Journal of Nuclear Materials 470 (2016) 119–127

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Non-equilibrium properties of interatomic potentials in cascade simulations in tungsten

A.E. Sand [a, *], J. Dequeker [b], C.S. Becquart [b], C. Domain [c], K. Nordlund [a]

[a] Department of Physics, P.O. Box 43, FI-00014, University of Helsinki, Finland
[b] Unité Matériaux et Transformations, UMET, UMR 8207, Université de Lille 1, F-59655, Villeneuve d'Ascq, France
[c] EDF-R&D, Département MMC, Les renardières, F-77818, Moret sur Loing, France

ARTICLE INFO

Article history:
Received 28 September 2015
Received in revised form
11 December 2015
Accepted 13 December 2015
Available online 15 December 2015

keywords:
Tungsten
Interatomic potential
Collision cascades
Molecular dynamics

ABSTRACT

The reliability of atomistic simulations of primary radiation damage hinges on the quality of the interatomic potential. However, irradiation induced collision cascades involve strongly non-equilibrium processes, and thus depend on properties of potentials not usually included in the potential fitting. Here, we compare the predictions of five interatomic potentials for tungsten in cascade simulations with primary knock-on energies ranging from threshold energies for defect production, up to 200 keV. The highest energies represent the energetic recoils induced by the 14 MeV fusion neutron irradiation. We further compare properties related to dynamic collisions predicted by the different potentials to DFT calculations, to assess the accuracy of these predictions. We also present two hardened versions of a recent EAM-type potential, and demonstrate explicitly the importance of carefully adjusting the range of the potential at interaction distances smaller than those included in the fitting of potentials to equilibrium properties.

1. Introduction

The formation of the primary radiation damage from collision cascades occurs on picosecond time scales, and is practically inaccessible by current experimental means. Hence simulations are an invaluable tool for predicting the magnitude and morphology of cascade damage. This primary damage is key to the subsequent microstructural evolution, and thus strongly affects the response of materials to radiation. Radiation effects in tungsten materials in particular are of current interest due to their planned use for both armor and structural components in future fusion reactors [1]. Tungsten exhibits several attractive properties, including high temperature strength and stability, resistance to erosion, and good thermal conductivity [2]. However, little is known of the effects of the 14 MeV fusion neutron irradiation on the mechanical properties of this already intrinsically brittle metal.

Primary radiation damage in W has been studied with molecular dynamics (MD) methods for some time [3,4]. However, large variations exist in the primary damage predicted by different interatomic potentials, especially regarding the clustering behavior of defects. As the efficiency of computers has grown, simulations of larger systems and higher primary knock-on atom (PKA) energies have become feasible. These have uncovered increasingly large discrepancies between the predictions of different potentials [5]. At the same time, the continuous development of new, more extensively fitted potentials offers possibilities for improved accuracy in simulations.

Collision cascades involve processes spanning a large range of temperatures, pressures, and states of the irradiated material, and the specific aspects of potentials that affect the formation of the primary damage is not known. Traditionally, the formation energies of point defects have been considered to be of paramount importance, and the use of density functional theory (DFT) calculations to predict such properties have made it possible to develop more accurate potentials. However, in combined BCA and MD studies, Becquart et al. [6] showed that the interactions at distances below those of the fitted part of a potential played a crucial role in the development of cascades. They introduced the parameters range R (the distance at which the interaction energy is equal to 30 eV) and stiffness S (the slope of the potential at that point) to describe the repulsive part of the potential at interaction distances relevant to collision cascades in Fe. These parameters influenced

* Corresponding author..
E-mail address: andrea.meinander@helsinki.fi (A.E. Sand).

http://dx.doi.org/10.1016/j.jnucmat.2015.12.012
0022-3115/

the length and frequency of replacement collision sequences (RCSs) in BCA simulations, and were clearly related to a difference in cascade morphology seen with different potentials in MD [6].

Most equilibrium potentials must be hardened for use in cascade simulations. This is typically done by joining the fitted "equilibrium" potential to a repulsive potential at short distances. The joining is done at distances below those included in the fitting procedure of the equilibrium potential. The form of the potential at such intermediate distances strongly affects the predicted threshold displacement energy (TDE), which can thus be used to tune the joining. However, in a wide survey of available data from MD cascades in Fe, Malerba [7] notes that the correct prediction of TDEs does not necessarily guarantee reasonable results from cascade simulations, as suggested already earlier by Becquart et al. [8]. A subsequent, systematic comparison of cascades in Fe [9] shows an effect of the range and stiffness of a potential at 30 eV interaction energy. More defects, attributed to a shorter recombination time, are seen for stiff repulsive potentials with a small R value.

Nevertheless, it is difficult to draw clear conclusions from these results, due to the large number of varying parameters involved in the different interatomic potentials which have been compared. Hence it has not been possible to ascertain with certainty which aspects of a given potential are responsible for differences in the predicted damage.

In this work, we have for the first time compared cascades simulated with two versions of the same potential, that differ only in the interval over which they are joined to the short-range repulsive potential. Each version gives reasonable TDEs, but the stiffness and range for 10–300 eV interaction energies is very different. Comparison of these potentials is made in relation to the performance of a number of established interatomic potentials, used frequently in studies of collision cascades. Furthermore, we compare a number of non-equilibrium properties of these potentials, which are related to dynamic collisions, including the energy transfer to target and surrounding atoms in head-on collisions in the lattice. Such properties are not typically used in the fitting of equilibrium potentials, nor when hardening them, yet they involve interactions over ranges which are directly relevant for collision cascade simulations. We compare the results to DFT calculations, both including and excluding semi-core electrons in the *ab initio* calculations.

