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Molecular dynamics simulation of displacement cascades in Fe–Cr alloys

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Abstract

An embedded atom method (EAM) empirical potential recently fitted and validated for Fe–Cr systems is used to simulate displacement cascades up to 15 keV in Fe and Fe–10%Cr. The evolution of these cascades up to thermalisation of the primary damage state is followed and quantitatively analysed. Particular attention is devoted to assessing the effect of Cr atoms on the defect distribution versus pure Fe. Using the Wigner–Seitz cell criterion to identify point defects, first results show that the main effect of the presence of Cr in the system is the preferential formation of mixed Fe–Cr dumbbells and mixed interstitial clusters, with expected lower mobility than in pure Fe.
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1. Introduction

The development of models to assess the mechanical stability under intense neutron irradiation of high-Cr reduced-activation ferritic/martensitic (RAFM) steels is an important part of fusion reactor materials research. The starting point for any neutron-irradiation-damage modelling effort is the study of the primary damage state produced by displacement cascades in the relevant material. Molecular dynamics (MD) is well known to be the simulation tool 'par excellence' for the study of displacement cascades, provided that a valid and adequately stiffened many-body interatomic potential is available for the system of interest [1]. In the past, much work has been done on MD simulation of displacement cascades, using pure Fe as model alloy for steels, described by a variety of interatomic potentials [2–6]. However, to take a step forward toward modelling real engineering materials, the assessment of the effect of Cr, the main alloying elements, in RAFM steels, on the primary damage state is of critical importance. Thus, this paper reports a first set of results on displacement cascades initiated by Fe recoils up to 15 keV in Fe–10%Cr. The simulations have been performed using a recently fitted and validated many-body potential for the Fe–Cr system [7,8]. For comparison, cascades in pure Fe were simulated as well, using the same potential. The objective of the reported work is (i) to further validate this potential by comparing the obtained results with earlier work for pure Fe, (ii) to analyse and discuss the main effects of the presence of Cr in terms of primary defect population.

2. Simulation method

The details of the fitting procedure and validation of the embedded atom method (EAM) [9] interatomic potential for Fe–Cr used in this work can be found elsewhere [7,8]. Briefly, the Fe–Fe potential was fitted following the approach described in Ref. [10] and stiffened using the same method as in Ref. [2]. The Cr–Cr potential was taken from the literature [11]. The Fe–Cr crossed pair contribution was fitted, using the procedure

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described in [12], to the experimental values of bulk modulus (1544 kbar), cohesive energy (−4.262 eV) and lattice parameter (2.866 Å) of the Fe–10%Cr alloy, as well as to the mixing enthalpy for the same alloy (5.16 meV), calculated by ab initio methods to take into account the effect of ferromagnetism [13]. This potential has proven to provide a reasonably good description of the interaction between Cr atoms and point defects in a ferritic matrix compared to ab initio results obtained with the VASP code [8,14,15]. In particular, the stability of the Fe–Cr and Cr–Cr dumbbells is correctly reproduced, as well as the negligible binding energy of Cr atoms with vacancies [8]. In addition, the description of Cr diffusivity in Fe is also in good agreement with experimental results [16,17]. The main limitation is that this potential should not be used in the range of compositions where α'-phase nano-segregation is expected.

The potential was implemented in the classical MD code Dymoka, which is suitable for the simulation of displacement cascades [6]. Prior to initiating the cascade, a block was equilibrated for 1 ps at 300 K. This atom block was then used as starting point for the cascade simulation and defect detection. The cascade was started by imparting a kinetic energy E_{MD} to the selected primary knock-on atom (PKA) along a high-index direction $\langle 1\,3\,5\rangle$, in order to avoid channelling [2,5]. Cubic box size, simulated time and number of cascades versus E_{MD} are summarised in Table 1. Following common practice [1–3,5,6], no rigorous attempt was made to control the temperature of the system and all presented results were obtained working in the NVE microcanonical ensemble with periodic boundary conditions [3,4,6]. Only in the case of high recoil energy (10 and 15 keV) were the boundary atoms damped using the rescaling velocity algorithm, to partially extract heat. It is anyhow accepted that the simulation temperature scarcely influences the defect population produced in displacement cascades in Fe [18,5]. The evolution of the cascades was monitored using appropriate visualisation tools. The final atomic configuration was then analysed to detect and count defects, using a Wigner–Seitz cell method: an empty cell corresponds to a vacancy, two atoms in the same cell correspond to an interstitial configuration. Replacements were also accounted for and the number of displaced atoms was defined as the sum of replaced and interstitial atoms. Clusters were defined using a third nearest neighbour (nn) criterion for interstitials and second nn for vacancies.

