

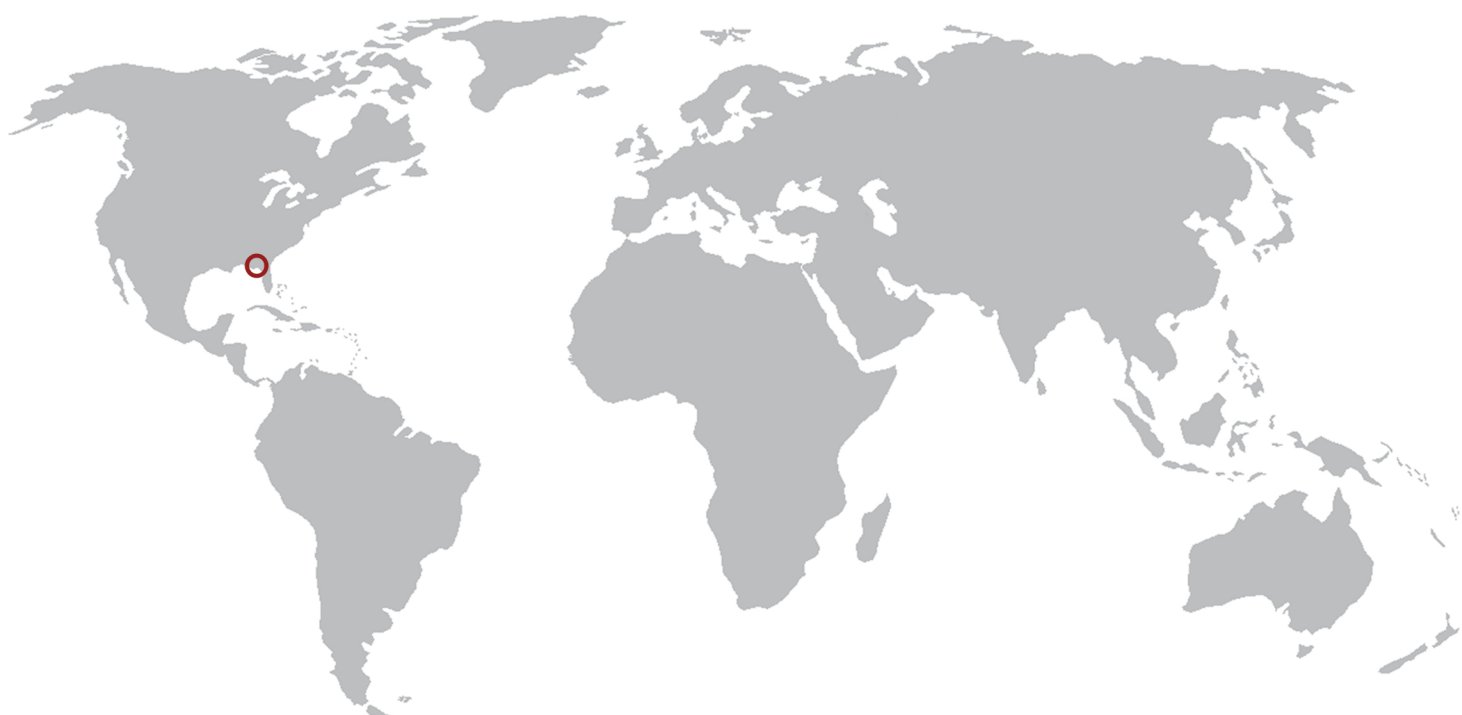
P R O C E E D I N G S

MMM 2008
Fourth
International Conference

MULTISCALE MATERIALS MODELING

OCTOBER 27-31, 2008 • TALLAHASSEE, FLORIDA, USA

*Tackling Materials Complexities
via Computational Science*



Hosted by the Department of Scientific Computing and Florida State University

DEPARTMENT OF
Scientific
COMPUTING



Proceedings of

MMM 2008
*Fourth
International Conference*
MULTISCALE MATERIALS MODELING
OCTOBER 27-31, 2008 • TALLAHASSEE, FLORIDA, USA

Anter El-Azab
Editor

**Organized and Hosted by
The Department of Scientific Computing and
Florida State University**

DEPARTMENT OF
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Papers published in this volume constitute the proceedings of the Fourth International Conference on Multiscale Materials Modeling (MMM-2008). Papers were selected by the program committee for oral or poster presentation. They are published as submitted, in the interest of timely dissemination.

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Forward

The field of multiscale modeling of materials promotes the development of predictive materials research tools that can be used to understand the structure and properties of materials at all scales and help us process materials with novel properties. By its very nature, this field transcends the boundaries between materials science, mechanics, and physics and chemistry of materials. The increasing interest in this field by mathematicians and computational scientists is creating opportunities for solving computational problems in the field with unprecedented levels of rigor and accuracy. Because it is a part of the wider field of materials science, multiscale materials research is intimately linked with experiments and, together, these methodologies serve the dual role of enhancing our fundamental understanding of materials and enabling materials design for improved performance.

The increasing role of multiscale modeling in materials research motivated the materials science community to start the Multiscale Materials Modeling (MMM) Conference series in 2002, with the goal of promoting new concepts in the field and fostering technical exchange within the community. Three successful conferences in this series have been already held:

- The First International Conference on Multiscale Materials Modeling (MMM-2002) at Queen Mary University of London, UK, June 17-20, 2002,
- Second International Conference on Multiscale Materials Modeling (MMM-2004) at the University of California in Los Angeles, USA, October 11-15, 2004, and
- Third International Conference on Multiscale Materials Modeling (MMM-2006) at the University of Freiburg, Germany, September 18-22, 2006.

The Fourth International Conference on Multiscale Materials Modeling (MMM-2008) held at Florida State University comes at a time when the wider computational science field is shaping up and the synergy between the materials modeling community and computational scientists and mathematicians is becoming significant. The overarching theme of the MMM-2008 conference is thus chosen to be “*Tackling Materials Complexities via Computational Science*,” a theme that highlights the connection between multiscale materials modeling and the wider computational science field and also reflects the level of maturity that the field of multiscale materials research has come to. The conference covers topics ranging from basic multiscale modeling principles all the way to computational materials design. Nine symposia have been organized, which span the following topical areas:

- Mathematical basis for multiscale modeling of materials
- Statistical frameworks for multiscale materials modeling
- Mechanics of materials across time and length scales
- Multiscale modeling of microstructure evolution in materials
- Defects in materials
- Computational materials design based on multiscale and multi-level modeling principles

- Multiscale modeling of radiation effects in materials and materials response under extreme conditions
- Multiscale modeling of bio and soft matter systems

The first five topical areas are intended to cover the theoretical and computational basis for multiscale modeling of materials. The sixth topical area is intended to demonstrate the technological importance and industrial potential of multiscale materials modeling techniques, and to stimulate academia-laboratory-industrial interactions. The last two topical areas highly overlap with the earlier ones, yet they bring to the conference distinct materials phenomena and modeling problems and approaches with unique multiscale modeling aspects.

This conference would not have been possible without the help of many individuals both at Florida State University and around the world. Of those, I would like to thank the organizing team of MMM-2006, especially Professor Peter Gumbsch, for sharing their experience and much organizational material with us. I also thank all members of the International Advisory Board for their support and insight during the early organizational phase of the conference, and the members of the International Organizing Committee for the hard work in pulling the conference symposia together and for putting up with the many organization-related requests. Thanks are due to Professor Max Gunzburger, Chairman of the Department of Scientific Computing (formerly School of Computational Science) and to Florida State University for making available financial, logistical and administrative support without which the MMM-2008 would not have been possible. The following local organizing team members have devoted significant effort and time to MMM-2008 organization: Bill Burgess, Anne Johnson, Michele Locke, Jim Wilgenbusch, Christopher Cprek and Michael McDonald. Thanks are also due to my students Srujan Rokkam, Steve Henke, Jie Deng, Santosh Dubey, Mamdouh Mohamed and Jennifer Murray for helping with various organizational tasks. Special thanks are due to Bill Burgess and Srujan Rokkam for their hard work on the preparation of the proceedings volume and conference program.

I would like to thank the MMM-2008 sponsors: Lawrence Livermore National Laboratory (Dr. Tomas Diaz de la Rubia), Oak Ridge National Laboratory (Dr. Steve Zinkle) and Army Research Office (Drs. Bruce LaMattina and A.M. Rajendran) for the generous financial support, and thank TMS (Dr. Todd Osman) for the sponsorship of MMM-2008 and for advertising the conference through the TMS website and other TMS forums.

I would also like to thank all plenary speakers and panelists for accepting our invitation to give plenary lectures and/or serve on the conference panels. Lastly, I would like to thank the session chairs for managing the conference sessions.

Anter El-Azab
Conference Chair

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Symposium 6

Multiscale modeling of radiation effects in materials

Ab-initio Based Modeling of Diffusion and Radiation Induced Segregation in Ni-Fe-Cr alloys

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ABSTRACT

High concentrations of point defects created in radiation environments can cause severe material degradation as they migrate and cluster. Radiation induced segregation (RIS) is the process by which the local composition of an alloy is altered due to transport of point defects to sinks. RIS has concerned the nuclear industry for decades since it can change materials properties at critical regions near grain boundaries, voids, and dislocations. However, a complete understanding of the temperature and composition dependence of RIS has remained elusive, primarily due to a lack of knowledge about the complex diffusion mechanisms of point defects in multi-component alloys. Here we use ab initio energetics, statistical mechanics, kinetic Monte Carlo, and rate equation modeling to identify the atomic scale processes underlying RIS. For the first time we clearly demonstrate which solute-defect mechanisms lead to RIS in Ni rich fcc Fe-Ni-Cr alloys. We find weak binding between Fe and Cr solutes and vacancies, which rules out vacancy drag mechanism as a dominant driving force. We find strong binding between Cr solutes and the $\langle 100 \rangle$ interstitial, which is counterintuitive to former assumptions based on solute size. These atomic scale effects are integrated into statistical and kinetic Monte Carlo models to determine diffusion coefficients associated with both vacancy and interstitial migration. These diffusion coefficients are then used to parameterize a rate theory type model for quantitative RIS predictions.

Interaction of Interstitial Clusters With Solute Atoms in Ferritic Alloys and its Consequence for Microstructural Development Under Neutron Irradiation

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ABSTRACT

Up to now the theory of microstructure evolution in irradiated materials has been developed mainly for pure metals. It succeeds in explaining several observations in metals with fcc crystal structure and the difference between swelling under neutron and electron irradiations. The theory emphasises the role clusters of interstitial atoms play in the microstructure development and importance of the steady production of small interstitial clusters directly in displacement cascades under neutron irradiation. Despite this significant success, the understanding of distinguishing features of microstructure development specific to bcc alloys, e.g. low swelling rates, and more generally its sensitivity to material composition is still poor. In this paper, a summary of the information on the interaction of irradiation-produced defects, vacancies and clusters of interstitial atoms, with interstitial carbon, substitutional chromium and copper solute atoms, and copper precipitates in ferritic alloys is presented. This was obtained in recent years from molecular dynamics simulations using existing empirical potentials for the description of interatomic forces and compared with published data from *ab initio* calculations. It is discussed on the basis of the rate theory that this interaction, especially for interstitial clusters, may be the key for understanding of many features of microstructure development, such as copper precipitate and void growth, under neutron irradiation.

The research was sponsored by a research grant from the UK Engineering and the Physical Sciences Research Council.

An ab initio based AKMC model of ferritic Fe alloys under irradiation

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ABSTRACT

Neutron irradiation of pressure vessel steels lead to the formation of point defects and solute clusters that are responsible for the vessel embrittlement. In order to model the evolution of the microstructure of Fe-CuNiMnSiP alloys a multiscale modelling approach is followed based on ab initio and kinetic Monte Carlo simulations. Ab initio results in the framework of density functional theory related to point defect clusters will be presented and in particular the effect of solutes on the relative stability of small interstitial clusters will be discussed. The ab initio calculations indicate also that solute transport via an interstitial mechanism is very likely for Mn. Our atomistic kinetic Monte Carlo (AKMC) model treats thus self interstitials in addition to vacancies. The parameterisation for the vacancy diffusion has been done using thermal annealing experiments on dilute Fe alloys of growing complexity, while the parameters regarding the self interstitial atoms were adjusted using isochronal annealing data. The AKMC model has been used to simulate isochronal annealing experiments in model Fe based alloys as well as electron and neutron irradiation in dilute Fe-CuNiMnSiP alloys.

The challenge to bridge scales in computational modeling Fe alloys

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ABSTRACT

One of the main challenges in the multiscale approach to computational materials science is to bridge the scales in a meaningful and consistent way, which requires being able to capture the main processes at every scale and communicate them to the next.

For several materials of interest this simple statement has been challenged at the first bridge between scales, namely the electronic structure and molecular dynamics. In particular for steels, the situation is such that we can not say today that we have a satisfactory link between these scales, and the reason is that many of the unique properties of Fe and its alloys are based on electronic properties that can not be translated into classical counterparts. In this talk I will review the significant progress done in recent years in understanding the most elementary properties of Fe and some its alloys which is a requirement for a successful implementation of a multiscale approach to their properties.

Ion irradiation of nanocrystals

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ABSTRACT

Ion irradiation can be used to modify the structure of nanoparticles. For instance, light ions can be used to enhance the magnetic properties of FePt particles due to increased ordering, an effect that is widely used for bulk systems. In some cases, however, nanoparticle systems respond to irradiation quite differently than their bulk counterparts.

The effects of irradiation of nanoparticles can be examined by a multi-scale combination of methods similar to those used for bulk materials. In my talk I will describe our recent work on ion beam modification of nanoclusters carried out with a combination of molecular dynamics methods and analytical models. We have shown that the sputtering yield from nanoclusters can be much higher than that from the bulk, an effect which may be useful for enhancing SIMS. We have also examined the experimental observation that irradiation can be used to transform multiply twinned nanoparticles into a single crystalline morphology, and shown that the most likely explanation to the effect is a transient amorphization stage.

Microstructural Evolution of Binary Alloys under Irradiation

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ABSTRACT

The evolution of a compositional microstructure of a concentrated binary alloy under irradiation is simulated using a continuum kinetic model, which considers a vacancy and interstitial transport mechanism for atomic diffusion. The model is based on the coarse graining of a simple atomistic model, where point defects, both isolated and clustered, are included. The effect of irradiation-induced displacement cascades on the chemical composition is modeled by the introduction of a controlled number of single and clustered defects, which will evolve accordingly to local atomic fluxes. A recombination term is also included to take into account the annihilation of vacancies and interstitials due to thermal diffusion. Simulations in two and three dimensions are employed to investigate the effect of material properties, including point-defect and solute coupling, as well as irradiation parameters, namely the primary recoil spectrum, on irradiation-induced segregation and precipitation.

Molecular dynamics simulation of irradiation induced phase transition in MgAl_2O_4

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ABSTRACT

The kinetic of phase transitions under irradiation of MgAl_2O_4 has been studied using molecular dynamics (MD) simulation by means of continuous cation Frenkel pair (FP) accumulation. Three different cases have been considered: cation FP accumulation starting (i) from the normal (N) spinel, (ii) from the amorphous (A) spinel and (iii) from the interface between rock-salt (NaCl) spinel and A-spinel.

When submitted to FP accumulation, the N-spinel transits directly towards the NaCl-spinel for temperature lower than 600K. For temperatures higher than 600K, the dose needed to obtain the rock-salt structure increases with temperature, and the structure transits to an intermediate disordered (D) structure prior to the NaCl-spinel. No amorphization of the spinel under FP accumulation is obtained for doses up to 68 displacements per cation at 30K. The dose – temperature dependence relies on thermally activated FP recombination processes.