2. Methods and analysis

2.1. Interatomic potentials

Many interatomic potentials for W exist today (see Ref. [10] for an extensive survey). The selection included in this work is not meant to reflect the quality of the various potentials. Rather, we have chosen to compare a number of established potentials which have frequently been used for cascade simulations, as well as new candidates for such simulations. We focus on EAM-type potentials due to their computational efficiency, which make them ideal for simulations of high-energy collision cascades requiring systems with millions of atoms.

The potentials we have considered here include the well-established Finnis-Sinclair (FS) potential [11] with modifications by Ackland and Thetford [12] and short range stiffening by Zhong et al. [3] (hereafter referred to as AT-ZN), the potential by Derlet et al. [13] with the repulsive part fitted by Björkas et al. [4] (hereafter denoted by DND-BN), the FS potential as recently modified by Juslin et al. (JW) [14], and one of the new potentials developed by Marinica et al. [15], stiffened for cascade simulations in two different versions in this work (denoted by M-S_s for a "soft" interpolation and M-S_h for a "hard" interpolation). The universal Ziegler-Biersack-Littmark (ZBL) potential [16] is used to describe the short range interactions for all potentials.

Table 1 lists some of the properties of these five potentials, compared to experimental predictions and *ab initio* values. Simulation cells with 38400 atoms, initially with half liquid and half crystal structure, were used here to find the predicted melting points of the potentials.

2.2. Stiffening of the M–S potential

In order to make the M–S potential suitable for cascade simulations, the pair interaction part $V(r)$ was first splined to the universal ZBL potential [16] for short ranges. The interpolation was done using a fifth degree polynomial $V(r)_{int} = a_0 + a_1 r + a_2 r^2 + a_3 r^3 + a_4 r^4 + a_5 r^5$, as in Ref. [4], with the following considerations: (i) the new $V(r)$ and its first and second derivatives must be smooth, (ii) no spurious minima should appear in the new $V(r)$, (iii) the electron density of the EAM potential should not interfere with the ZBL at short ranges, (iv) the formation energies of point defects should not be affected by the modification, and (v) the resulting full potential should yield reasonable TDEs. Since the original electron density function of the M–S exhibits a maximum at a distance close to 2 Å, it was allowed to saturate to that value for shorter distances.

Two different interpolations were found, which differ significantly from each other in the intermediate interaction range. The harder M-S_h resembles the other potentials, while M-S_s deviates clearly from the rest (see Fig. 1), with a softer outer shell down to a distance of 1.15 Å, and a stiffer core at distances closer than 1.15 Å. Table 2 gives the parameters used in the interpolations. These two versions make it possible to test explicitly the effect of the intermediate interaction range of the potential on cascade behavior, since all equilibrium properties of the two versions remain the same.

The TDE is the lowest PKA energy by which a stable defect can be created, and is often strongly anisotropic. This is especially the case in W [22]. The predicted TDEs of the potentials in the main crystallographic directions were determined here by the method described in Ref. [28]. In Ref. [6], the range R and stiffness S were

Table 1
Properties of different potentials for W, compared to experiment and *ab initio* values. Note that these properties are all identical for the two versions of M-S. The lattice constant a_0 (Å), cohesion energy E_{coh} (eV), melting point T_{melt} (K), maximum interaction distance R_{cut} (Å) (at separation distances beyond this the atoms do not see each other), vacancy formation energy E_v^f, interstitial formation energy for the ⟨111⟩ crowdion and ⟨100⟩ and ⟨110⟩ dumbells, and the vacancy migration energy E_v^m, in eV units. In some cases two values from different sources are given, to illustrate the level of uncertainty in the calculated values. [a] [4], [b] [17], [c] [18], [d] [19], [e] [20], [f] [21], [g] [22], [h] [14], [i] [23], [j] [24], [k] [25], [l] [26], [m] [27].

	DND-BN	AT-ZN	JW	M-S	exp.	*ab initio*
a_0	3.1652	3.165[e]	3.1652	3.14339	3.165[c]	3.14[f] 3.17[b]
E_{coh}	−8.90	−8.90	−8.90	−8.90	−8.9[d]	−9.97[f], −8.49[b]
T_{melt}	3900 ± 50	3950 ± 50	4100 ± 50[h]	4600 ± 100	3695[l]	
R_{cut}	4.4000	4.4002	4.4002	5.5000		
E_v^f	3.56	3.63	3.63	3.82	3.7 ± 0.2[j]	3.34[b] 3.56[i]
$E_{\langle 100\rangle}^f$	12.44	8.65	10.27	12.81		11.49[i] 12.87[m]
$E_{\langle 110\rangle}^f$	9.77	8.45	10.16	10.88		9.84[i] 10.82[m]
$E_{\langle 111\rangle}^f$	9.46	7.84	9.50	10.40	9.1 ± 0.6[k]	9.55[i] 10.53[m]
E_v^m	2.1	1.4	1.4	1.8		1.7