3. Results and discussion

3.1. Defect number

In Fig. 1(a) the average number of surviving Frenkel pairs at the end of the cascade (N_{FP}) in both pure Fe and Fe–10%Cr resulting from the MD simulations is plotted as a function of E_{MD}. The points have been interpolated using the empirical power law proposed by Bacon et al., $N_{FP} = A \cdot E_{MD}^{m}$ [1], whose validity has been determined to cover the range from about 1 to 20 keV [5]. Prefactors and exponents obtained in a least-square fit are in close agreement with previous work on Fe [1,5].

Traditionally, to estimate the number of Frenkel pairs produced per cascade the NRT formula is used [19,20]: $N_{NRT} = 0.8E_D/2E_d$, where E_d is the average displacement energy for all crystallographic directions and E_D is the damage energy. E_D is the fraction of recoil energy that goes into displacive damage, after subtracting the portion dissipated for electron excitation. Since in the present MD simulations the interaction between ions and electrons is not included, it is assumed that $E_D = E_{MD}$, as is customarily done [1]. The defect production efficiency is defined as the ratio of the MD surviving Frenkel pairs to the number of NRT displacements ($N_{FP}N_{NRT}$). For the sake of simplicity and following Ref. [20], we took $E_d = 40$ eV to estimate the number of defects with the NRT formula for both pure Fe and Fe–10%Cr. In both cases (Fe and Fe–Cr) the defect production efficiency decreases with recoil energy down to a more or less asymptotic value of about

Table 1
Summary of number of cascades, simulation time and box size versus E_{MD}

E_{MD} (keV)	No. of cascades		Other parameters	
	Fe	Fe–10%Cr	Time (ps)	Box size (atoms)
0.5	10	10	10	54 000
0.7	10	10	10	54 000
1	10	10	10	54 000
2	10	10	10	128 000
5	10	10	20	128 000
8	10	10	20	250 000
10	10	10	30	432 000
15	5	10	30	432 000

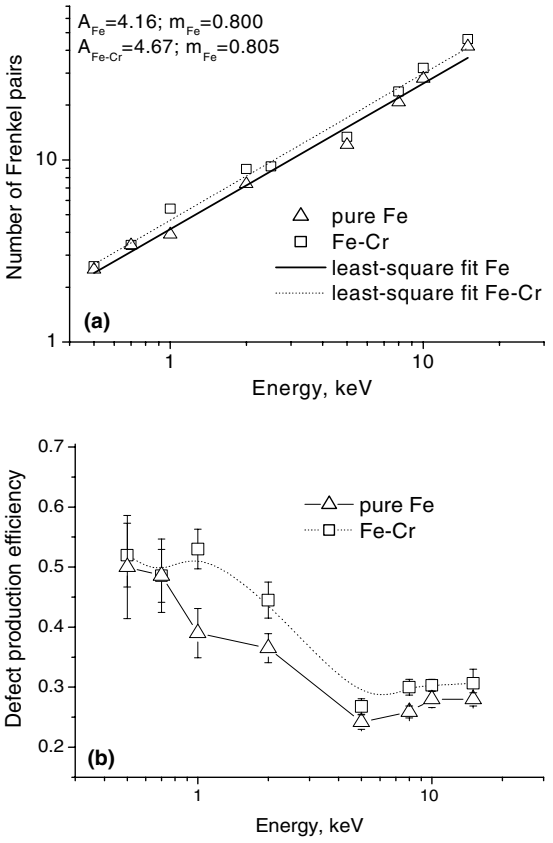

Fig. 1. Number of surviving Frenkel pairs (a) and defect production efficiency compared to NRT (b) at the end of the cascade versus recoil energy, in Fe and Fe–10%Cr. In (a) the power law proposed by Bacon et al., $N_{FP} = A \cdot E_{MD}^{m}$ [1] has been used for interpolation.

