Multi-scale Modeling of Radiation Damage: Large Scale Data Analysis

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Abstract. Modification of materials in nuclear reactors due to neutron irradiation is a multi-scale problem. These neutrons pass through materials creating several energetic primary knock-on atoms (PKAs) which cause localized collision cascades creating damage tracks, defects (interstitials and vacancies) and defect clusters depending on the energy of the PKA. These defects diffuse and recombine throughout the whole duration of operation of the reactor, thereby changing the micro-structure of the material and its properties. It is therefore desirable to develop predictive computational tools to simulate the micro-structural changes of irradiated materials. In this paper we describe how statistical averages of the collision cascades from thousands of MD simulations are used to provide inputs to Kinetic Monte Carlo (KMC) simulations which can handle larger sizes, more defects and longer time durations. Use of unsupervised learning and graph optimization in handling and analyzing large scale MD data will be highlighted.

1. Introduction

Materials in nuclear reactors are subjected to neutron fluxes. The neutrons pass through the material losing energy by elastic and inelastic collisions with the nucleus of the atoms constituting the material [1]. This creates energetic primary knock-on atoms (PKAs) which cause local collision cascades in the material. This results in atoms getting displaced from their crystal positions, creating defects (interstitials and vacancies). The timescale of these collision cascades are of the order of a few pico-seconds [2]. Depending on the energy of the PKA, the extent of the cascades can be from less than a nanometer to several hundred nanometers. The created defects can then diffuse throughout the material. As they diffuse they can recombine and reduce damage, or form defect clusters which change the material micro-structure and cause degradation in its properties: viz. radiation induced amorphization, radiation induced hardening, thermal and electrical conductivity degradation, radiation induced segregation, high temperature embrittlement and dimensional instabilities like radiation growth, creep and swelling ([3] and references therein).

The changes in micro-structure happen throughout the material for the duration of reactor operation. Therefore Multi-scale simulations are necessary to model the change in the material micro-structure [4, 5, 6]. The sequence of steps in such a study are:
(i) Creation of PKAs by neutrons. The energy spectrum of the PKA is obtained from neutron total scattering cross-sections [1].

(ii) Creation of interstitials and vacancies by the energetic PKA, thier in-cascade clustering and distribution. The creation of interstitials and vacancies can be modelled by Binary collision Approximation (BCA) Monte Carlo methods [7, 8] or by Molecular Dynamics (MD) [9, 10, 11].

(iii) The number of Frenkel pairs, their configuration and spatial distribution from the cascade simulations will be an input to lattice and/or object Kinetic Monte Carlo (KMC) simulations [6, 12] of diffusive–recombination of the interstitials, interstitial clusters and vacancies to form voids and precipitates. Note that the diffusion and precipitation can occur within the grains or along grain boundaries.

The micro-structural and compositional changes due to irradiation obtained by the above sequence of simulations can be inputs to dislocation dynamics [13] and phase field simulations [14] which can be used to obtain the changes in material properties.

Binary collision approximation Monte Carlo codes (BCA-MC) have been widely used for sputtering, range and back-scattering calculations. However better models are needed in modeling the creation of interstitials and vacancies [15]. We have used MD to study the directional effects of PKA bombardment on the number of displaced atoms in a collision cascade in crystal Cu, W and Fe [15, 16] for modest PKA energies of 1-5 keV. We have carried out several thousand MD simulations with the PKA directed along random crystal directions and shown that typically a hundred random directions in the crystal have to be explored before the average number of displaced atoms become constant. We have obtained the direction averaged (i) number of displacements, (ii) distributions of interstitials and vacancies and (iii) in-cascade cluster size distributions of these defects in [15]. The requirement for large scale post processing of several tera-bytes of data from the MD runs is described in Section.2.

In order to obtain the diffusion co-efficient of the defects, MD simulations for interstitialcy diffusion were carried out [17, 18]. In interstitialcy diffusion the interstitial atom is same as the host material atom and the interstitial displaces a lattice atom and the lattice atom becomes the new interstitial. Therefore the identity of the atom keeps changing. In order to identify the interstitial and create a random walk from which the diffusion can be parametrized, we use unsupervised learning [19] and the graph-tree data structure [17, 20]. This is described in Section.3. We illustrate the multi-scale model by using the average values gleaned from the MD simulations in a kinetic Monte Carlo model for diffusive-recombinations of defects. This is presented in Section.4. Finally we present our conclusions.