The epitaxial and homogeneous irradiation induced re-crystallization of amorphous MgAl_2O_4 was obtained by continuous FP accumulation. Present results point out that the re-crystallization induced by FP accumulation appears in both cases to be thermally enhanced but non diffusive. It is governed by a local rearrangement of each point defect in the homogeneous case, while spontaneous FP recombination process in the crystalline part or at the interface drives the re-crystallization in the epitaxial case.

In summary, the thermally enhanced – *but non diffusive* – Frenkel pair recombination or local relaxation of point defects may play a central role to the radiation resistance of MgAl_2O_4 .

1. Introduction

Magnesium aluminate spinel MgAl_2O_4 is known to have an excellent radiation resistance. Its radiation-induced behavior has been studied extensively using both experimental techniques [1–3] and recently numerous atomic scale modeling [4, 5]. It has been shown that MgAl_2O_4 undergoes amorphization under high-dose heavy ion irradiation at cryogenic temperatures. A meta-stable phase has been recognized as a transient state prior to the phase transformation to amorphous state [1–3]. Recent results of displacement cascades [4, 5] have shown that single events do not induce direct impact amorphization of the spinel structure, but rather produce Frenkel pairs (FP) and disordering in the sub-lattices. Those results indicate that point defect accumulation may play an important role for the phase transformation of MgAl_2O_4 under irradiation. Conversely, the amorphous (A) state of MgAl_2O_4 may re-crystallize into a defective

rock-salt (NaCl) structure under electron irradiation [6]. The re-crystallization has been observed to occur both at the amorphous/crystalline interface by epitaxial (E) growth or directly in the amorphous region by nucleation and growth (H). The effect of ballistic displacement has been invoked as a driving process for the re-crystallization under irradiation [6] at low temperature. We have investigated the effects of irradiation in N-spinel (normal spinel), A-spinel (amorphous spinel) and for the NaCl – A spinel (rock-salt – amorphous spinel) interface by means of continuous cation FP accumulation in the framework of Molecular Dynamics (MD) using empirical potentials. The aim of the present investigation is to understand the processes that induce the phase transformation of MgAl_2O_4 submitted to irradiation.

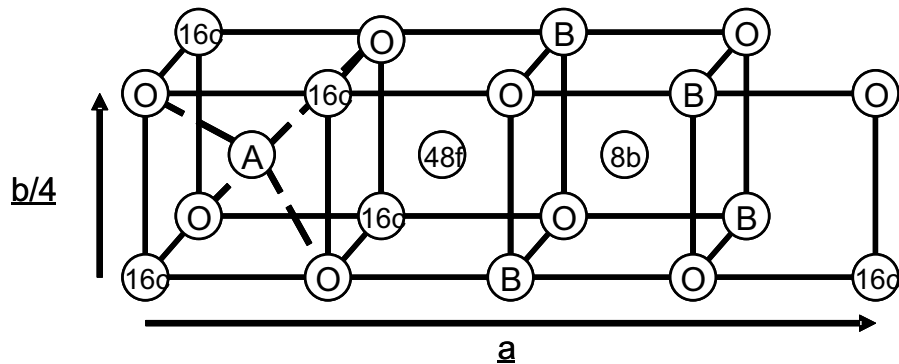


Figure 1: Schematic representation of the partial spinel structure. The \underline{a} and \underline{b} symbols indicate the axis of the conventional cell. The A, B and oxygen (O) sites of the N spinel are plotted (see the text for details) along with the octahedral interstitial sites (16c) and tetrahedral interstitial sites (8b and 48f).

2. Modeling the phase transition under irradiation.

1.1 Crystal structures.

At low temperature the normal spinel (N-spinel) crystallizes in the $Fd\bar{3}m$ (227) space group. The magnesium ions are at the tetrahedral sites 8a while the aluminum ions are at the 16d octahedral sites. The oxygen ions are organized as a fcc sub-lattice in the 32e sites (see Fig. 1). Upon increase of temperature, the cation sub-lattices are disordered. The perfect disordering (called D-spinel) in the cation sub-lattice is attained for $x = 2/3$ in $\text{Mg}_{1-x}\text{Al}_x(\text{Al}_{2-x}\text{Mg}_x)\text{O}_4$. The I-spinel (inverse spinel $\text{Al}[\text{MgAl}]_2\text{O}_4$) occurs when all the Mg ions lie in the 16d octahedral sites, the tetrahedral sites being filled by Al cations. Note that the oxygen ions keep their own site 32e. The defective rock-salt structure of spinel (NaCl-spinel) can be described using the other fcc sub-lattice built up by the 16c sites (see Fig. 1). Both interpenetrating fcc sub-lattices 16c and 16d and the 32e sub-lattice form a defective rock salt structure when the Al and Mg cations are randomly distributed in the 16c and 16d sites. Finally, there are two other tetrahedral special sites (48f and 8b, see Fig. 1), that are interstitial sites for spinel as well as for the defective rock salt structure. They mainly remain empty during all the simulations performed; and will not be considered in the following.

1.2 Two-body potentials assessment

We have used the two-body empirical potentials fitted by Morooka *et al* [7] to describe MgAl_2O_4 . They are composed of a Buckingham potential and a Morse potential that complete the Coulomb interactions. Those potentials properly describe the thermal expansion as well as the elastic properties as a function of temperature for MgAl_2O_4 , but also for the end-members of the $\text{MgO} - \text{Al}_2\text{O}_3$, i.e. MgO and Al_2O_3 .

We further checked Morooka's potentials by evaluating the relative energies of the D-, I-, NaCl- and A-spinel. The physical properties as well as the radial distribution functions obtained for each phase of spinel will also be used as references for the on-line analysis of the evolution of the structure during the accumulation of cation FP. We built up super-cells by doubling the lattice parameter of the conventional cell along the three crystallographic directions, obtaining a 448-ion cell, except for the case of epitaxial re-crystallization which contains the 448-ion cell of NaCl-spinel and the A-spinel. Thirty (30) different random configurations of the I-spinel, D-spinel and NaCl-spinel were generated and the lattices were relaxed by static energy minimization. Their relative energies, volumes and bulk modulus, were obtained by averaging over the 30 configurations. We have also generated the amorphous spinel (A-spinel), by quenching a 448-atom super-cell from 6000 K towards 300 K with a linear cooling rate of 10 K/ps at constant pressure. The properties of the A-spinel at 0 K were then calculated by static energy minimizations of the A structure obtained at 300 K.

Structure	Volume (nm^3)	Bulk Modulus (GPa)	Energy (eV/atom)	Energy ab initio (eV/atom) [8]
N-spinel	0.5196	192.5	0.000	
D-spinel	0.5181 (-0.3%)	191.3 (-0.6%)	0.041	0.036
I-spinel	0.5143 (-1.0%)	202.0 (+4.9%)	0.054	0.039
NaCl-spinel	0.5130 (-1.3%)	153.4 (-20%)	0.144	
A-spinel	0.5316 (+2.3%)	139.9 (-27%)	0.183	

Table 1: Physical properties of the different phases of spinel calculated using static energy minimization.

Overall, the results (quoted Table 1) are consistent with the available experimental and *ab initio* data. In particular, the relative energies of the four different phases show an evolution consistent with the experiments. More details on the relative stability are available in a separate papers [8, 9].

1.3 Continuous cation Frenkel pairs (FPs) accumulation

The irradiation induced phase transition of MgAl_2O_4 has been simulated by means of continuous cation FP creation with MD simulations in the NPT ensemble, using a method similar to the one used by Crocombette *et al.* [9]. In both cases, the cation FPs are introduced one after another. For each FP introduction, one cation was randomly chosen and was displaced randomly to an interstitial site. The interstitial (either for crystalline states or for amorphous states) is defined as a position located further than 1 Å distance from any other atoms of the cell. The target temperature was strictly maintained using a Nosé-Hoover thermostat in order to remove the excess of thermal energy produced by the introduction of FPs in the system. Such a method avoids any local heating caused by the FP production in the cell throughout the simulations. We assumed 1 ps and 10 ps for the time intervals between subsequent FP introductions. They

correspond, respectively, to dose rates Φ of 5×10^9 and 5×10^8 displacements per cation per second (dpc/s) for a 448-atom cell. Those values of Φ are far higher than the maximum dose rate that may be attained in experiments (10^{-3} dpa/s). Such short time intervals between FP introductions prohibit any long range migration, while they allow the FP to recombine spontaneously or each defect (interstitial and vacancy) to relax locally.

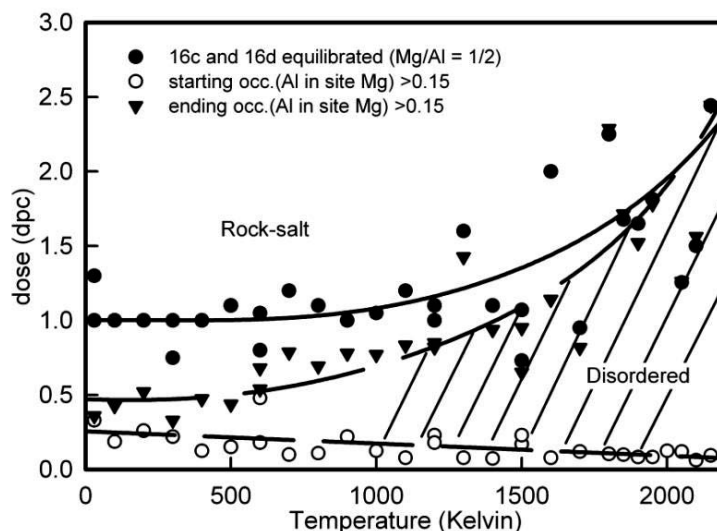


Fig. 2: Critical doses D_C (in displacement per cation, dpc) for the phase transitions of N-spinel under the accumulation of cation FPs (with a time interval between two cation FP of 10 ps) as a function of the temperature.

3. Normal spinel under cation FP accumulation.

We have performed the accumulation of cation FPs in normal spinel, as a function of temperature [10]. The analysis of the evolution of the structure as a function of the critical dose D_C for transition was monitored by calculating the occupation fraction of Al and Mg ions at all the site positions. The analysis was done just before the introduction of the FP in the cell, such that the previous one is already relaxed. The transition dose D_C (in dpc) towards the D-spinel was defined when the occupation fraction of aluminum in the magnesium sites is above 0.15 (as can be seen on the Fig. 2). The transition dose towards the NaCl-structure was defined when the occupation fraction of all the tetrahedral sites is below 0.1 and the 16d and 16c sites are randomly occupied by Mg and Al.

We have reported on Fig. 2 the evolution of the structure as a function of dose of cation FPs with a time interval of 10 ps. We observe that the steady state of spinel under FP accumulation is the rock-salt structure. No amorphization has been observed under FP accumulation, even for the accumulation with the higher flux (time interval of 1 ps) up to a dose of 68 dpc, at 30K.

At low temperature (below 600 K), the N-spinel structure almost directly transits towards the NaCl-spinel structure. For temperature higher than 600 K, the N-spinel structure transits first towards the D-spinel structure, and then towards the rock-salt structure. We recall that no diffusion is allowed during the cation FP accumulation because the time intervals between two successive FP introductions are too short (1 or 10 ps). Consequently, such behavior is related

with the thermal activation of recombination of cation FPs above 600 K, as in pyrochlore [9]. The cation interstitials in the tetrahedral sites recombine with vacant sites either in the original tetrahedral sub-lattice (the 8a sites) or in the original octahedral sub-lattice (the 16d sites). It induces a disordering in the cation sub-lattices, prior to the stabilization of the NaCl-spinel. Finally, the transition sequence of N-spinel under irradiation may then be summarized as N-spinel \rightarrow D-spinel \rightarrow NaCl-spinel, in agreement with experimental evidences [1].

4. Amorphous spinel under cation FP accumulation.

We have calculated the re-crystallization behavior of A-spinel under the accumulation of cation FPs [11]. Two different starting configurations have been considered: the A-spinel and the interface between NaCl-spinel and A-spinel. We have chosen the NaCl-spinel for the interfacial configuration because the steady state of MgAl_2O_4 under irradiation is precisely the NaCl-spinel and too because the homogeneous re-crystallization under irradiation (detailed below) leads to the NaCl-spinel. Two different fluxes Φ (i.e. time interval of 1 and 10 ps between cation FP introductions) were investigated in both cases.

The analysis of the evolution of the structure as a function of dose D_C was done just before each FP creation. The transition of the A-spinel cannot be monitored by means of site occupations analysis. Instead, the transition of the structure was identified by following the evolution of the enthalpy and the volume as a function of dose. We also cross-controlled the analyses using the total radial distribution function. More details are available in a previous paper [11].

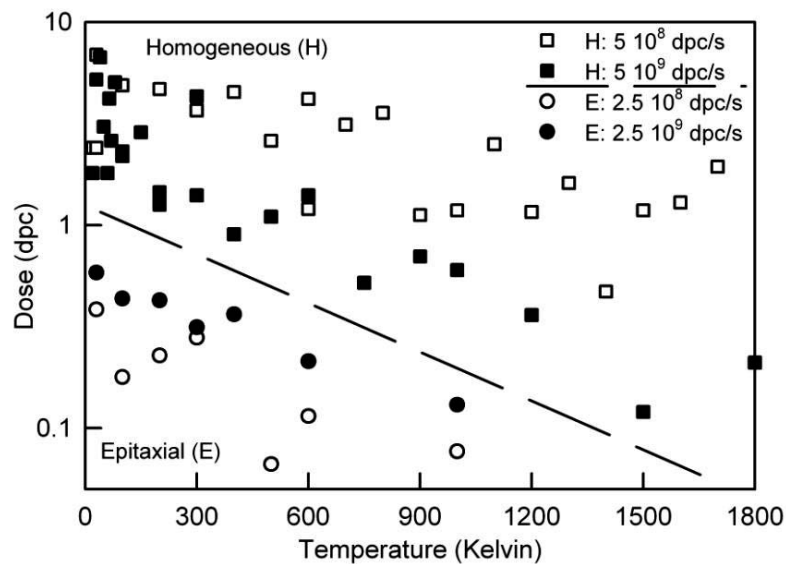


Figure 3: Dose of re-crystallization of the A-spinel (homogeneous (H) squares) and of the epitaxial amorphous/NaCl-spinel (epitaxial (E) circles) as a function of temperature. The dotted line is separates the homogeneous from the epitaxial ranges.

The re-crystallization towards the NaCl-spinel was obtained beyond a certain dose of FP accumulation in the homogeneous as well as in the epitaxial case, at all the temperatures and fluxes considered.

We have reported for both cases (epitaxial E and homogeneous H) the critical dose D_C for re-crystallization as a function of temperature for the two different fluxes in Fig. 3. We can observe that for both homogeneous and epitaxial re-crystallization the critical dose decreases with temperature. The epitaxial (E) re-crystallization doses are one order of magnitude lower than the doses for the homogeneous (H) re-crystallization irrespective of the temperature. It means that E and H re-crystallization may be interpreted independently in the present simulations. The other main characteristic of those results is that the critical doses D_C for re-crystallization depend upon the FPs flux Φ . The higher Φ induces the lower D_C in the H case ($D_C \propto \Phi^\beta$ with $0 < \beta < 1$). This trend is reversed in the E case ($D_C \propto \Phi^\beta$ with $\beta > 1$). It is, therefore, straightforward that different microscopic mechanisms pilot the re-crystallization of spinel in the homogeneous nucleation and growth and in the epitaxial processes. Following the proposed model by Heera *et al* [12] (equation 15 in their paper), and since no thermal diffusion of the point defects is allowed, we may deduce that homogeneous re-crystallization is driven by the local reorganization induced by the cation displacements while in the epitaxial case the re-crystallization is governed by the FP annealing.