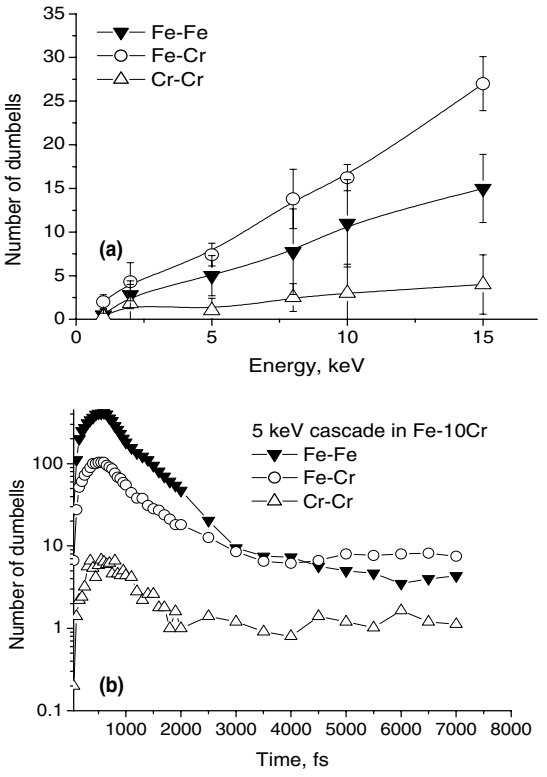

Fig. 2. Number of Fe–Fe, Fe–Cr and Cr–Cr dumbbells in Fe–10%Cr cascades versus (a) recoil energy and (b) time in 5 keV cascades (average evolution of 10 cascades).

0.3 (0.28 in Fe and 0.31 in Fe–Cr), in agreement with previous work [1,5,6] (see Fig. 1(b)).

A close inspection of Fig. 1 reveals a slightly higher defect production when Cr is present. A study of the threshold displacement energies of a Cr atom in a ferritic matrix along the three main crystallographic directions showed that only in one direction, $\langle 1\,1\,0 \rangle$, the threshold is 5 eV lower than for Fe atoms, independently of the Cr concentration [21]. Thus, the resulting slightly lower average threshold energy for Cr atoms cannot alone explain the somewhat higher defect production efficiency in Fe–10%Cr. It is more likely that the higher production results from stable Fe–Cr and Cr–Cr dumbbell formation during cascade cooling, which limits recombination. Indeed, in agreement with ab initio calculations [8] the present potential predicts a positive binding energy for Fe–Cr and Cr–Cr dumbbells (larger than 0.3 eV). This feature leads to the formation of a number of mixed dumbells that is remarkably higher than the number of Fe–Fe dumbells at the end of the cascade, as is shown in Fig. 2(a). Independently of the recoil energy, 67% (±6% of standard deviation) of the final dumbbells contain Cr atoms, to be compared with the 10% Cr concentration of the alloy. Fig. 2(b) shows clearly, in the case of 5 keV cascades, that the formation of mixed dumbbells is a post-collisional-phase phenomenon. Initially, as is to be expected, more Fe atoms than Cr atoms are displaced and counted as interstitials. However, during the cooling stage most of these Fe–Fe dumbbells, possibly assisted by the still high temperature, glide till they become trapped at the closest Cr atoms, thereby determining a cross-over of the curves of the number of mixed and self-interstitial dumbbells versus time in Fig. 2(b). The same process has been seen to occur for all cascade energies investigated in this work.

In summary: Cr atoms do not appear to significantly affect the collisional stage of the cascade, as may be expected, considering the negligible difference in mass between Fe and Cr atoms. Yet, the presence of Cr determines a redistribution of dumbbell species during the post-collisional stage, where most of the dumbbells end up containing Cr atoms, in percentage far higher than the Cr concentration in the alloy. The thermal stability of these dumbbells is likely to be responsible for

the somewhat reduced recombination observed during the cooling phase of Fe–10%Cr cascades, as compared to pure Fe.

3.2. Cluster formation

All previous papers on displacement cascades in Fe agree on that very little in-cascade vacancy clustering occurs in this material, while sizeable interstitial clusters are found to form [1,2,5,6]. Our results agree with these findings, as is shown in Fig. 3(a) and (b), where the fraction of, respectively, surviving vacancies and interstitials in clusters (containing at least two elements) is plotted versus E_{MD} for both Fe and Fe–10%Cr. We find that the average fraction of clustered vacancies hardly exceeds 30%, the mean cluster size being rather small (2–3 vacancies). Conversely, up to 50% of the interstitials are found in clusters, of sizes up to 11. These results, allowing for the large scatter, are in broad agreement with most previous work [1,2,4–6], provided that the same criterion is used for the definition of clusters (second nn for vacancies, third nn for interstitials).