2. Large scale data analysis: MD studies of directional effects

We use the Large-scale Atomic Molecular Massively Parallel Simulator (LAMMPS) [21] for our MD simulations of collision cascades. MD outputs the atomic positions as a function of time at specified intervals and this data has to be post-processed in order to identify defects and classify them as belonging to clusters or not. This classification is further complicated by existence of “crowdion configuration” in which several neighbouring interstitials and vacancies crowd around and the number of unpaired defects have to be identified [2]. A typical MD simulation of a collision cascade requires that the simulation box size be large enough to accommodate the cascade. Since the PKA is launched in random direction and secondary cascades can take any random direction, we take the simulation box size to be around 2.5 times the expected range of the PKA. The expected range is found by initial scoping runs. The collision cascade has a time scale of a few pico–secs. Typically, a $50 \times 50 \times 50$ unit cell simulation of bcc W is sufficient to
simulate collision cascades of PKA of energy 5 keV. If the atomic identities and positions of such a cascade simulation of 10 pico–secs are output every 100 MD time steps, it will lead to a 1.25 Giga-byte file. If 200 to 1000 PKA directions have to be explored, this means a 250 Giga-byte to 1 Tera-byte of data for post-processing at one PKA energy.

The defects were identified by an algorithm similar to the method of spheres [2] and is described in [16, 15]. In this method, an interstitial–vacancy pair is said to have been created if the interstitial is away from its lattice position by a value \( r_{\text{disp}} \). We have used an unsupervised clustering algorithm (called Max-space clustering) to identify the value of \( r_{\text{disp}} \) such that the displaced interstitials could be separated from the natural vibrational amplitudes of the atoms [17, 18]. This was done as follows:

- Find the displacements (hitherto called offsets) of all atoms from their original lattice positions.
- Sort the atoms based on their offsets.
- Find the difference between offsets between consecutive atoms from the sorted list and therefore the pair of atoms (say \( i \) and \( i+1 \)) having maximum difference in their offsets.
- Set \( r_{\text{disp}} = \text{average value of the offsets of atoms } i \text{ and } i+1. \)

It is shown that this method of choosing offsets is better than existing methods [18].

Using the \( r_{\text{disp}} \) obtained from this method, in-situ identification of the displaced atoms was carried out by adding two new “computes” in LAMMPS, one for outputting the number of displacements, the maximum range, the range of the PKA, etc and the other to dump the positions of the identified displaced atoms for post-processing the in-cascade clustering of defects. This results in only around 500 MB of data output for post-processing 1000 random directions of a PKA at a specified PKA energy compared to the earlier 1TB data output. It also did not lead to extra computational time because the extra time for in-situ data processing was offset by the input/output (I/O) time required to write the several Giga–bytes of data files to disk. Typical direction averaged distributions of interstitials and vacancies for PKA energies of 1-5 keV in Cu is shown in Fig.1 and Fig.2 respectively. The averaging was done for 200 MD simulations of randomly directed PKA at each energy. It was seen that 200 random directions is sufficient for the variances in the number of defects produced to stabilize [15]. This can now be a realistic input for initializing defect positions in a KMC code (Section.4).

3. Interstitialcy diffusion from MD

The defects obtained from the above described collision cascades thermalize with the material atoms at the end of the cascade and diffuse. The main channel of diffusion of the interstitial atoms is by interstitialcy diffusion, in which, the interstitial displaces a lattice atom and occupies its position. The displaced lattice atom is the new interstitial. In an MD simulation, in order to identify the random walk of the interstitial, the changing identity of the interstitial atom also has to be tracked. The interstitial can be identified from each time frame output by the Max-space clustering algorithm described in the previous section. However, many a time, the interstitial is part of a cluster of atoms in a crowdion or dumbbell configuration (hitherto referred to as ICA - Interstitial Configuration Atoms). In order to identify the interstitial from the ICA we use the graph-tree data structure [20]. All ICA in a time-step are represented by nodes in a level (fig.3). Nodes across levels, i.e. ICA at the previous and next time-step, are connected by an edge with weight \( w \) equal to the displacement between the positions. All the nodes at level \( l \) (parent-nodes) are connected to all the nodes at level \( l + 1 \). We link every node to a parent node which has least weighing edge to it. Specifically, for every node \( nl \) of the level \( l \), we calculate the cost function \( c \ (nl) = \min( w \ (ni \ (l+1), \ nl) ) \) for \( i \ [1,m] \), where \( m \) is the
Figure 1. Distribution of the interstitials as a function of distance from the origin of the PKA for energies 1-5 keV in Cu

Figure 2. Distribution of the vacancies as a function of distance from the origin of the PKA for energies 1-5 keV in Cu

number of parent nodes, and mark a link to the parent node which yields minimum cost. Details of the graph-tree algorithm and various cost functions explored are presented elsewhere [17].