Acknowledgements

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First principles modeling of defects behavior in UO_2 and CeO_2

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ABSTRACT

To investigate the microscopic mechanism of the defective structure behavior in the ceramics fuel UO_2 under irradiation, a comprehensive first principles calculation has been performed for UO_2 , as well as CeO_2 , a simulation materials of UO_2 used in the accelerator experiments due to its similar structural and thermodynamic properties. For supercells containing various types of point defects including one of main fission product Xe in both UO_2 and CeO_2 , the electronic structure, the defect formation energies and the lattice derivation are evaluated using PAW-GGA/LDA+U. Combing with Cluster Expansion Method (CEM), the effective cluster interaction energies for two systems are extracted. Based on the resultant ground state properties, the concentrations of various types of point defects are further estimated in the framework of the point defects model. These calculations give an explanation on the nonstoichiometric features manifested in the phase diagrams of U-O and Ce-O, that is, UO_2 is hyperstoichiometric, whereas CeO_2 is hypostoichiometric, and revealed the origin of similarities and differences between these two systems from electronic structure level. The calculations are further extended to the clustering behavior of oxygen in the nonstoichiometric UO_{2+x} . Based on the local stability of five basic clustering modes of oxygen defects, the clustering pattern for anion-excess uorites is suggested, which provided a united picture for understanding the structure of Willis type and cuboctahedral clusters in UO_{2+x} and $\beta\text{-U}_4\text{O}_9$.

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Magnetic Cluster Expansion Study of Magnetism and Thermodynamic Properties of Iron and Iron-Chromium Alloy

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ABSTRACT

Central problem with the description of FeCr alloys is the presence of two contributions to the energy of the alloy, the magnetic and the non-magnetic (configurational) energies, related to each other in a non-trivial way. The existing approaches, such as the Cluster Expansion, do not distinguish between these two contributions. The absence of magnetic moments in a conventional Cluster Expansion does not make it possible to apply the method to the study of the full magnetic and structural phase diagram of FeCr. We present more realistic approach for treating the energy and magnetic properties of solid solutions: the Magnetic Cluster Expansion (MCE). The expression for the energy of an alloy in the MCE, in addition to the conventional discrete occupational variables, explicitly includes magnetic moments associated with the constituent atoms of the alloy. These magnetic variables take any vector values. For pure iron, this allows to restrict the range of interactions, as compared to the CE, to nearest and second nearest neighbours only, in agreement with direct DFT evaluation of exchange parameters of the Heisenberg model for ferromagnetic iron. For iron-chromium interactions, we retain terms up to the fourth nearest neighbour, and for chromium-chromium – up to the fifth. The values of the magnetic moments for each alloy structure are found by minimizing the total energy of the system described by the MCE. We present a derivation of MCE interaction parameters, obtained via fitting to the DFT enthalpies of formations, and to magnetic moments evaluated for a set of reference structures of the binary FeCr system. We apply the method to the Monte Carlo investigation of the energy and magnetic properties of pure iron and FeCr alloy in the whole region of concentrations.

This work was supported by the UK EPSRC, by EURATOM, and by EXTREMAT integrated project under contract number NMP3-CT-2004-500253.

Systematic Study of Interactions between Irradiated Defects and Impurities in Bcc Transition Metals

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ABSTRACT

Tungsten and other bcc transition metals are the main candidate plasma facing materials for the divertor and other components of future fusion power plants. There is ample evidence for that impurity atoms play a significant part in micro-structural evolution under neutron irradiation. For example, impurities have strong degrading effect on mechanical properties due to strong interaction with radiation point defects, the so-called trapping effect in metals. We apply density functional theory (DFT) to the investigation of the interaction between impurity atoms and point defects (vacancies and self-interstitial atoms (SIAs)) in these metals. The benefits of doing systematic DFT studies of defects in all bcc transition metals have now become clear. Our recent investigations of SIA configurations show that in all the non-magnetic bcc transition metals, including bcc-W, the most stable defect configuration has the $\langle 111 \rangle$ (crowdion) orientation, in contradiction with earlier empirical potential results suggesting that the $\langle 110 \rangle$ orientation. Our benchmark DFT calculation of the migration energy of mono-vacancy (1.78 eV) and the prediction of positive binding energy between two vacancies in the first-nearest neighbor of bcc-W are also in excellent agreement with the available experimental observations. We have carried out a systematic DFT study of impurities (C, N, O and H) in all the bcc transition metals (W, Ta, Mo, Nb, Cr, V and Fe) and found that C and N atoms are more stable in the octahedral interstitial site, whereas O and H atoms likely occupy the tetrahedral sites. We have also investigated the interaction of mono-vacancies and SIA configurations with these four impurities assuming that they occupy the most favorable lattice site predicted by the DFT calculations. A particular case of interaction between carbon and nitrogen atoms and a mono-vacancy in bcc-W, the binding energies characterizing bound states formed by the carbon and nitrogen atoms located at octahedral sites, and a vacancy on a nearest neighbor site are very large (1.39 eV and 1.91 eV, respectively). This is expected to have a strong effect on the mobility of vacancies in tungsten. We discuss implications of the new set of data for the binding energies of impurity and point-defect interactions for the interpretation of experiments on diffusion of radiation defects in bcc transition metals.

This work was supported by the UK EPSRC, by EURATOM, and by EXTREMAT integrated project under contract number NMP3-CT-2004-500253.

Atomistic studies of properties of helium in BCC iron

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ABSTRACT

In fusion applications, helium caused by transmutation plays an important role in the response of RAFM steels to neutron radiation damage. The growth, migration and coalescence behavior of helium bubbles is very sensitive to the properties of individual He interstitials and helium-vacancy clusters [1]. We have performed atomistic simulations using the Ackland [2] Fe potential combined with different potentials for the iron-helium interaction. Interstitial helium is very mobile and coalesces together to form interstitial clusters. With the ORNL 3-body Fe He potential [3-5], the interstitial clusters show lower binding energy than with the Wilson potential [6], in agreement with the DFT calculations of ref [7]. This lower binding energy increases the maximum number of He atoms that can exist in a mobile interstitial cluster. When the He cluster reaches a sufficiently large size the cluster will push out an Fe interstitial, creating a Frenkel pair. The resulting helium-vacancy cluster is not mobile. The reverse process is also studied. Multiple helium atoms can be trapped in a single vacancy, and if there are few enough, the vacancy can recombine with an Fe interstitial to create a helium interstitial cluster. Mobile helium interstitial clusters may provide a mechanism by which helium bubbles in iron can become over-pressurized. Over-pressurized bubbles are known to punch out interstitial dislocation loops, and such loops have been observed - for example in ref [8].

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Interaction of He_nV_m Clusters with Coherent and Semi-Coherent Fe/Cu Interfaces

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ABSTRACT

In the fusion neutron environment copious quantities of He will be produced by transmutation reactions in addition to point defects associated with displacing atoms from their lattice sites. Helium forms gas bubbles in the matrix, on dislocations, at precipitate-matrix interfaces, and in grain boundaries. Helium bubbles can act as formation sites for growing voids at lower irradiation temperatures and creep cavities on stressed grain boundaries at high temperatures. In order to design microstructures that are resistant to high levels of He and displacement damage it is important to develop predictive models of the interaction of He atoms, vacancies and He-vacancy complexes with the microstructural features mentioned above. Previous computational modeling shows that interstitial and substitutional He as well as He-divacancy clusters are strongly bound to extended defects in Fe such as dislocations, grain boundaries and particle-matrix interfaces. A finding from our past work is that He binds with coherent Cu particles in an Fe matrix. Our earlier research also established that the core of an edge dislocation strongly traps He. Thus, we hypothesized that a semi-coherent interface might be a stronger trap for He than a coherent interface due to the array of misfit dislocations needed to accommodate the lattice parameter mismatch between the particle and the matrix in the semi-coherent case. In the present study we employ atomistic simulations to compare the binding of He_nV_m complexes to coherent and semi-coherent bcc Fe/bcc Cu interfaces. The simulations show that the binding energy of He_nV_m complexes to a coherent Fe/Cu interface ranges from 0.35 eV for a single vacancy up to 0.70 eV for a He₁V₂ complex. A semi-coherent interface was found to be a much stronger trap for He near the core of a misfit dislocation. Binding energies varied from 0.86 eV for a substitutional He atom up to 2.38 eV for a He₁V₂ complex. These binding energies were found to be significantly larger than the values obtained for simple edge dislocations in Fe. The trend in binding energies can be rationalized in terms of the spatial dependence of excess atomic volume for each interface.

Combined Atomistic and Dislocation Dynamics Modeling of Dislocation-SFT Interactions in Cu

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ABSTRACT

When subject to irradiation, fcc metals with low stacking-fault energies are known to develop a microstructure characterized by a large number density of small stacking-fault tetrahedra (SFTs). Upon deformation, these materials undergo a very heterogeneous plastic behavior, with most of the strain localized along a few shear bands wherein SFTs are removed. The SFT removal mechanisms by dislocations presumably governs the width and spatial distribution of these shear bands, and considerable effort has been devoted to their study by computer simulation. Although atomistic simulations have revealed numerous useful insights about these processes, they are too intensive to explore the full parameter space present in real systems (dislocation character and length, SFT size, reaction geometry, etc). In this work, we perform dislocation dynamics (DD) simulations of dislocation-SFT interactions for a wide range of conditions and SFT sizes. Our DD methodology is fitted from data obtained by specifically-tailored atomistic simulations and explicitly takes into account partial dislocations in fcc crystals. We provide strength curves as a function of the distance from the base of the dislocation glide plane, and identify the different mechanisms operating within each regime. We show that the strength of SFTs to dislocation passage can be universally described with a simple power law that depends only on the SFT area intersected by the dislocation glide plane

Core Structure, Peierls Potential and Kinks of Screw Dislocations in Iron from First Principles

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ABSTRACT

A quantitative description of straight and kinked $\langle 111 \rangle$ screw dislocations in iron from first principles is presented. The calculations are carried out within the density functional theory (DFT) framework using the SIESTA code and simulation cells containing up to 800 atoms. The construction of simulation cells appropriate for such extended defects has been optimized for cell sizes accessible to DFT calculations. We have rationalized the cell-size dependences of the energetics evidenced both in the cluster approach and in the dipole approach for various cell and dipole vectors; they are due respectively to surface-dislocation and core-core interactions. It is concluded that a quadrupolar arrangement of dislocation dipoles is best suited for such calculations.

In agreement with previous DFT calculations in body centered cubic transition metals, a non-degenerate structure is found for the core. From a detailed comparison with anisotropic elasticity a significant dilatation effect has been evidenced in addition to the Volterra field. Two high symmetry directions of the Peierls potential have been sampled: the line joining two easy-core positions – which gives the Peierls barrier – and that from an easy to a hard-core position. The behavior of the Ackland-Mendelev potential for iron, which gives the correct non-degenerate core structure unlike most other potentials, has been tested against the present results [1].

Finally, the structure and the formation and migration energies of single kinks have been investigated. The methodology for constructing triperiodic cells with a single kink on each dislocation line has been developed. Results obtained with empirical potentials and the perspectives to perform similar calculations within DFT are presented [2].

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Multiscale Modeling and Experimental Validation of Radiation Effects in Silicon Carbide

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ABSTRACT

Multiscale computer simulations and ion-beam irradiations are used to investigate primary damage states and radiation damage evolution in SiC as functions of temperature, ion mass and energy. Multiscale computation methods are used to determine stable defect configurations, defect production, damage efficiency, cascade-overlap effects, close-pair recombination, and defect migration energies and pathways. Simulations of ion mass and energy effects indicate that the damage efficiency for C ions decreases rapidly with energy to a value of about 50%. In the case of Si ions, the damage efficiency is 100% up to energies of 1 keV, above which the damage efficiency decreases with energy to 50%. For Au ions, the damage efficiency is 100% to about 5 keV before decrease with energy to 62% at 50 keV. These damage efficiencies are consistent with the experimental damage production behavior observed in SiC irradiated at 150 K with C, Si and Au ions. The disordering behavior, volume change and mechanical properties obtained experimentally and from molecular dynamic simulations are in good agreement. The point defects produced by radiation are dominated by close Frenkel pairs, and atomistic simulations indicate that the activation energies for recombination of most close pairs range from 0.24 to 0.38 eV. Atomistic simulations have also determined that the activation energies for long-range diffusion of C and Si interstitials are 0.7 and 1.5 eV, respectively. Using these activation energies and ab initio results as input parameters, kinetic lattice Monte Carlo annealing simulations of a 10 keV MD cascade demonstrate the roles that close-pair recombination and long-range interstitial diffusion play in the thermal recovery stages observed experimentally. This synergistic integration of experimental and computational efforts is providing atomic-level understanding and necessary parameters to model dynamic defect processes and the evolution of irradiation damage in SiC as functions of time, temperature and dose rate.

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Radiation Damage in Nanocrystalline UO₂

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ABSTRACT

Displacement cascades in nanocrystalline UO₂ are studied using large-scale molecular dynamics (MD) simulation. Point-defect formation from radiation damage in single-crystal UO₂ has been studied previously [1, 2]. To understand the role of grain boundaries in radiation damage, we performed simulations on nanocrystalline UO₂. The microstructure consists of 4 grains of identical shape and diameter ($d = 10$ nm) in a three-dimensional periodic simulation cell. A heavy cation (Uranium) is used as a primary knock-on atom (PKA) with energy ranging from 1 keV to 5 keV. We characterize the defect formation and migration, with particular emphasis on the interaction of the radiation cascade with the grain boundaries. We also determine the effect of initial crystallographic direction of PKA on the defect formation. The effect of the temperature on the defect formation/diffusion is also examined.

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Molecular Dynamics Study of Frenkel Pairs in Cerium Dioxide

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ABSTRACT

Molecular dynamics (MD) simulations of oxygen Frenkel pairs (FPs) in cerium dioxide (CeO₂) were carried out in order to understand their kinetic behavior. The results show that an oxygen FP annihilates by recombination of the vacancy and the interstitial after the vacancy jump along the $\langle 100 \rangle$ direction. When multiple oxygen FPs are introduced, the interstitials aggregate into a (111) plate-like cluster at relatively lower temperature, while they annihilate by recombination with vacancies at elevated temperatures. The theoretical results are consistent with the transmission electron microscopy (TEM) observations of formation of {111} oxygen interstitial platelets in CeO₂ under electron irradiation.

1. Introduction

Recent transmission electron microscopy (TEM) studies have shown that electron irradiation produces plate-like defect clusters in several fluorite structure materials, such as ZrO₂ [1], CeO₂ [2] and CaF₂ [3]. Fig.1 gives an example of TEM bright-field images of CeO₂ irradiated with 1 MeV electrons at 300 K. Here, one may recognize dark contrast due to oxygen platelets with the nature of $1/9\langle 111 \rangle\{111\}$ [2]. Under electron irradiation, light O ions are selectively displaced from their lattice sites, while the sublattice of heavy Ce ions remains stable. Thus displaced oxygen interstitials are considered to aggregate into {111} platelets. Following the above experimental results, molecular dynamics (MD) simulations have been carried out in the present study to understand recombination of displaced oxygen interstitials with vacancies as well as aggregation of interstitials.