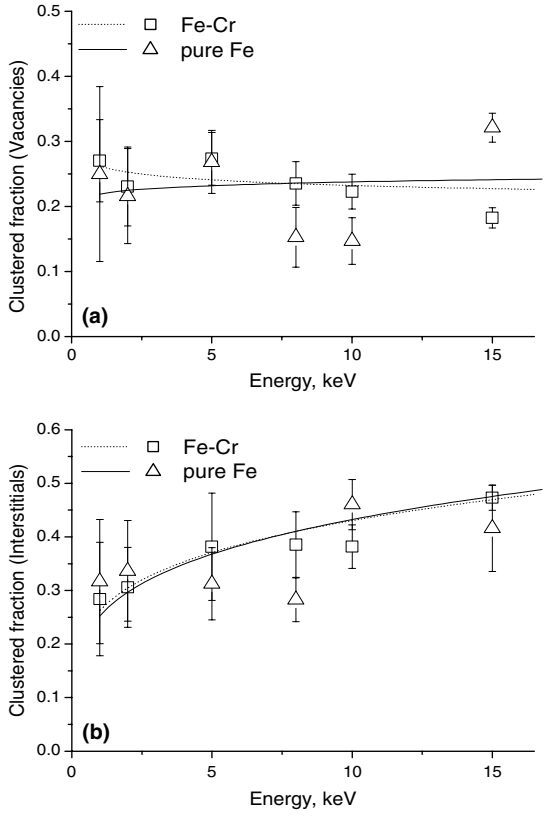

Fig. 3. Fraction of defects in cluster at the end of the cascade, in both Fe and Fe–10%Cr, versus recoil energy: (a) vacancies (2 nn criterion); (b) interstitials (3 nn criterion). The lines represent trend power.

The presence of Cr does not seem to influence the fraction of defects in clusters. Also the cluster size distribution is comparable in both materials. No detailed analysis was conducted to detect a possible preferential association of vacancies and vacancy clusters to Cr atoms, but this is not likely to be a large effect, due to the very low Cr–V binding energy. On the contrary, a large portion of the interstitials in cluster in Fe–10%Cr are Cr atoms. Although further investigation is required, the promotion of the formation of mixed interstitial loops, stabilised by the presence of Cr, will have a large impact on the subsequent evolution of radiation damage if, as one should expect, their mobility is lower than in pure Fe. A first simulation study of single interstitial diffusivity in Fe and Fe–Cr has shown a decrease of the interstitial diffusion prefactor by about one order of magnitude, due to the presence of Cr atoms [16]. The experimental observation of slowing down of interstitial loop motion in Fe due to the presence of Cr is also reported in these proceedings [22]. Moreover, Okada et al. observed, in neutron and electron irradiation experiments performed between 200 and 500 °C, that the addition of even small percentages (0.1 at.%) of Cr to ultra-pure Fe induces more frequent nucleation of small loops of interstitial nature than in ultra-pure Fe [23]. These experimental findings seem to be correctly reflected by the results of our simulations.

4. Summary and conclusions

Displacement cascades up to 15 keV recoil energy were simulated by molecular dynamics in Fe and Fe–10%Cr, using a recently fitted and validated EAM many-body potential. The different phases of the cascades were monitored and analysed using ad hoc post-processing tools. In this paper, results concerning the number of surviving defects and their distribution in clusters have been reported. In the case of pure Fe, our results are consistent with previous ones. The presence of 10% Cr atoms does not seem to affect the collisional stage of the cascade, but it does determine a redistribution of dumbbell species during the post-collisional stage. Most interstitial atoms are Cr, in percentage far higher than the concentration in the alloy. This effect seems to reduce recombination, thereby leading to a slightly higher defect production in the alloy compared to pure Fe. No substantial difference is detected between Fe and Fe–10%Cr in the fraction of clustered defects. However, interstitial clusters contain a large percentage of Cr atoms which stabilise them. This feature may drastically reduce the mobility of interstitial loops in Fe–Cr compared to pure Fe, as confirmed by recent experiments, with significant impact on the subsequent evolution of radiation induced defects in the alloy.

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