We have carried out MD simulations to determine (i) $r_{\text{disp}}$, (ii) the migration energy $E_m$ and (iii) the diffusion coefficient for interstitialcy diffusion [18, 17]. A $10 \times 10 \times 10$ unit cell simulation box of various fcc and bcc metallic crystals of interest with a randomly placed single interstitial of the same specie was first equilibrated using a NPT MD simulation at various temperatures below the melting point of the crystal. NVE simulations were then carried out for nano–seconds with a time step of a femto–sec. Atomic positions were output every 100 MD time steps. Post-processing of the data to identify the ICA and the diffusion trajectory was carried out. Fig.4 shows a typical trajectory of Cu interstitial. The diffusion coefficient of the interstitial at each temperature was then obtained from the diffusion trajectory. Assuming an Arrhenius relation for the diffusion with temperature, the migration energy was determined. Fig.5 shows the variation of the interstitialcy diffusion coefficient of Cu with temperature. From the plot the $E_m$ for interstitialcy diffusion of Cu is calculated to be 0.08 eV which compares well with published results of 0.12 eV [22]. The pre–factor for the diffusion is $7.9 \times 10^{-8}$. It was also seen that almost all the jumps in the random walk are 1st nearest neighbor (1NN) jumps. These diffusion parameters can and characteristics can also be used as inputs to a KMC code (Section 4).

4. Scaling up with Kinetic Monte Carlo

We have developed a simulation code for Object KMC (OKMC) simulations based on the residence time algorithm [23]. The code was validated by reproducing the interstitialcy diffusion obtained from the MD simulations in Section 3. We simulate cascade evolution of a uniform PKA energy spectrum in the energy range 1-5 keV, uniformly distributed over a $100 \times 100 \times 100$ unit cell W crystal lattice. The interstitials and vacancies are distributed from the distributions obtained.
from the MD cascades described in Section.2, but for W from [15]. The migration energies and the jump sizes were specified from similar MD simulations as described in Section.3, but for W [17]. New defects were introduced in the simulation volume at the rate of 0.1/micro-sec for 10 micro-secs. Clusters of defects of size lesser than four are allowed to diffuse uni-directionally since rotation of a cluster is very rare [24]. The larger clusters are not allowed to diffuse in these simulations. The simulation was carried out till there were no more particles that could diffuse. The evolution of the clustering of interstitials and vacancies at 10 micro-secs, 28 milli-secs and at the end of the simulation (5.6 hours) are shown in Fig.6, Fig.7 and Fig.8 respectively. The results demonstrate that basic diffusion-agglomeration can be carried out by the KMC code. Work is going on to further validate it with published results.

5. Conclusions
We have described MD simulations to obtain (i) directional averaged values of the number of defects, their distribution around the PKA origin and their in–cascade clustering during a collision cascade in Section.2 and (ii) diffusion parameters and characteristics of single interstitials in Section.3. The data gleaned from the MD simulations are passed as inputs to a KMC simulation which simulates the diffusive-recombination of the defects to form clusters as described in Section.4. Initial results of the clustering for a randomly distributed, uniform spectrum of 1-5 XXVII IUPAP Conference on Computational Physics (CCP2015) IOP Publishing Journal of Physics: Conference Series 759 (2016) 012078 doi:10.1088/1742-6596/759/1/012078
keV PKA is presented thereby making the link from atomistic calculations to defect clustering at higher spatial and time scales, thereby demonstrating a multi-scale model for radiation damage.

A Max-space algorithm was used to identify the defects and a graph-tree data structure was used to obtain the interstitial from a group of interstitial cluster atoms so that the random walk of the interstitial can be obtained. One of the themes of this conference was on Computational Physics Education. The methods for data analysis used in this paper are not normally part of a Computational Physics curriculum. We therefore feel the need to emphasize that perhaps data structures and Artificial Intelligence (AI) algorithms need to be taught in advanced Computational Physics courses because they give a physicist structured ways to handle and analyze large data.

Acknowledgments
We would like to acknowledge the support lent to us by N. Shaktivel, H. Hemani and others maintaining the Computer Clusters at BARC-Visakhapatnam. The large scale MD simulations would not have been possible without them. One of the authors (S. Bukkuru) acknowledges a BRNS grant (No:2013/36/32-BRNS) which helped him carry out this work.

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