2. Procedure of molecular dynamics (MD) simulations

Buckingham potentials, complementing the Coulomb interactions, have been employed in the present study to describe the two-body short range ionic interactions. The potential parameters were set as the same as used by Arima *et al* [4]. The charge states of the constituent ions were

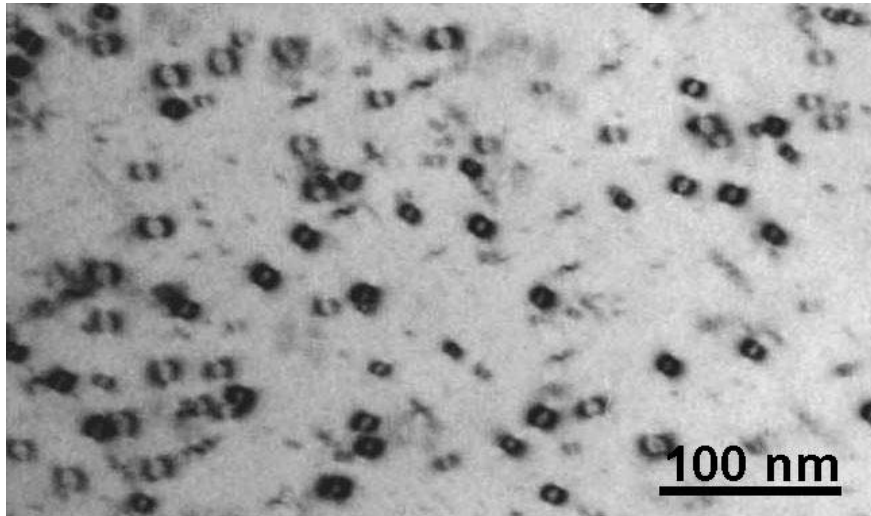


Figure 1. Dislocation loops formed in CeO₂ under 1250 keV electron irradiation with a fluence of about 2×10^{26} e/m² at 296 K [2].

assumed as $+4e$ and $-2e$ ($e=1.602 \times 10^{-19}$ C) for Ce and O ions, respectively. A super-cell containing $5 \times 5 \times 5$ unit cells was used in this study, including 500 Ce and 1000 O ions. The MD simulations were performed with the GULP (General Utility Lattice Program) code [5] in the NPT ensemble at temperatures of 300, 600, 900 and 1200 K. The super-cell was first equilibrated during 10 ps at a given temperature, and then Frenkel pairs (FPs), or pairs of oxygen interstitials and vacancies, were introduced into the super-cell. The atomic configurations were monitored every 0.2 fs up to 10 ps.

3. Results and discussion

3.1 Recombination of a single oxygen Frenkel pair (FP)

Tab.1 lists the arrangements of FP and their recombination feasibility at 300 K within 10 ps. Here the distance between a vacancy and an interstitial of a FP ranges from 1st nearest neighbor (1NN) to 7th nearest neighbor (7NN). According to the crystal symmetry, there are two different orientations for 4NN and 7NN FPs, or types A and B, while the others are characterized by one crystallographic orientation, as categorized in Tab.1.

It was found that some configurations of oxygen FPs, namely 1NN, 2NN, 4NN-type A and 7NN-type A, recombine rapidly within 0.01 ps after their creation. The 1NN FP recombines and annihilates immediately in one jump process, whereas FPs with longer separation, or 2NN, 4NN-type A and 7NN-type A FP, recombines after several steps of vacancy jump. One may notice that the rapidly recombining FPs are separated as $1/4 \langle 111 \rangle + n \langle 00 1/2 \rangle$. The MD simulations demonstrated that a vacancy readily replaces a lattice O ion by a jump toward $\langle 00 1/2 \rangle$. The replacement is repeated along the same direction until the vacancy arrives at 1NN position to the interstitial counterpart, and then they recombine with each other as described for an 1NN FP. On the other hand, the other types of FP could not recombine within the simulation period of 10 ps.

Table 1. Recombination feasibility of a single oxygen FP

type	distance [nm]	separation	recombination
1NN	0.234	$1/4\langle 111 \rangle$	Yes
2NN	0.449	$1/4\langle 113 \rangle$	Yes
3NN	0.590	$1/4\langle 133 \rangle$	No
4NN	type A	$1/4\langle 115 \rangle$	Yes
	type B	$1/4\langle 111 \rangle$	No
5NN	0.800	$1/4\langle 135 \rangle$	No
6NN	0.887	$1/4\langle 335 \rangle$	No
7NN	type A	$1/4\langle 117 \rangle$	Yes
	type B	$1/4\langle 155 \rangle$	No

The separations of the latter types of FP, or 4NN-type B and 7NN-type B, contain combination of perpendicular $\langle 1/2 00 \rangle$ directions, making the recombination hard in such a short time.

3.2 Aggregation of multiple oxygen FPs

In order to investigate interacting behavior between multiple FPs, plural 7NN-type B FPs were introduced at the same time in the simulations. The MD simulations with double oxygen FPs indicated that the oxygen interstitials come close to each other at 2NN distance from a more distant separation position at 300 and 600 K. The introduction of triple and quadruple oxygen FPs was found to aggregate interstitials. The oxygen vacancies, on the other hand, were settled at the initial positions during the simulations. Fig.2 shows the temporal change of the configuration of four interstitials at 300 K. The four interstitials encircled with white dotted lines aggregate rapidly in 0.06 ps, and then they keep their configuration up to 10 ps. One may recognize in the right figure from the $\langle 112 \rangle$ view that the oxygen interstitials form a plane cluster inserted between two adjusting (111) oxygen layers. This result suggests the tendency of formation of oxygen platelets with $1/9\langle 111 \rangle\{111\}$ type of dislocation loops under electron irradiation at 300

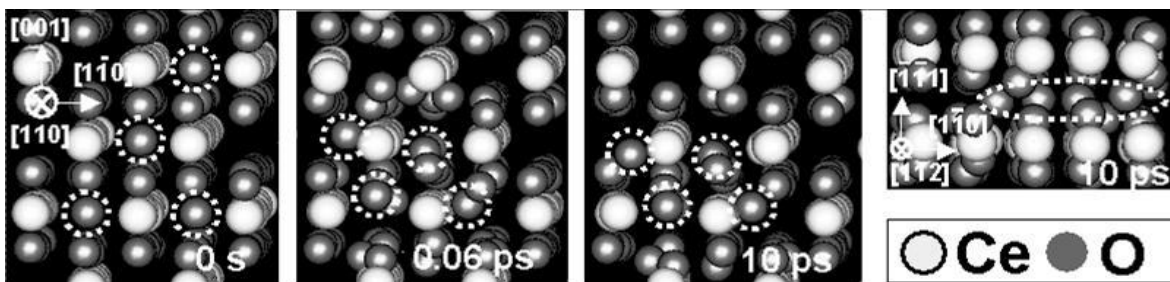


Figure 2. Snapshots of configuration of quadruple oxygen FPs at 0, 0.06 and 10 ps viewed from $\langle 110 \rangle$ and $\langle 112 \rangle$ (only right figure) directions. Oxygen interstitials are encircled by white dotted lines.

K [2]. In contrast, the FPs annihilate by recombination with vacancies in the MD simulations at elevated temperatures 900 and 1200 K following a diffusion process. Correspondingly no incidence of formation of the oxygen platelet is recognized in the TEM observation of electron-irradiated CeO₂ at a temperature higher than 500 K.

The formation of an oxygen platelet between oxygen layers surely results in enhancement of the Coulomb repulsive interactions among the O ions. It was however found that the total lattice energy of the system decreases with the aggregation of oxygen interstitials, stabilizing the platelet. It suggests that the lattice strain energy would play an important role in the formation of the oxygen interstitial platelets.

4. Conclusions

The MD simulations of a single, double, triple and quadruple oxygen FPs were performed to investigate the behavior of oxygen interstitials and vacancies in CeO₂. The results show that an oxygen FP annihilates by recombination of the vacancy and the interstitial after the vacancy jump along the <100> direction. When multiple oxygen FPs are introduced, the interstitials aggregate into a (111) plate-like cluster at relatively lower temperature, such as 300 and 600 K, while they annihilate by recombination with vacancies at elevated temperatures 900 and 1200 K. The results are consistent with the TEM results of formation of 1/9<111>{111} oxygen platelets in CeO₂.

Acknowledgements

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Clustering process and precipitation hardening in Fe-Cu alloys: First-principle and empirical model evaluations

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ABSTRACT

It is well known that the mechanical properties of reactor pressure vessel have changed due to ageing and neutron irradiation. Neutron irradiation enhances the nucleation of lattice defects such as the formation and growth of Cu precipitates. In the present study, the clustering process and interaction between precipitation and pure edge dislocation in Fe-Cu alloys are investigated. Fundamental physical properties associated with classical nucleation theory are first estimated by first-principle calculations based on the projector augmented wave method, and the diffusion processes of impurity ions in α -Fe is comparatively evaluated by first-principle transition state analysis. The heat of solution and the activation energy for the migration of Cu is smaller than other ion species. It is found that these characteristic features are derived from the weak directional influence and low binding energy between valence electrons. On the other hand, a large number of atomistic simulations have been studied to investigate the self-interstitial atom and their clusters in α -Fe under radiation damage environment. We perform a sequence of molecular dynamics simulations to investigate the interaction between a pure edge dislocation and spherical Cu precipitate. The hypothetical rigid body precipitations as well as Cu precipitates are considered for comparison of the characteristics of the precipitation types. Accordingly we can specify the size and spacing effects on the yield stress and the interaction mechanisms between precipitate and dislocation.

1. Introduction

Cu-alloyed steels are widely used as the structural materials for reactor pressure vessel (RPV) in nuclear power plants. It is well known that the mechanical properties of RPV have changed due to ageing and neutron irradiation. Neutron irradiation enhances the nucleation of lattice defects such as the formation and growth of Cu precipitates which result in the hardening and embrittlement of these materials [1, 2]. These self-interstitial atom and resultant Cu cluster exist as barriers to the movement of dislocations under applied stress. The interaction between Cu precipitates and dislocations is one of the most dominant strengthening mechanisms. A large number of studies have been performed to understand the hardening mechanism of hardening and embrittlement derived from Cu precipitate in binary Fe-Cu alloys and RPV steels [1-3].

Recently, hardening effects on the interaction between precipitate and dislocation are investigated by atomistic simulations [4-6]. On the other hand, the reason why Cu impurities peculiarly come together in a cluster and the fundamental process of the formation of these clusters in Fe-Cu alloys have not been sufficiently understood. In the present study, elementary migration process and energetic stability of Cu cluster are comparatively investigated by first principle transition state analysis. In addition, in order to study the hardening and embrittlement processes due to Cu-precipitation, the effects of size and spacing on the yield stress and the mechanisms of cutting and bypass behaviors in spherical precipitation of both Cu and virtually-constructed solid are comparatively investigated via empirical atomistic simulations.

2. Physical mechanism of clustering process

Classical nucleation theory (CNT) [7, 8] associates the onset of a homogeneous phase transition with production of clusters, in which a free energy barrier of the simple form, $\Delta G(r) = (4/3)\pi r^3 \Delta\mu + 4\pi r^2 \gamma$, is predicted. The first term of the right side is the bulk term which is negative and decreases as r^3 , and the second term is the surface term which is positive and increases as r^2 . Impurities are likely to aggregate and make cluster when the interfacial energy is relatively smaller than volume energy. We investigate the interfacial energy and volumetric strain energy by first-principle density functional calculation using Vienna ab Initio Simulation Package (VASP) with Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation (GGA) exchange-correlation density functional, projector augmented wave (PAW) method. When octahedral unit cluster is embedded in the Fe-matrix, the interfacial energy of close-packed octahedral plane is 1.7 J/m^2 , and mismatch strain energy is 0.019 eV/atom . Compared with ground states, strain energy of Cu is found to be smaller than other impurity ions such as Ni and Cr while interfacial energy is not smaller than others. On the other hand, stable interfacial energy is smaller than grain boundary energy of high- Σ coincidence site lattice and random grain boundary structure in α -Fe. Therefore, the homogeneous nucleation favorably occurs in preference to the heterogeneous nucleation at the boundary.

Although the CNT has been an excellent theory to estimate whether the impurities aggregate and make clusters, it is not absolutely enough to explain the clustering of impurity ion in Fe matrix under irradiation environment. Solid solubility limit should be considered in discussing the segregation characteristics. In addition, the nucleation and growth of the solid solution precipitations are influenced by the diffusion rate of impurities in the matrix. We investigate migration energy of impurity Cu and Ni in the Fe-matrix via first-principle transition state analysis. We take the most stable case as a typical example that an impurity moves to the neighboring vacancy in the direction of nearest neighbor atom. The minimum energy path (MEP) of this process is shown in Fig. 1, where atomic configurations are given at the corresponding states. Migration energy of Cu impurities in Fe-matrix is found to be 0.50 eV , which is 0.68% smaller than Ni impurity. The characteristic features of Cu are derived from the weak directional influence and low binding energy between valence electrons, and thus Cu has a greater tendency to migrate between lattice positions than Ni. Similar discussion can apply to the interpretation of low heat of solution and solid solubility limit of Cu in Fe-matrix.

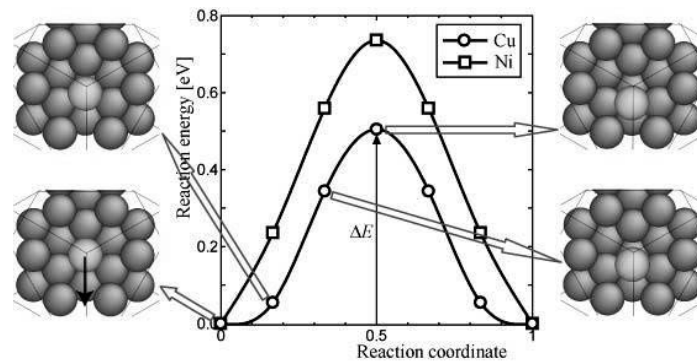


Fig. 1 Minimum energy path of the migration of impurity Cu and Ni.

3. Spacing and size effect of precipitation hardening

It is well known that Cu-rich precipitates cause the increase of yield stress and reduction of ductility which are derived from the interaction of dislocations and Cu precipitates. Recently, precipitate formations of other impurities ions such as Nb and Mo have been observed. Moreover, as dislocations beside the heterophase boundary experience specific Peach-Koehler force depending on the elastic properties of both phases, comprehensive understanding including precipitation type, radius and density should be in order to study the precipitation hardening in Fe-matrix. As the initial step, in this study we consider three types of precipitations, different radii and spacing, where well-known bcc, hypothetical fcc phase and rigid precipitations are embedded in the Fe-matrices, respectively. Many body potential proposed by Ackland et al. is employed to express Fe-Cu interaction, and repulsive potential which produce force component indicated by circle, square and rhomboid, respectively. For purpose of comparison, spacing of d

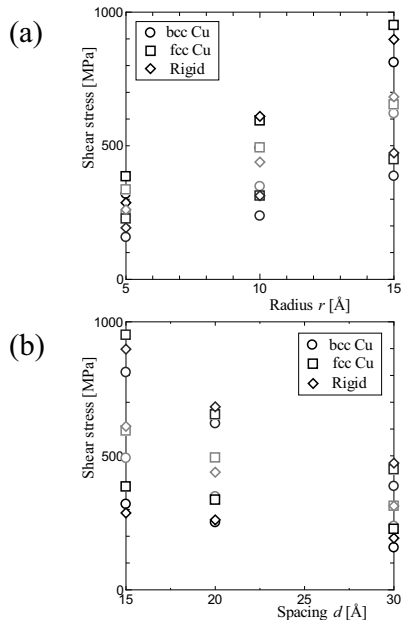


Fig. 2 (a) Critical stress vs. radius and (b) Stress vs. Spacing.

