimens of Fe of different purities, as well as various FeCr alloys, were irradiated with 100 or 150 keV Fe^+ and Xe^+ heavy ions at room temperature (RT) and 300 °C (573 K) to doses up to 2×10^{18} ions m^{-2}.

This paper reports on results from these two parallel investigations, focusing on those elements which allow for a direct link to be drawn between the two. In particular, we consider the vacancy clusters which are present in thin-foil irradiations. At the low doses of the present experiments many if not all of the clustered defects are likely to be vacancy in nature. Interstitial damage is less commonly seen in thin foils at low doses because the expected high mobility of single interstitials and small clusters causes most of them to be lost from the foil, although interstitial clusters are present at higher doses [1]. A more detailed description of the present results can be found in [4] for the atomistic simulations, which also consider tungsten, and in [1] for the experiments, where full results from irradiations of FeCr alloys are also presented.

2. Mobility of vacancy dislocation loops

In thin-foil experiments the vacancy or interstitial nature of very small clusters is difficult to determine. However, the so-called black–white (B–W) contrast technique does allow for the nature of some loops lying close to the foil surface to be determined. When

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imaged under two-beam dynamical imaging conditions loops lying within layers close to the surface may exhibit B–W contrast, characterised by a B–W contrast vector **l**. The technique relies on the direction of **l** relative to the diffraction vector **g** and a determination of the depth of the loop in the foil (see [5] for full details of the technique). An experiment was performed in Fe-11%Cr irradiated with 30 keV Ga^+ ions. Because of the low energy of the ions, it was expected that most loops would be produced close to the surface, within the so-called first depth layer. It was found that all loops which exhibited strong B–W contrast had the same direction of **l** and were of vacancy nature, if they lay in the first depth layer. Whilst we have not yet analysed loops in pure Fe irradiated with Fe^+ or Xe^+ ions, it seems likely that a proportion of the loops observed in thin foils are also vacancy in nature in these cases.

MD simulations such as those by Calder and Bacon [6] have demonstrated that interstitial clusters can form directly in cascades, and so it is widely held that the reason for the absence or scarcity of interstitial loops in experiments such as ours is that such clusters are highly mobile, and so most are lost from the foil, either via migration to surfaces or by recombination. However, this prompts the question as to how mobile are vacancy clusters relative to interstitial clusters.

In the present work cluster mobility was investigated using long-timescale (in MD terms) simulations of equivalently-sized interstitial and vacancy loops in pure bcc Fe. For each loop, 2 ns simulations were performed at temperatures ranging from 100 to 800 K. The position of the loop was measured at 0.1 ps intervals throughout each simulation and then the diffusion coefficient D was evaluated by taking the statistical average over many subsets of the full set of positions. A typical trajectory is shown in Fig. 1 for a vacancy loop with Burgers vector $1/2\langle 1\,1\,1\rangle$ at 700 K. Here the position of the vacancy loop was determined by averaging the positions of the atoms that bounded the loop and therefore had energies significantly divergent from that of bulk atoms. In the case of interstitial loops, the loop position was the average of the atoms making up the loop. Fig. 2 shows the variation of diffusion coefficient D with temperature for $1/2\langle 1\,1\,1\rangle$ loops of diameter 1.9 nm (comprising 61 vacancies or interstitials). Note that the trend lines drawn in the graph here, and elsewhere in the paper, are continuous interpolations of the discrete data.

Fig. 2 shows that interstitial loops are systematically more mobile than vacancy loops at the same temperature. However, the results also indicate that vacancy loops do diffuse at rates of the same order of magnitude as interstitial loops. A similar pattern was observed in simulations of 3.1 nm diameter $\langle 0\,0\,1\rangle$ loops (185 vacancies/interstitials), although in this case the difference between the two loops types was less pronounced. The simulations also indicated that a $1/2\langle 1\,1\,1\rangle$ vacancy loop could be lost to a surface on a timescale of 100 ps once it approached the surface closer than 5 nm. Therefore, in experiments, it is probable that both interstitial and vacancy dislocation loops are lost to surfaces. This will be more significant for interstitial loops due to their higher mobility and hence higher escape probability.