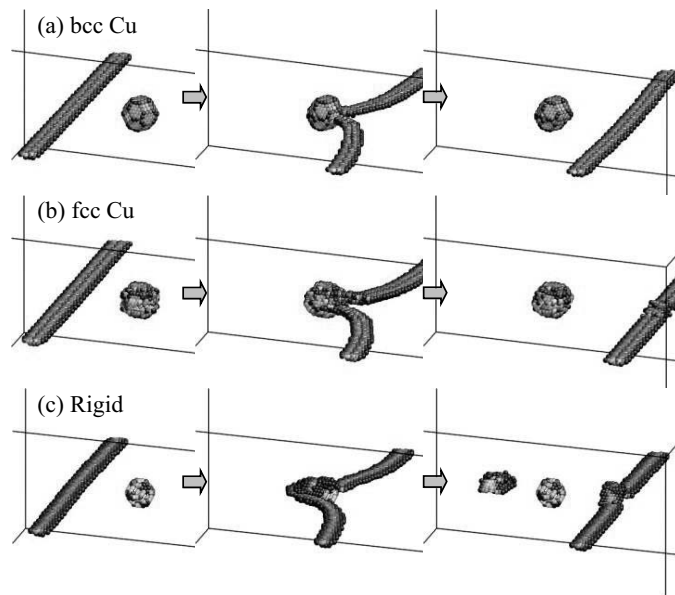


Fig. 3 Atomic images of interaction process between dislocation and three types of precipitation.

= 15 Å (black), 20 Å (red) and 30 Å (blue) in Fig. 2 (a), and radius of $r = 5$ Å (black), 10 Å (red) based on the harmonic function. The relationships of critical stress—radius and stress—spacing are shown in Fig. 2 (a) and (b), respectively, where bcc-Cu, fcc-Cu and rigid precipitation are and 15 (blue) in Fig. 2 (b) are shown. Critical stress is corresponding to the applied stress for dislocation pass through each precipitation. It can be said in all cases that the critical stress increases in approximate proportion to the cluster size, and that they are reduced relative to the spacing. These characteristics are derived wholly from the interaction between dislocation and precipitations including stress distribution around the boundary, lattice inconsistency and mismatched interface. In Fig. 3, atomic images of the same specific radius and spacing ($r = 10$ Å, $d = 30$ Å) in the process of interaction are given to explain the correlation between critical stress and dislocation motion. Interaction between edge dislocation and bcc Cu produces comparatively-weak pinning effect due to the preferable lattice consistency, and dislocation remains unchanged after the interaction. On the other hand, atomic configurations are disturbed after the dislocation pass through fcc Cu precipitation with the strong pinning. Therefore, fcc Cu shows the higher critical stress than bcc Cu. Moreover, rigid precipitation reproduces the highest critical stress among them, where dislocation experiences cross slip on the secondary slip system to overpass the precipitation and prismatic mobile dislocation left behind before the precipitation.

Acknowledgements

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Atomic-scale Modelling of Dislocation-obstacle Interactions in Irradiated Metals

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ABSTRACT

Irradiation of metals with high-energy particles produces nano-scale defect clusters, such as voids, dislocation loops, stacking-fault tetrahedra and irradiation-induced precipitates. They are obstacles to dislocation glide during plastic flow and give rise to hardening and, in some conditions, deformation localisation. Approximations based on the elasticity theory of defects offer the simplest treatment of strengthening, but are deficient in many respects. Atomic-scale computer simulation of dislocation dynamics has therefore been developed to provide detail of mechanisms and quantitative information on the influence of stress, strain rate and temperature which cannot be determined at the continuum level. Recent results of modelling dislocations gliding under stress against obstacles in a variety of metals across a range of temperature are considered. The effects observed include obstacle cutting, absorption and drag. Simulations of OK provide a direct comparison with results from continuum treatment of dislocations, and although some processes can be represented within the continuum approximation, others cannot.

Study of Cavity Evolution in Iron under Neutron and α -Particle Irradiations

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ABSTRACT

The expected high helium generation rate in metals and alloys under fusion irradiation conditions gives rise to a serious concern regarding the performance and lifetime of materials used in the structural components of a commercial fission and fusion reactor. At temperatures above the recovery stage V the concurrent generation of helium and displacement damage may enhance volumetric swelling in the grain interior and may induce grain boundary embrittlement. Although it is well known that the presence of helium enhances the nucleation of voids the mechanisms controlling the scale of nucleation and the level of resulting swelling are still not well understood. The present work was intended to investigate aspects of damage accumulation, in particular cavity formation, at low and higher temperatures (i.e. below and above the recovery stage V) and the role of helium on the cavity formation both experimentally and theoretically. Both un-implanted and He-implanted specimens were irradiated with fission neutrons in the BR-2 reactor at Mol (Belgium) at 50°C and 350°C with a displacement dose rate of $\sim 4.5 \times 10^{-8}$ dpa/s to doses between 0.01 and 0.23 dpa. The experimental investigations were carried out using Positron Annihilation Spectroscopy (PAS) as well as Transmission Electron Microscopy (TEM) and were designed to validate the basis of existing models. Nucleation and growth of He-vacancy clusters were calculated by using a new grouping method for numerical solution of the mean field Master Equation. The calculations are capable of elucidating quantitatively the role of different mechanisms that may be operating during cluster evolution.

First-passage Monte Carlo for materials under irradiation

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ABSTRACT

The key challenge in simulations of irradiated materials is that of time scale. Typically, atomistic simulations extend to less than one nanosecond whereas kinetic Monte Carlo (kMC) simulations struggle to reach hours of simulated irradiation. Based on a time-dependent Green's function formalism, our new kMC algorithm [1] extends the simulated time horizon from minutes to tens and hundreds of years while retaining uncompromising accuracy. This presents an exciting opportunity to extrapolate, through accurate numerical simulations, the material behavior observed under the short and violent irradiation exposures used in the accelerated material tests, to the much longer reactor material lifetimes.

[1] T. Opperstrup, V. V. Bulatov, George H. Gilmer, M. H. Kalos, and B. Sadigh
First-passage monte carlo algorithm: Diffusion without all the hops
Physical Review Letters, **97**(23):230602 (2006)

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An Atomistic Study of Fission Gas Bubble Re-solution in UO₂

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ABSTRACT

With the development of generation IV reactor technology, the impact of intragranular fission gas in uranium dioxide and mixed oxide fuel elements has gained considerable interest. Gaseous fission products and their precipitation (bubble formation), transport, and segregation are known to influence the thermal and mechanical properties of reactor fuels. Release of gaseous species from the fuel can lead to cladding failure at high burn-up. Given the importance of heat removal and the fact that thermal conductivity is influenced by the microstructure, predicting the evolution of the size distribution of fission gas bubbles as a function of temperature and burn-up rate is crucial in determining fuel performance in Gen-IV reactors.

The available analytical bubble population models, discussed in a recent review article by Olander and Wongsawaeng [1], build on fundamental mechanisms for bubble nucleation and re-solution. Two generally accepted mechanisms for re-solution are homogeneous re-solution through single fission gas atom knock-out, and heterogeneous re-solution where a fission product dissolves bubbles instantaneously along its track. These mechanisms, however, have not been directly validated, and furthermore the corresponding cross-sections are determined phenomenologically.

We incorporate these seemingly different mechanisms in a unified description of the ballistic mixing, where, for a given recoil energy, the full histogram of relocation distances is taken into account. Using a combination of classical MD simulations and a binary collision Monte-Carlo software, adapted to support bubble-in-matrix sample geometries, we obtain such a histogram for the system Xe in UO₂.

The contribution of electronic stopping of fast fission fragments in the fuel matrix is modeled by classical MD using a thermal spike approach. A quantitative analysis is performed in light of thermal spikes being a potential physical explanation for a heterogeneous re-solution mechanism.

[1] D. R. Olander and D. Wongsawaeng, “Re-solution of fission gas – A review: Part I. Intragranular bubbles”, *Journal of Nuclear Materials*, **354**, 94 (2006).

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Ab initio Calculations of Defects, Defect Clusters and Defect Creation in SiC and GaN

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ABSTRACT

SiC and GaN have attracted extensive theoretical and experimental interest due to their potential applications for electronic devices. GaN and SiC are both covalent materials, but with GaN being more ionic and SiC more covalent. SiC is of additional interest because of its potential technological applications in high-temperature structural components for fission and fusion reactors. The defects created by energetic particles during ion implantation or neutron irradiation and their subsequent evolution give rise to important microstructural changes that affect many of the macroscopic properties of electronic devices and nuclear components. Large-scale ab initio simulation methods (up to a few thousand atoms) have been developed for the study of ion-solid interactions in materials, and these methods have been employed to investigate defect properties in SiC and GaN. Relative stabilities of these defects have been determined, and their electronic structures provide insight into the configurations and binding properties of these native defects. Atomic structures, formation energies and binding energies of small clusters (both vacancy and interstitial clusters) have been investigated, and their relative stabilities determined. Furthermore, ab initio molecular dynamics methods have been used to calculate threshold displacement energies and to simulate the primary damage states for the PKA (primary knock-on atom) energies up to 1 keV in SiC and GaN. These simulations provide important insights into electronic effects on ion-solid interaction processes, and reveal that significant charge-transfer occurs between atoms. The charge variation of the recoil atom can decrease the energy barrier for stable defect formation, and the corresponding dynamics evolution is a charge-assisted process, which is expected to have significant effects on defect creation in covalent and ionic materials. Thousand-atom ab initio simulation provides a feasible path to study low-energy ion-solid interaction, charge transfer and charge-redistribution, with first-principle accuracy, in covalent materials.

New Approach To Model Point Defects Migration Using A Monte-Carlo Paradigm Coupled With Artificial Intelligence.

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ABSTRACT

In this work, we propose to construct an Atomistic Kinetic Monte Carlo paradigm where the point defects migration barriers are calculated with few approximations on the basis of a Molecular Dynamics (MD) based method. The algorithm is speeded up with an Artificial Neural Network, trained to reproduce the migration barriers on the basis of the Local Atomic Environment, thus avoiding the need of an on-the-fly use of MD.

1. Introduction

It is widely accepted that the formation of copper-rich precipitates plays a key role in nuclear reactor pressure vessel steel hardening and embrittlement [1]. With this respect, any method capable of predicting the evolution of solute atom precipitation versus radiation dose helps at designing or monitoring nuclear reactor components (see e.g. [2][3]).

The Atomistic Kinetic Monte Carlo (AKMC) simulation [4] is a compromise between Molecular Dynamics (MD), which considers events at the atomic time and length scale, and coarse-grained tools, such as Object KMC [5] and rate theory [6], that are necessary to extend the simulation to the macroscopic scale. AKMC techniques retain the atomic level description, but reduce the number of possible events to the very basic mechanisms of single-defect diffusion and can thus encompass a timeframe (much) larger than MD.

In the AKMC algorithm, the events are vacancies and/or interstitials migration at a close neighboring position. The probabilities are calculated with the eqn. (1), where $\nu_{0,j}$ is a prefactor (assumed to be constant as Debye's frequency in Fe), k_B is Boltzmann's constant, T is the absolute temperature and E_j is the Migration Energy (ME) at 0K. The index j denotes the event for which the probability is calculated. The summation over k in the denominator is over all possible events. When the event is chosen, the simulation time is incremented with a mean residence time algorithm.

$$p_j = \frac{v_{0,j} \exp(-E_j/k_B T)}{\sum_k v_{0,k} \exp(-E_k/k_B T)} \quad (1)$$

The ME's are therefore the only unknowns that must be calculated on the fly during the simulation, depending on the instantaneous Local Atomic Environment (LAC) of the migrating point defects. They are classically estimated with empirical formula based on the difference on energy induced by the migration, or numerically fitted on the basis of a limited amount of *ab initio* data.

It is clear that the ME quality (depending on the method used to calculate it) is strongly influencing the kinetic path followed by the system. Empirical formulas based on the energy difference have the advantage to be very easy to do on-the-fly, but give a poor description of the energy barriers, mainly because there are in reality very poorly correlated with the energy difference. On the other extremity, *ab initio* methods are unquestionably the state-of-the-art but can only provide a rather limited amount of calculations because of their inherent complexities.

2. Methodology

In our method, we propose to calculate the ME with an MD based tool. The Nudged Elastic Band (NEB) method [7] optimizes the Minimum Energy Path (MEP) of the migrating particle from the initial to the final state of the system (that are both relaxed with a conjugate gradients method), by constructing a chain-of-states linked with fictive springs. The saddle point in the MEP, corresponding to the ME, is found with a good accuracy. This method therefore provides an automated procedure to calculate the ME, entirely based on the inter-atomic potential used in an MD framework to calculate the total energy of the system.

The NEB method allows us to calculate any ME in a reasonable timeframe. As an order of magnitude, the calculation of the vacancy migration energy takes about 100 seconds on a modern mono-processor machine. This is, however, much too slow to envisage an on-the-fly use in the AKMC simulation. But the generation of considerably large databases of examples, corresponding to original LAC's, is possible.

Once large databases of NEB calculated examples have been generated, we train an Artificial Neural Network (ANN) to construct a mathematical regression between the LAC and the corresponding ME. The ANN input variables are categorical integers, describing the chemical nature of the neighboring atoms situated at fixed positions in a rigid lattice grid of coordinates. Our ANN is a Multi-Layer Perceptron with only one hidden layer, trained with a Levenberg-Marquardt algorithm [8]. Only a small part of the ME database is used as a training set, to adapt the degrees of freedom in the ANN. The rest is used as a validation set, to assess the ANN extrapolation qualities on never seen examples. Once trained, the ANN is by 6 to 7 orders of magnitude faster than NEB in providing an estimate of the ME.

3. Results

Figure 1 shows the ANN quality of predictions for a simple FeCu binary system, with only one vacancy. A database of more than 100,000 randomly generated examples has been NEB calculated. The ANN has been trained using only 15,000 of them, taking the third nearest neighbors of both the migrating atom and its destination into account (that makes 39 variables for a BCC crystallographic structure). The average ANN error measured on the rest of the database is of the same order as the estimated NEB accuracy. We can therefore consider that the use of an ANN, in this case, is fully equivalent than if NEB is used on the fly during an AKMC simulation.

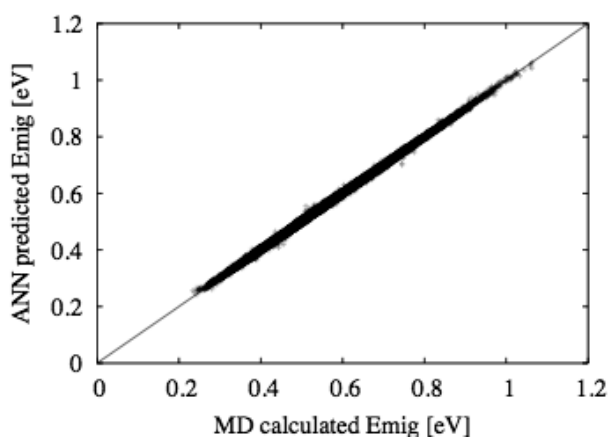


Figure 1 – ANN quality of prediction for the 1nn migration of a vacancy in an FeCu alloy. 39 atomic sites are taken into account. The average error is 0.51% and $R^2 = 0.999$. The points shown were not used for training.

The problem of Self-Interstitial Atom (SIA) migration is much more complicated than for vacancies, because of the strong and highly anisotropic interaction between close ones. We still have some problems to calculate their ME with a sufficient accuracy, because the relaxation of a small system containing several SIA's, with the constraint to keep their orientations and positions, can be a delicate issue. The problem is to guarantee that the migration path found by the NEB method gives truly the lowest possible saddle point energy for a given transition. Meanwhile, we have managed to train the ANN to predict the difference of (relaxed) energy for the 1nn migration of a $\langle 110 \rangle$ dumbbell, in pure Fe with maximum 4 other SIA's in the box, as shown on figure 2. 145 atomic sites were taken into account and 80,000 examples were used for training.