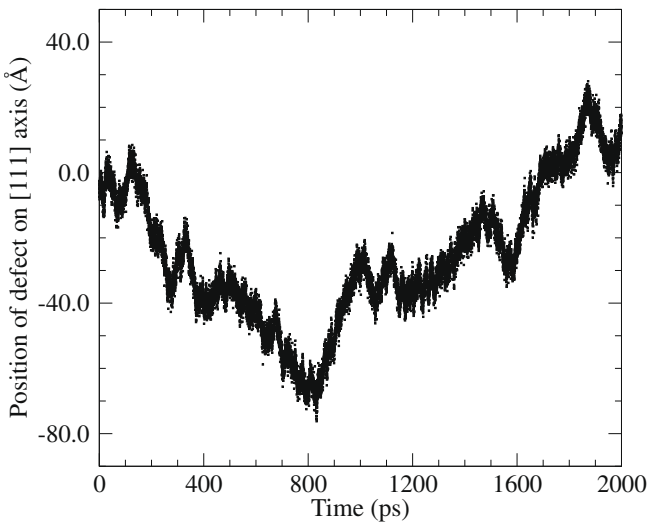

Fig. 1. Defect trajectory of a 1.9 nm diameter $1/2\langle 1\,1\,1\rangle$ vacancy loop (61 vacancies) during 1 D vertical motion in the [1 1 1] direction in a MD simulation at 700 K. Loop position marked every 0.1 ps.

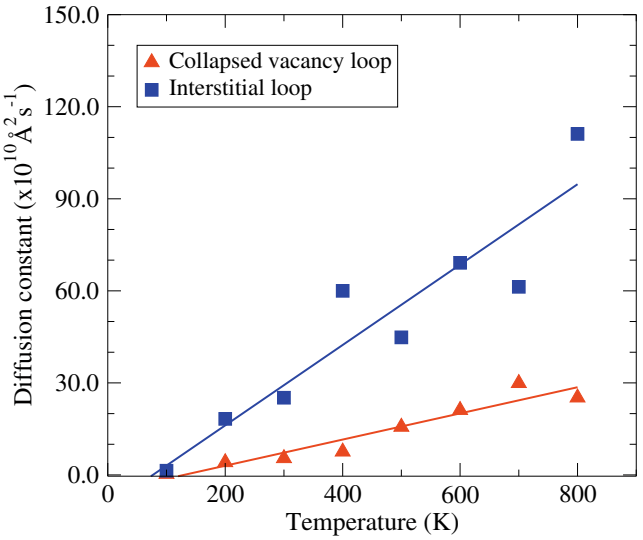

Fig. 2. Plot of calculated diffusion coefficients as a function of temperature in Fe for 1.9 nm $1/2\langle 1\,1\,1\rangle$ (61 vacancies/interstitials) interstitial and vacancy dislocation loops. Here 'collapsed vacancy loop' is an alternative description of a vacancy dislocation loops.

In some of the *in situ* experiments is was possible to perform dynamic observations using a CCD camera connected to a video (DVD) recorder, which allowed recording of images during and immediately after irradiation. The motion of individual loops was observed in all materials, including FeCr alloys, although it was most pronounced in Ultra-High Purity (UHP) Fe. The motion consisted of sudden, discrete hops over distances of several nanometres or more, and occurred both during irradiation and afterwards under observation in the electron microscope and at both RT and 300 °C.

Fig. 3(a) shows an example of a loop in UHP Fe which moved one-dimensionally over the course of three image frames. This motion occurred under electron illumination alone with the specimen at 300 °C. The direction of hopping is parallel to the projection of the $1/2\langle 1\,1\,1\rangle$ Burgers vector. The time spacing between frames is approximately 34 ms and a clearly visible hop occurs between the first and second frames. From this observation, and many others like it, it is clear that many of the loops formed under irradiation are mobile. Whilst this does not confirm directly the MD results discussed above, because the nature of the hopping loops is uncertain, there is a significant probability that some of the mobile loops were vacancy in nature. In addition, we have also observed that in some cases the loops formed during ion irradiation were lost from the foil, as demonstrated by the sequence of images shown in Fig. 3(b), in which a loop disappeared over the course of three frames in UHP Fe in the same specimen as Fig. 3(a). These losses occurred both during and after irradiation, and may (but not necessarily) be caused by glide of the loop to the surface [1]. The timescale of loss was longer than in the atomistic simulations of migration to surfaces, but if escape to surfaces is the preferred