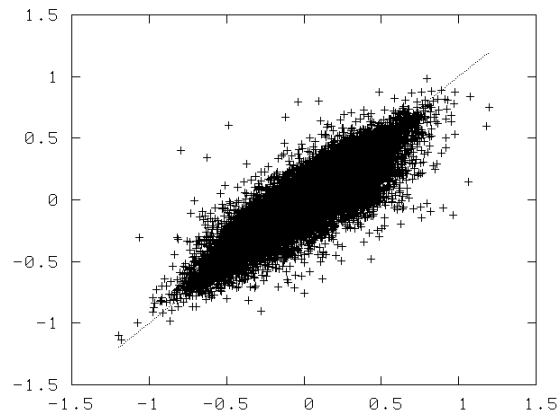


Figure 2 – ANN predictions of the relaxed energy difference for the 1nn migration of a $\langle 110 \rangle$ dumbbell in pure Fe. The X axis is the MD value, the Y axis is the ANN predicted. The average error is 0.07 eV and $R^2 = 0.89$.

Acknowledgements

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Influence of Electronic Stopping on Sputtering Induced by Cluster Impact on Metallic Targets

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ABSTRACT

Using large scale molecular-dynamics simulations, we model the sputtering of a Au (111) crystallite induced by the impact of Au projectiles with total energies up to 500 keV. Due to the uncertainty in particular of the electronic stopping of Au moving at small velocities, we performed several simulations, in which the electronic stopping parameters are systematically changed. Our results demonstrate the dominating influence of the cut-off energy E_c , below which the high-energy electronic stopping of atoms is switched off in the simulation. If E_c is smaller than roughly one half the cohesive energy of the target, sputtering ceases after a few ps; the spike contribution to sputtering (also called phase-explosion, or gas-flow contribution) is entirely quenched and the sputtering yield is up to an order of magnitude smaller than when electronic stopping is taken into account only at higher atom energies. Our results demonstrate the importance of a careful modeling of electronic stopping in simulations of spike sputtering from metals. We discuss the necessity to include the electronic temperature, and a possible multi-scale modeling implementation.

Interaction of dislocations with Cr-rich precipitates: a molecular dynamics study

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ABSTRACT

FeCr alloys are the base matrix of high-Cr ferritic/martensitic (FM) steels for industrial applications. FeCr binary alloys as well as FM steels exhibit α - α' phase separation, if Cr content exceeds 10%at., resulting in the formation of fine-dispersed nano-meter size precipitates, in the range of temperatures relevant for technological applications (500-900K). This precipitates obstruct motion of dislocations leading to additional hardening and risk of embrittlement. In order to better understand the basic mechanisms of hardening and embrittlement of aged steels one can quantify precipitate-dislocation interaction in terms of critical stress and its temperature dependence. In the present work, we apply molecular dynamics (MD) techniques to study the interaction of dislocations with Cr precipitates of different and sizes at different temperatures in Fe and Fe-10Cr matrix (typical content of Cr in FM steels). The corresponding stress-strain curves are thereby obtained and the mechanisms of reactions are investigated. Theoretical models based on elasticity theory, which account for different strengthening mechanisms (size-misfit, modulus misfit and chemical strengthening) are applied, being parameterized using the interatomic potentials. The ultimate goal of the work is to confront the data obtained using both theoretical and simulation methods and thus to reveal prevailing mechanisms of precipitate strengthening in Fe-Cr alloys.

Oxygen interstitial migration in hyperstoichiometric uranium dioxide studied by kinetic Monte Carlo simulations

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ABSTRACT

A kinetic Monte Carlo (kMC) model has been developed to study the point defect mobility and their interaction in hyperstoichiometric uranium dioxide (UO^{2+x}). Kinetic properties of point defects govern radiation tolerance, fission product accommodation, fission gas release and micro-structural evolution in-pile. The kinetics of this evolution will impact the stability of the fuel during service. Under operating conditions, uranium fuel is typically hyperstoichiometric, i.e., there are excess oxygen atoms in the material to accommodate U^{5+} charged ions. In hyperstoichiometric uranium dioxide, UO^{2+x} , the excess oxygen atoms occupy interstitial sites. The thermal conductivity and oxygen diffusivity are inter-related. Under operating conditions, the oxygen atoms re-distribute in accordance with temperature gradients and also serve as carriers for the heat conduction in the uranium fuel rod. Thus, understanding oxygen interstitial diffusion is essential to understanding fuel rod properties both in equilibrium and under operating conditions. The kMC model performs simulations of the oxygen interstitials on the octahedral sites in the fluoride sublattice of the calcium fluoride crystal structure. A blocking model was employed to describe the interactions between two oxygen interstitials i.e., if an oxygen atom was one lattice spacing apart from another oxygen interstitial, their respective paths in those directions were blocked. The diffusivity is calculated by calculating the mean square displacement of the oxygen interstitials. Oxygen diffusivity is calculated as a function of non-stoichiometry and temperature and agrees with experimental values at low non-stoichiometry values.

Irradiation of helium implanted iron studied by multiscale modeling and experiments

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ABSTRACT

We present the results of experimental and modeling studies of the effect of irradiation in He implanted single crystal iron. Microstructural defects are introduced in materials upon irradiation with energetic particles. These defects can cause degradation of mechanical properties and contribute to material failure. Transmuted helium in irradiated stainless steels exerts deleterious effects on material properties. We have performed kinetic Monte Carlo (kMC) simulations of point defect diffusion and clustering in bcc alpha iron. The model includes helium and vacancy diffusion and spontaneous clustering and dissociation of the point defects from the clusters. We employ the kMC simulations to investigate the time evolution of the point defect configuration leading to defect clustering and bubble formation. The concentration of embryonic point defect clusters is determined as a function of the simulation time. The results from the Monte Carlo model illustrate the competition of the vacancies and clusters as sinks for the interstitial He in the system and as possible nucleation sites for bubbles. Order kinetics of transmutation gas mechanisms are studied with the kMC method and analyzed using diffusion limited reaction rate theory. Experimentally, in situ transmission electron microscopy measurements were performed of Fe⁺ ion irradiation of He doped single crystals and polycrystals. The temperatures selected were 300 and 450C with ion energies of 50 and 150 keV. Also, Positron annihilation spectroscopy was used to analyze the open-volume defects created in single crystal, body-centered cubic iron irradiated with 1.0 MeV protons. The effects of irradiation dose and temperature were investigated. A novel technique utilizing a Bremsstrahlung beam to activate and induce positron decay in the bulk specimens, followed by Doppler broadening spectroscopy, was employed. Results from the simulation and experiments will be compared and methods for validating computational models discussed.

Fe-Cu-Ni many-body potential consistent with thermodynamics

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ABSTRACT

The Fe-Cu-Ni ternary alloy is of interest for nuclear applications because Cu and Ni are considered to have major effects on the embrittlement under irradiation of reactor pressure vessel steels. In this work a ternary Fe-Cu-Ni many-body potential consistent with thermodynamics is developed for the first time. Thermal ageing of an Fe-Cu-Ni alloy is simulated using atomistic kinetic Monte Carlo as a validation of the potential.

1. Introduction

It has been recognised for many years that the embrittlement of reactor pressure vessel steels is due to the formation of ultrafine copper-enriched precipitates [1]. In addition to Cu, atom probe analysis has revealed that Ni as well as Mn, Si and P are encountered in these nano-defects. In order to improve our understanding of this radiation induced and enhanced precipitation, large scale atomistic simulations to support experimental observations in Fe-Cu-Ni model alloys are desirable. For such a purpose, a ternary many-body interatomic potential for the Fe-Cu-Ni system is here developed and tested.

2. Methodology

The formalism used to describe the atomic interactions is the embedded atom method (EAM) [2]. Within this approach, each atom is assigned an energy consisting of pair contributions V and an embedding term F , the latter function of the local electron density ρ .

For the pure species, state of the art interatomic potentials from the literature were chosen, so only the cross pair potentials need to be fitted. For Fe, Cu, and Ni the potential developed by Mendeleev et al. [3], Mishin et al. [4] and Voter and Chen [5], respectively, was chosen. Prior to fitting the cross potentials, the pure potentials were transformed into the effective gauge, so that $F(\rho_{eq})=0$ and $\rho_{eq}=1$ for all the species on the bcc lattice. This transformation does not affect any property of the pure species and ensures an equal contribution to the local electron density of every species. At the fitting stage, however, the relative weight of ρ_{eq} between the species is modified, introducing an extra degree of freedom. The cross potentials

for Fe-Cu, Fe-Ni and Cu-Ni were fitted to (i) thermodynamic properties, (ii) kinetic properties, and (iii) point defects in the bcc matrix, using a cubic spline expansion.

In particular, the Fe-Cu cross potential was fitted to the bcc mixing enthalpy and two points of the experimental Fe-Cu equilibrium phase diagram. The latter was realised using the cluster variation method at the fitting stage [6]. The vacancy migration barriers were fitted so that experimentally observed dragging of Cu by vacancies could be reproduced [6]. In addition the binding energy of some defect configurations was fitted to *ab initio* data.

In the case of Fe-Ni special care was taken to make $L1_0$ FeNi and $L1_2$ FeNi₃ the only ground states of the system, as observed in experiments. To do so, both intermetallics were compared to 99 bcc and 87 fcc intermetallics, representing possible ground state structures, at the fitting stage. In addition to the constraints on the formation energies of these intermetallics, the heat of mixing of Ni in bcc Fe and of Fe in fcc Ni were fitted. A correct reproduction of the mentioned properties assures a reasonable thermodynamic behaviour. In addition some vacancy migration barriers and binding energy of defect configurations in the bcc Fe matrix were fitted to *ab initio* data.

Finally, for Cu-Ni the fcc mixing enthalpy was fitted, which proved sufficient to closely reproduce the experimental Cu-Ni miscibility gap, and also *ab initio* data was used to fit the binding energy of some Cu-Ni complexes in the Fe matrix.

3. Results

As a validation of the ternary potential, an attempt was made to follow the microstructural evolution of alloys Fe-1.13at.%Cu and Fe-1.13at.%Cu-1.36at.%Ni under thermal ageing, for which experimental data is available [7]. From these experiments and others, it has been shown that Cu-precipitates are coherent with the bcc Fe matrix up to a size of 4-5 nm, then transform into 9R and eventually into the fcc structure. It was also found that in the case of Fe-Cu-Ni, the Cu-precipitates were enriched with Ni at their surface.

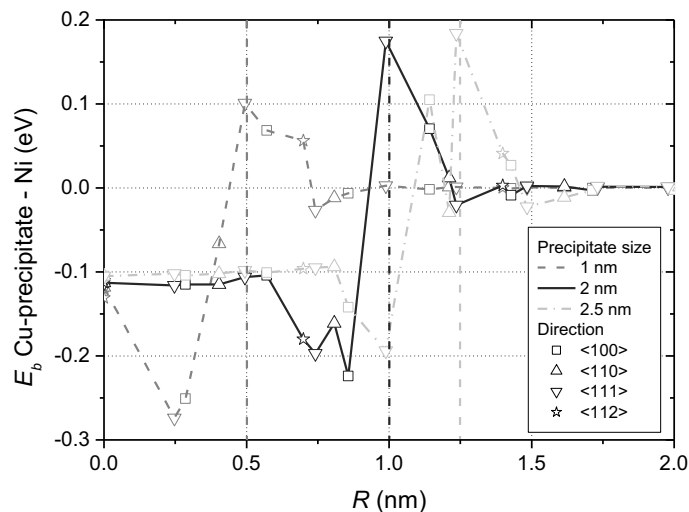


Figure 1. Binding energy between a Ni-atom and Cu-precipitate vs. distance.

Initially, molecular statics calculations were performed to examine the interaction between a Cu-precipitate and a Ni atom in the bcc Fe matrix. To do so, the binding energy (positive values meaning attractive interaction) between a Ni atom and a Cu-precipitate (of size 1, 2 and 2.5 nm) was calculated as a function of the distance between the Ni atom and center of the Cu-precipitate. To increase statistics, the Ni atom was moved along different crystallographic directions ($\langle 100 \rangle$, $\langle 110 \rangle$, $\langle 111 \rangle$ and $\langle 112 \rangle$). The results of these calculations are presented in Fig. 1. The figure clearly shows a strong, attractive, and highly localized interaction at the precipitate surface. For a Ni atom inside the precipitate, the interaction is always repulsive. Fig. 1 is in clear agreement with the experimental observations of Ni-enrichment at the Cu-precipitate surface. It is important to note that the unrelaxed binding energy is almost coincident with the relaxed one presented in Fig. 1, suggesting that the binding of Ni on the Cu-precipitate surface is a chemical effect, rather than an effect due to the stress field around the (oversized) precipitate. Finally, note that results are presented for precipitate sizes up to 2.5 nm only. For larger precipitates the coordination number of Cu atoms increased under relaxation, pointing towards a crystallographic transformation. Therefore our potential predicts Cu-precipitates to be coherent with the bcc Fe matrix up to 2.5-3 nm.

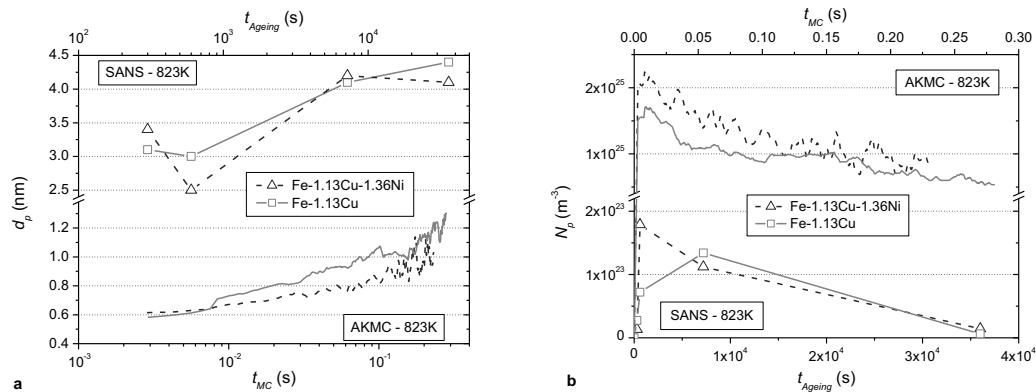


Figure 2. (a) Average precipitate diameter, and (b) precipitate density , as a function of time.

On a 2nd stage, the thermal ageing of the above alloys was simulated using atomistic kinetic Monte Carlo (AKMC). AKMC was performed on cubic samples containing 128,000 atoms. The atomic redistribution was driven by single vacancy jumps with local barriers estimated using a neural network trained on the present Fe-Cu-Ni potential [8].

The results of the AKMC simulations in terms of average precipitate diameter, d_p , and density, N_p , are presented in Figs. 2a and 2b, respectively. In the same figures the experimentally obtained values [7] are superposed. From the experimental results presented in Fig. 2a it is clear that the addition of 1.36 at.% Ni to Fe-1.13at.%Cu has little or no effect on the evolution of the precipitate size. Clearly, our results are in qualitative agreement with that observation. In quantitative terms, however, the precipitate size is underestimated, which is due to the limited box size used for the simulations. The experimental precipitate densities presented in Fig. 2b suggest an increase in the peak density by about 34% due to the addition of 1.36 at.% Ni. With increasing ageing time, t_{Ageing} , however, the effect of Ni on the density diminishes to zero. Both trends are clearly reproduced by the AKMC results, showing a 29% increase in peak density due to the addition of Ni and a fading effect with increasing MC time, t_{MC} . Quantitatively, however,

the precipitate density seems to be overestimated by two orders of magnitude, again, due to the limited box size.

Additional investigation of the AKMC configurations has shown that the Cu-precipitates were enriched with Ni at the surface, in agreement with Fig. 1. It was also observed that Ni was expelled from the Cu-precipitates as soon as the Cu-rich clusters became stable, estimated to be at a size of 5-8 atoms.

4. Conclusions

An embedded atom method interatomic potential for the ternary Fe-Cu-Ni alloy was developed with the emphasis on the correct reproduction of the microstructural evolution of the alloy under thermal ageing and irradiation. The potential was validated using molecular statics and atomistic kinetic Monte Carlo simulations, and a qualitative agreement with experiments was established. In particular, Cu-precipitates were found to be enriched by Ni on the precipitate surface and the effects of the addition of Ni on average precipitate size and density showed similar trends as observed in experiments.

5. Acknowledgements

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KMC Simulations for Formation Kinetics of Vacancy Clusters in beta-SiC during Irradiation

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ABSTRACT

SiC/SiC composites are one of candidates for the structural material of future fusion reactors. Currently, experimental investigations of the property change and performance of this material due to irradiation are extensively done to construct material's database required for reactor design. Recent experiments show that the clusters of vacancies (i.e., voids) are formed in the material during irradiation at higher temperatures than 1000 °C. This lower temperature limit is surprisingly much greater than that for void formation in metals. The objective of this study is to clarify the formation kinetics of void formation in SiC during irradiation. Radiation damage processes are essentially multiscale phenomena, which occur over a wide spectrum of time and length scales. Hence, linking several computer simulation techniques are often made to understand these complicated, athermal, multiscale processes. In the present study, molecular dynamics (MD) and molecular static (MS) calculations with the empirical interatomic potential functions developed for beta-SiC [1] were performed to obtain the formation energy of voids as a function of their size and vacancy composition (i.e., the fraction of the number of Si-vacancies to that of C-vacancies *in voids*). The formation energy obtained here was used to evaluate the binding energy of a vacancy to voids, which determines the thermal stability of voids. It is found that voids with the fraction of 1 show the most stable configuration when void size is given. Also, kinetic Monte-Carlo (KMC) simulations were performed to investigate the nucleation and growth process of voids as a function of the concentrations of vacancies *in the matrix* and temperature. Independent on the fraction of the concentration of Si-vacancies to that of C-vacancies *in the matrix*, voids grow while keeping the vacancy composition of voids as 1. Thus, linking several methods is useful to understand multiscale processes in materials during irradiation.

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MD simulations for defect properties in β -SiC under irradiation – Energetics of interstitial clusters –

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ABSTRACT

Molecular dynamics and molecular static calculations have been performed to evaluate the formation energy of self-interstitial atom (SIA) clusters in β -SiC. An attempt has been made to formalize the formation energy of disc-shaped SIA-clusters with stoichiometric composition using a polynomial function of cluster size n . Resultant equation $E_F=0.94n^1+3.29n^{1/2}$ may indicate the applicable capability to a wide range of cluster size.

1. Introduction

Cubic silicon carbide (β -SiC) is a component of SiC/SiC composites that are candidates for blanket structural materials of nuclear fusion reactor because of high mechanical strength and low activation properties, where β -SiC has the zincblende structure with the stacking sequence of the silicon-carbon dimmers, described as ABCABC... along the $\langle 111 \rangle$ direction. A variety of experiments have been performed to evaluate the irradiation effects in β -SiC. Some experiments using transmission electron microscopy show that SIA-clusters with the diameter of 2.0~3.6 nm are observed in irradiated β -SiC at higher temperatures than 600 °C [1-3]. Such microstructural changes cause the degradation of material's properties; therefore, they should be predicted and controlled. However, formation kinetics of SIA-clusters in β -SiC is not yet understood well.

In the present study, an atomistic calculation was performed to investigate energetics of SIA-clusters in β -SiC, which is important to clarify the formation kinetics of the clusters during irradiation.

2. Procedure

Molecular dynamics (MD) and molecular static (MS) calculations were performed to evaluate the formation energy of SIA-clusters in β -SiC. The empirical interatomic potential employed for the present atomistic calculations was the Gao-Weber potential [4] which was developed to provide a good description of self-interstitials' properties in β -SiC. Firstly, an SIA-cluster was introduced into a computational box with the constant volume under 3-D periodic boundary condition. The box size was chosen to be $10a_0 \times 10a_0 \times 10a_0$ and $18a_0 \times 18a_0 \times 18a_0$ for the investigations of relatively small SIA-clusters and relatively large SIA-clusters, respectively, where lattice constant $a_0=0.436$ nm. The computational system was then fully relaxed by keeping it at finite temperatures ranging from 100 K to 1500 K. And then, the system was quenched to 0 K to obtain the converged value of total potential energy. It is noticed that this combined relaxation method [5] of MD and MS techniques can provide more relaxed configuration of

defects than those provided from a simple static relaxation method [4]. In fact, the lowest formation energy of defects in materials was obtained by this method, as described elsewhere [5,6].

Formation energy of an SIA-cluster is defined as energy required for introducing the cluster into an otherwise perfect crystal. When n_I^{Si} and n_I^{C} denote the numbers of silicon- and carbon-interstitials in an SIA-cluster, respectively, the formation energy of the cluster is given by

$$E_F(n_I^{\text{Si}}, n_I^{\text{C}}) = E^{\text{tot}}(N_{\text{Si}}, N_{\text{C}}) - N_{\text{Si}}\varepsilon_{\text{Si}} - N_{\text{C}}\varepsilon_{\text{C}}, \quad (1)$$

where N_{Si} and N_{C} denote the total numbers of silicon atoms and carbon atoms in the system, respectively. $E^{\text{tot}}(N_{\text{Si}}, N_{\text{C}})$ is the total potential energy of fully relaxed system mentioned above, $\varepsilon_{\text{Si}} = -6.21$ eV and $\varepsilon_{\text{C}} = -6.61$ eV are the calculated cohesive energies of a silicon atom and a carbon atom in β -SiC, respectively. An SIA-cluster formed by n_I^{Si} silicon interstitials and n_I^{C} carbon interstitials is hereafter described as SIA($n_I^{\text{Si}}, n_I^{\text{C}}$). Notice that SIA(1, 0) and SIA(0, 1) denote isolated silicon- and carbon-interstitials, respectively.

3. Results and Discussion

The calculated lowest formation energies of isolated silicon- and carbon-interstitials are 3.17 eV and 3.24 eV, respectively, and the most relaxed configurations of isolated interstitials are shown in Fig. 1. The most stable configuration of isolated silicon interstitial is the TC configuration where a silicon atom is located at the tetrahedral position surrounded by four regular carbon lattice atoms [4]. On the other hand, the energetically favorable configuration for isolated carbon interstitial is the C-C dumbbell configuration in which two carbon atoms share a regular carbon lattice site along $\langle 100 \rangle$ direction [4]. These stable configurations are consistent with those obtained by our previous ab-initio calculation [7].

The calculated lowest formation energy of SIA-clusters with the cluster size $n = n_I^{\text{Si}} + n_I^{\text{C}} = 2 \sim 6$ are also listed in Tab. 1. The formation energy strongly depends on the size and chemical composition of clusters. As to the most stable configuration of the clusters, surprisingly, each silicon interstitial in the clusters also maintains the TC configuration, while each carbon interstitial in the clusters maintains the C-C dumbbell configuration, as well.

Also, preliminary calculations were conducted to obtain the formation energy of relatively large SIA-clusters. Fig. 2(a) shows

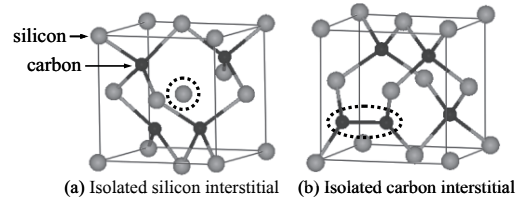


Figure 1. The most relaxed configuration of isolated (a) silicon- and (b) carbon- interstitials obtained from the MD and MS calculations.

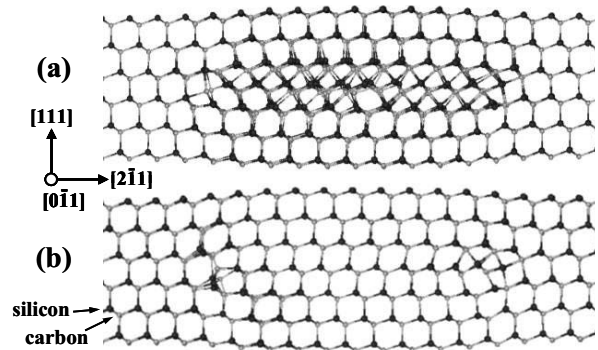


Figure 2. The configuration of SIA(91, 91) relaxed at (a) 300K and (b) 600K obtained from the MD and MS calculations. Some preliminary calculations show that a metastable SIA(91, 91) is a dis-shaped defect containing stacking fault on the closed-packed (111) plane, as shown in (a). When the relaxation is done at a higher temperature, the stacking fault is eliminated and the SIA-cluster is unfaulted to be a perfect loop, as indicated in (b).

the configuration of SIA(91, 91) which was relaxed at 300K. As shown in the figure, the SIA(91, 91) is a disc-shaped defect containing stacking fault on the closed-packed (111) plane, with the estimated diameter of 2.1 nm. When the relaxation was done at a higher temperature, the stacking fault was eliminated and the SIA-cluster was unfaulted to be a perfect loop, as indicated in Fig. 2(b). In order to clarify the mechanism of the unfaulted process, further investigation is underway.

The calculated lowest formation energies of SIA-clusters with stoichiometric composition ($n_I^{\text{Si}}/n_I^{\text{C}}=1$) are plotted in Fig. 3, where the formation energy is an increasing function of n . An attempt was made to formalize the formation energy of SIA-clusters with $n_I^{\text{Si}}/n_I^{\text{C}}=1$ using the following equation:

$$E_F = \pi R^2 \gamma^{\text{SF}} + 2\pi R \gamma^{\text{DIS}}, \quad (2)$$

where an SIA-cluster is assumed to be a disc-shaped defect with the radius R , γ^{SF} is the stacking fault energy, and γ^{DIS} is the energy for dislocation line per length. R has a relationship with n , as $\pi R^2 b = n\Omega$, in which Ω is the mean atomic volume, and b is the thickness of the disc in the $\langle 111 \rangle$ direction. By the relationship between R and n , Eqn. (2) is rewritten as follows:

$$E_F = An^1 + Bn^{1/2}, \quad (3)$$

where $A = \gamma^{\text{SF}} \Omega / b$, and $B = 2\gamma^{\text{DIS}} (\pi \Omega / b)^{1/2}$. The fitting of Eqn. (3) to the formation energies of SIA(91, 91), SIA(127, 127) and SIA(217, 217) leads to $A=0.94$ and $B=3.29$, and the resultant fitting curve is plotted in Fig. 4. Extrapolation of the fitting curve into the smaller region shows a close value to the calculated energy of small clusters. It may indicate that the derived equation is applicable to a wide range of cluster size.

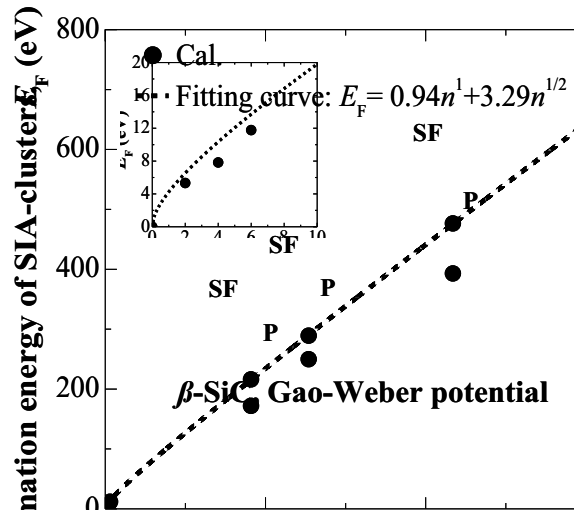


Figure 3. The size dependence of formation energy of SIA-clusters with stoichiometric composition ($n_I^{\text{Si}}/n_I^{\text{C}}=1$). The black dots denote the calculated lowest formation energies listed in Tab. 1. The symbol “SF” denotes a disc-shaped defect containing stacking fault on the closed-packed (111) plane. The symbol “P” denotes a defect unfaulted to be a perfect loop. The broken line denotes the fitting curve $E_F=0.94n+3.29n^{1/2}$.

Table 1. The calculated lowest formation energy of SIA-clusters formed by n_1^{Si} silicon- and n_1^{C} carbon-interstitials in β -SiC in eV, $E_{\text{F}}(n_1^{\text{Si}}, n_1^{\text{C}})$. The superscript “SF” denotes a disc-shaped defect containing stacking fault on the closed-packed (111) plane. The superscript “P” denotes a defect unfaulted to be a perfect loop.

		No. of silicon interstitials in a cluster, n_1^{Si}							
		0	1	2	3	4	91	127	217
No. of carbon interstitials in a cluster, n_1^{C}	0	0.00	3.17	4.41	6.32	6.98			
	1	3.24	5.33	6.85	8.15				
	2	5.07	7.27	7.83					
	3	3.89	8.80		11.77				
	4	7.69							
	91						216.07 ^{SF} 172.00 ^P		
	127							289.13 ^{SF} 249.77 ^P	
	217								476.45 ^{SF} 392.88 ^P

4. Summary

Formation energy of SIA-clusters in β -SiC was evaluated by means of the MD and MS combined technique with the Gao-Weber potential. The most stable configurations of isolated silicon- and carbon-interstitials are the TC- and C-C dumbbell-configurations, respectively. These configurations are maintained in the relatively small SIA-clusters. As to the relatively large SIA-clusters, a metastable cluster is a disc-shaped defect containing stacking fault on the closed-packed (111) plane. When the relaxation is well done, the stacking fault is eliminated and the SIA-cluster is unfaulted to be a perfect loop. An attempt was made to formalize the formation energy of disc-shaped SIA-clusters with stoichiometric composition using a polynomial function of cluster size n . Resultant equation $E_{\text{F}}=0.94n^1+3.29n^{1/2}$ may indicate the applicable capability to a wide range of cluster size.

Acknowledgements

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The calculating analysis of the relationship between specialities of gamma spectrum and source condition

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ABSTRACT

The message of shape, structure and the corresponding condition of experiment has been comprised in the gamma spectrum of the radioactive material, exploring the relationship between the specialities of gamma spectrum and the shape, structure and the wall thickness of measured source has great significance for the nondestructive verification and authentication of radioactive material. In this paper, The specialities of gamma spectrum of radioactive material with special shape and structure has been simulated by Monte Carlo method, in special axis line, the spatial distribution of gamma counting rate of radioactive material with different shape, size and the thickness of material has been calculated, and the relationship between linear spatial distribution and these different factors has been summarized. The relationship between the intensity ratio of gamma-rays with different energy in the spectrum and the value of wall thickness has been explored. The research results were described as follows. 1 The linear spatial distribution of counting ratio was sensitive to the size of source, and was not sensitive to the shape and wall thickness of source; 2 The message of the value of wall thickness of measured source could be acquired according to the intensity ratio of the gamma-rays with 413 and 769keV when the thickness of outer shield material has been known.

Characterization of dislocation loops, voids and Cr precipitates as competing sources of hardening in irradiated Fe-9Cr alloys

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ABSTRACT

High-Cr ferritic-martensitic (FM) steels being candidates for structural materials in advanced nuclear systems are expected to suffer from hardening and embrittlement induced by neutron radiation accumulating during operation. In reactor conditions, formation of nano-defects such as dislocation loops, voids and secondary phase particles (Cr-rich precipitates) penalizing dislocation motion is the main source of hardening in high-Cr FM steels. In order to develop predictive models of hardening and embrittlement it is important to quantify the resistance that each type of defect opposes to dislocation motion and to characterize the mechanism of reaction. Here we use molecular dynamics techniques to study the interaction of dislocations with interstitial dislocation loops (with both $\langle 100 \rangle$ and $\frac{1}{2} \langle 111 \rangle$ Burgers vectors), voids and Cr-rich precipitates, of sizes from 0.5 nm (invisible in transmission electron microscopy (TEM)) up to 10 nm (clearly visible in TEM), in FeCr alloys with Cr content equal to 9% . The corresponding stress-strain curves are thereby obtained and the mechanisms of reactions are investigated at different simulation conditions.

The obtained results show that, for a given defect size, voids are the strongest obstacles to dislocation motion, while dislocation loops can be easily absorbed. Large dislocation loops, however, can be absorbed only partially, depending on the mutual orientation of the Burgers vectors. As a general statement, $\frac{1}{2} \langle 111 \rangle$ loops are stronger obstacles than $\langle 100 \rangle$ loops. Precipitates appear to be weaker obstacles, but their effect will be non-negligible because of their expected high density.

Influence of helium on the clustering of self-interstitials in irradiated bcc iron

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ABSTRACT

The interaction between helium and self-interstitial clusters produced during displacement cascades have been investigated using molecular dynamics simulations. A fine analysis of the defects and clusters has shown that helium affects both the production of Frenkel pairs and the formation of self-interstitial clusters. In particular, helium essentially stabilizes self-interstitial clusters and hence favors the formation of larger clusters. However depending on its nature interstitial or substitutional and its concentration in the material, it has been demonstrated that helium can noticeably influence the structure of self-interstitial clusters. Contrary to pure iron, irradiation temperature plays an important role on the growth of these clusters in presence of helium.

Atomistic Modeling of the Interaction between Dislocation and Vacancy in Tungsten

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ABSTRACT

The interaction between the $\frac{1}{2}\langle 111 \rangle$ screw dislocation and a vacancy in body-centered cubic (bcc) metal tungsten is studied by means of atomistic simulations. In order to investigate the influence of the vacancy on dislocation mobility, the Nudged Elastic Band method is implemented to determine the changes of the energy barrier for the dislocation motion. Our simulation results show that the dislocation mobility may be strongly influenced by the presence of vacancies and the change of the energy barrier depends sensitively on the vacancy position. A comparison of the simulation results with predictions of the elasticity theory shows an excellent agreement between the two approaches when the separation between the vacancy and the dislocation core is larger than 1 nm but large deviation from the elastic limit when the vacancy approaches the dislocation core.

1. Introduction

It is well known that the plastic deformation of metals is primarily governed by dislocation generation and motion. While the onset of plastic yield corresponds to triggering of dislocation motion, subsequent hardening is mainly controlled by interaction of gliding dislocations with other lattice defects. These may include individual point defects and their clusters as well as other extended defects such as dislocations, grain boundaries, interfaces, and surfaces. Point defects can be considered as the simplest crystal defects that are always present within the material. Experimental results however show that if the density of point defects increases, e.g., under irradiation conditions, the yield point and the flow stress also change significantly. These changes of macroscopic properties are directly related to processes at the atomic scale where the dislocation motion becomes strongly influenced by the numerous pinning points. Unfortunately, such microscopic processes are difficult to be observed and analyzed experimentally. In the present work we employ atomistic simulations together with the nudged elastic band (NEB) [1] technique to study the interaction between the $\frac{1}{2}\langle 111 \rangle$ screw dislocation and a vacancy in bcc W.

2. Methods

The atomistic simulations in this work are performed using the Finnis-Sinclair potential [2,3]. This interatomic potential has been widely used in studies of crystal defects and even though it exhibits deficiencies in the description of dislocation core structure it serves as a valuable tool for a systematic qualitative analysis of complex phenomena and testing of computational approaches.

In the simulation cell used in the current study, the x , y and z axes are parallel to the $[11\bar{2}]$, $[1\bar{1}0]$ and $[111]$ directions, respectively. A straight screw dislocation with its line direction parallel to the z direction is introduced in the centre of the simulation block by displacing the atoms according to the anisotropic elastic displacement field of a screw dislocation in an infinite medium. Periodic boundary conditions are applied in the z direction with the size of the simulation cell set to $4b$. Within this setup the dislocation line remains straight during the simulation. The block is then relaxed, with outmost layers in the x and y directions fixed. After relaxation, the dislocation core possesses a three-fold symmetry and extends into three intersecting $\{112\}$ planes [4,5]. Due to the symmetry of the lattice two degenerate core configurations exist that are schematically shown in Fig. 1. The vacancy is introduced by removing an atom at different sites of the dislocated lattice.

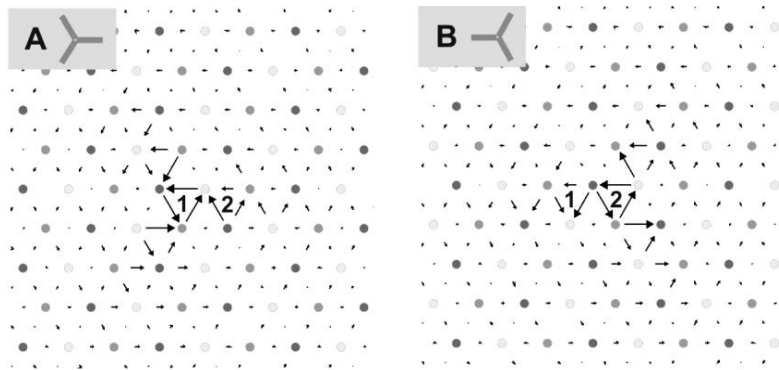


Figure 1. Differential displacement plots of two degenerate core configurations (at neighboring sites 1 and 2) in BCC W.

To study the influence of the vacancy on the dislocation mobility, the NEB method is used. The NEB method, rooted from transition-state theory, is an efficient technique for finding the minimum energy path (MEP) between two states. With this method, the transition between the neighboring dislocation positions is represented by an elastic band consisting of a set of replicas (intermediate states of transition) connected by springs. Force projections parallel and perpendicular to the entire band serve to relax the image configurations and to obtain the MEP. In the present study, only the A-to-B transition (see Fig. 1) is studied. The intermediate images are constructed by linear interpolation between the two neighboring core structures.

3. Results and Discussions

The first quantity of interest is the binding energy of the vacancy in the crystal containing a dislocation. This binding energy indicates whether it is more favorable for the vacancy to remain in the bulk or in the vicinity of the dislocation. The binding energy between the dislocation and the vacancy can be expressed as:

$$E_{bind} = (E_{vac,dislo} - E_{dislo}) - (E_{vac} - E_{ideal}), \quad (1)$$

where E_{ideal} is the total energy of ideal block without any defects and E_{dislo} , E_{vac} , $E_{vac,dislo}$ are the total energies of blocks containing a single dislocation, a single vacancy, and both the dislocation and the vacancy, respectively.

The computed vacancy binding energies as a function of the vacancy position are displayed in Fig. 2(a). From this binding energy map we can conclude two things. First, the binding energies between the vacancy and the straight screw dislocation are always negative, indicating that the vacancies will tend to be attracted by dislocations. Second, the most attractive positions for vacancies are located in the arms of the dislocation core.

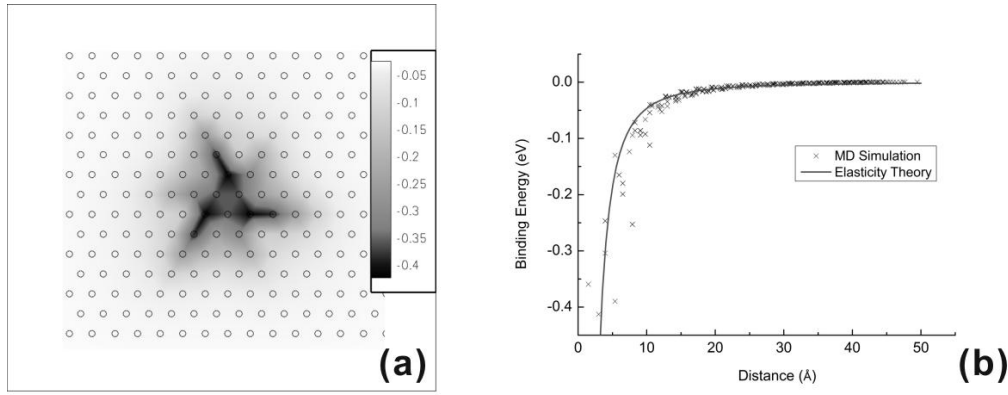


Figure 2. (a) Dislocation-vacancy binding energy map (in eV). (b) Comparison of the binding energy as a function of separation between the dislocation and the vacancy from atomistic simulations and elasticity theory.

In elasticity theory [6], the interaction between dislocation and vacancy can be written as a sum of the first-order size interaction, the second-order size interaction and the inhomogeneity interaction. For a screw dislocation the first-order interaction equals to zero. In addition, the lattice dilation due to the vacancy in W is very small and thus the second-order term can be also neglected. The elastic interaction is therefore in our case composed only of the inhomogeneity term as:

$$E_b^{Elas} = -\frac{5\mu b^2 r_0^3 (1-\nu)}{2\pi(7-5\nu)} \frac{1}{r^2} \quad (2)$$

In Fig. 2(b), we compare the binding energies from the elasticity theory and from the atomistic simulations. We can see that when the separation between the dislocation and the vacancy is larger than about 10 Å, the atomistic results agree well with the elasticity prediction. For shorter

separations the calculations reveal that the elasticity theory is no longer applicable and atomic-level interactions have to be taken into account. It should be noted that due the periodic boundary condition along the dislocation line the vacancy is also repeated infinitely in the z direction with a separation distance of $4b$. However, since the stress field of a point defect vanishes much faster ($\sim 1/r^3$) than the stress field of a dislocation ($\sim 1/r$), the interaction between the point defects can be neglected in both Eqs. (1) and (2) [7].

To study the influence of the vacancy on the dislocation mobility, we calculated the energy barrier of the A-to-B (see Fig. 1) transition using the NEB method. Both transitions for the dislocated lattice with and without vacancy were studied. When no vacancy is present, the predicted barrier energy is 0.090 eV/b. The change of the energy barrier due to the introduction of a vacancy is illustrated in Fig. 3. From the differential energy map we can conclude that the presence of vacancy may lead both to lowering and increase of the energy barrier for dislocation motion. The effect is rather significant and depends strongly on the separation distance and relative position between the vacancy and the dislocation. The vacancy positions that alter the barrier most lie again in the arms of dislocation cores. However, whether the barrier is lowered or increased depends sensitively on the vacancy position. While vacancies located in the arms of the initial core pointing away from the final dislocation configuration lead to an increase of the energy barrier, those in the arms of the final core will facilitate the dislocation movement.

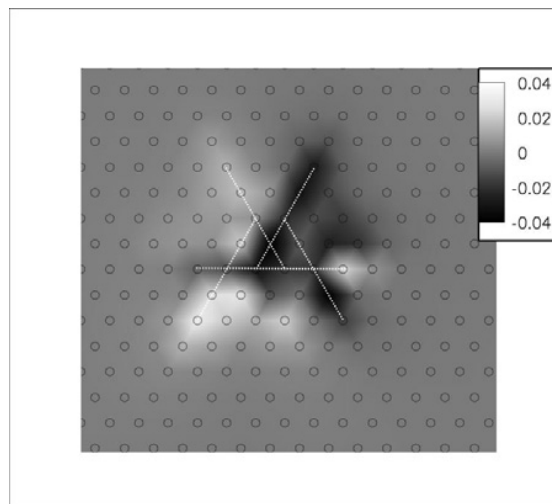


Figure 3. Change of the energy barrier for dislocation transition from A to B configuration as a function of vacancy position (in eV).

Our simulations are expected to give a detailed understanding of the fundamental steps of mechanical deformation processes and to assess the influence of the vacancy obstacle character on the dislocation mobility. Additionally, the atomistic findings can be used for better parameterization of dislocation mobility laws in mesoscopic modeling approaches such as discrete dislocation dynamics simulations.

Acknowledgements

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Kinetic Monte-Carlo simulation of Irradiation Damage Accumulation in Strain Field of Dislocation

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ABSTRACT

Accurate prediction of neutron irradiation embrittlement of highly-irradiated reactor pressure vessel (RPV) materials is very important for the structural integrity assessment of RPVs under long term operation. We have performed a computer simulation study to understand the radiation damage accumulation in bcc Fe using the kinetic Monte-Carlo (KMC) simulation technique. In the conventional approach, pre-existing dislocations were modeled as the sinks of point defects and the simulations were performed up to low to medium doses. However, in the high dose region, experimental results show that the decoration of pre-existing dislocations by dislocation loops occurs, and a new KMC model that can appropriately consider the interaction between the pre-existing dislocations and dislocation loops is required. In our study, a new KMC code is developed considering the change in strain energy due to the interaction between a dislocation and a dislocation loop. The migration energy change caused by the interaction between a dislocation and a dislocation loop is estimated by estimating the interaction energy. We applied the new KMC to the estimation of irradiation damage accumulation in bcc-Fe.

Modeling Study of Grain Sub-division Observed at High Burnup Nuclear Fuel

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ABSTRACT

Nuclear fuel pellets with high burnup undergo a characteristic grain sub-division in their peripheral region, and it is considered that this could increase the fission-gas release from the pellets in case of reactivity insertion accidents. Although the mechanism of this phenomenon has not been fully understood, a subgrain formation through dislocation patterning is widely recognized as a plausible scenario. However, modeling tools which view the microstructure based on the above scenario have been missing because dislocation pattern formations, in general, including those appearing in deformed metals are still too complex to have recourse to computational dislocation dynamics. This difficulty originates mainly in the long-range interaction between dislocations, which gives rise to dislocations' patterning as well as their disordering at the same time. In this paper, a new modeling method has been proposed to numerically construct microstructures caused by subgrain formations. The essence of the modeling is a balance between the two driving forces mentioned above, that is, pushing the dislocations into alignments and disorganizing them. The microstructure that realizes this balance is numerically searched for by using Voronoi tessellation and crystallographic rotation. The methodology can be generally applied to not only nuclear fuels but also deformed metals.

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Modelling of isochronal annealing in bcc metals

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ABSTRACT

From isochronal annealing experiments it is well known that Fe has a very particular behaviour compared to the other bcc metals. Thus three groups can be identified. For the 5B (V,Nb,Ta) elements, the recovery is almost athermal; for the 6B (Cr,Mo,W) elements, the recovery starts at 40-27K while in Fe it starts at 120K. We have here studied the interstitial stabilities of the bcc metals using density functional theory. The self-interstitial ground state for the 5B and 6B groups has hitherto been believed to be of $\langle 111 \rangle$ type. We here show that the 6B group is quite different and that new low-symmetry ground states are predicted that explain the observed difference with respect to the migration of self-interstitials in the 5B group. The information provided by the *ab initio* calculations has furthermore been used as input parameters for kinetic Monte-Carlo simulations of isochronal annealing of the bcc metals in order to compare to experimental results. The consequences for radiation damage in these metals are discussed.

A part of the calculations were performed on the supercomputers at Centre de Calcul Recherche et Technologie (CCRT) in the framework of an EDF-CEA contract.