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HPC atomic simulations of defect formation and kinetic Monte Carlo simulation of material ageing

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Abstract. Kinetic Monte Carlo (KMC) simulations are developed to simulate microstructure evolution under irradiation of structural materials of nuclear power plants. Methods based on rigid lattice atomic KMC and object KMC, despite some approximations, present the advantage to reach significant time, hence irradiation doses. The physical inputs such as the primary damage and defect cluster properties are the results of intensive atomic simulation on high performance computers. Here, atomic KMC is applied to model microstructure evolution of reactor pressure vessel steels and object KMC is applied to zirconium cladding materials.

1 Introduction

Under neutron irradiation, the interaction of neutrons with the atoms of the crystal lead to the formation of point defects (vacancy and self-interstitial atoms) and more extended defects (small dislocation loops or small cavities) within the picosecond time scale. The diffusion of these defects leads to some recombination (e.g. annihilation between vacancy and self-interstitial atom (SIA)) and to defect cluster growth (e.g. when defects of the same type aggregate) to form large dislocation loops for example. Under a flux of neutrons, the kinetics of the evolution of the irradiated microstructure has consequences on the evolution of the mechanical properties (e.g. hardening, embrittlement).

The shift of the brittle to ductile transition temperature in RPV steels is obtained at the mesoscopic level from the evolution of their nanostructure, ascribed mainly to matrix damage, solute enriched clusters, and grain boundary segregation. Therefore, the study of the formation, evolution, and thermal stability of these features is of great importance for the identification of the mechanisms to be included in physical based macroscopic scale models.

For some materials and in some specific conditions, the deformation is isotropic (irradiation swelling of cubic alloys) and for others, like zirconium alloys, the growth is anisotropic. Zirconium alloys are widely used as fuel cladding tubes in the nuclear industry

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due to their low absorption cross-section for thermal neutrons and good corrosion properties. However, during in-reactor operations, these components exhibit dimensional instabilities due to neutron irradiation and complex mechanical loading. Understanding and predicting the strain behaviour of these components is an important issue.

The challenge of modelling the evolution of a microstructure under irradiation is due to the different time and length scales involved. The primary damage is formed 10 picoseconds after the interaction of a neutron with the material that generates a displacement cascade which is modelled by molecular dynamic simulations at the atomic scale [1][2]. The medium to long term evolution is controlled by the diffusion of the defects formed (isolated and in clusters) which can be simulated by kinetic Monte Carlo (KMC). KMC allows to consider spatial correlations between defects and thus takes naturally into account the morphology of the displacement cascades. KMC simulation are time consuming because of the large gap between atomic migration process and time operation of the components. In order to accelerate these simulations, approximations are done, such as the use of a rigid lattice for atomic KMC (AKMC), or the development of coarse grain KMC, such as object KMC (OKMC). The inputs of these coarse grain approaches can be obtained from direct molecular dynamics for fast processes, or from accelerated molecular dynamics or off lattice KMC [3] results.

2 HPC atomic simulations of defect formation

Atomic simulations of the formation of defects following the interaction of a material with neutrons and obtaining their properties (i.e. their thermodynamical and diffusion properties) take a large advantage of HPC resources and massive parallelization.

2.1 Primary damage

First, the spectrum of the primary knocked on atom (PKA) is determined from the neutron spectrum, which allow to take into account the local neutron spectrum in the components. Secondly, the primary damage for a large range of PKA energies has been simulated in different materials (Fe for RPV, Zr for zirconium, as well as FeNiCr as model for austenitic stainless steel) by molecular dynamics based on empirical potentials (according to the Embedded Atom Method – EAM – formalism). A large number of displacement cascades has been modelled in order to have enough statistics to characterize the cluster size distribution as well as the spatial correlations of the defects within the cascades. Typically, a sampling of 1000 cascades is required to obtain significant statistics [4].

Figure 1 represents an 80 keV PKA energy cascade in Fe, with the formation of <111> SIA and <100> vacancy dislocation loops. Large statistic allows to characterise the defect size distribution, which is an important input for the KMC microstructure simulations.

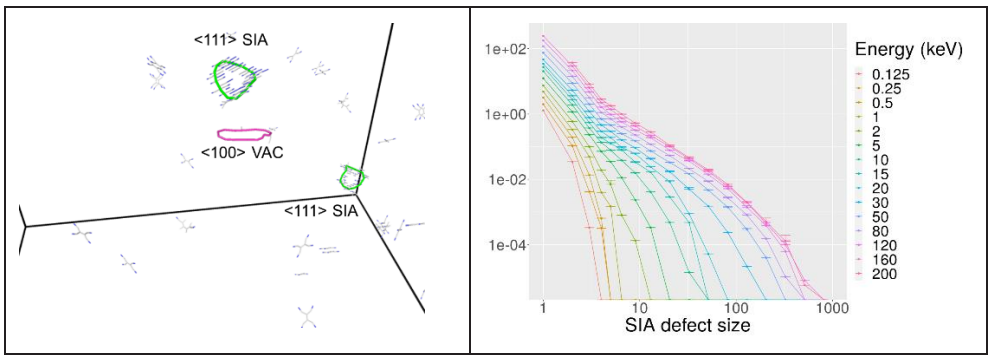

Fig. 1. Left: cascade debris of one 80 keV PKA cascade in Fe (SIA are represented by "bonds" between short distance atoms). Right: SIA cluster size distribution averaged on series of several hundred cascades of different PKA energies.

2.2 Defect clusters properties

2.2.1 Defects in Fe matrix

The interstitial defect clusters in Fe have complex structures (<110> dumbbells are the most stable mono SIA configuration in Fe due to magnetism, parallel or non-parallel organisation of many SIAs can form, for instance the C15 structure SIA clusters (Fig 2. a) up to a transition size towards interstitial loop formation (Fig 2. b)) [5] at the nanometre size. Density Functional Theory (DFT) calculations investigating the respective stability of SIA clusters of different shapes, using HPC resources, have allowed important progress in the interpretation of the experimental data. To model dilute FeCuNiMnSi alloys by AKMC, a cohesive model based on concentration dependent pair interactions on rigid lattice has been developed and parameterized based on DFT calculations [6]. While the new cohesive model is numerically heavier than simple pair potential models, it allows to capture the energetics of complex MnNiSi clusters in Fe matrix (Fig 2. c).

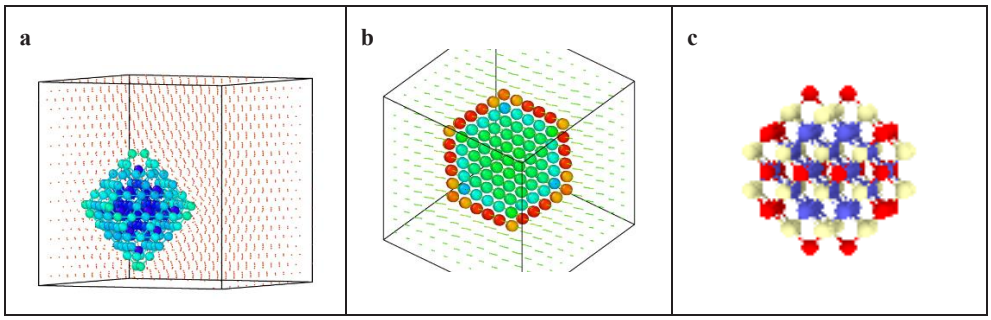

Fig. 2. Complex defects in Fe. a) C15 cluster (large atoms in blue). b) interstitial <111> loop (large atoms). c) MnNiSi cluster (Ni: yellow, Mn: blue, Si: red).

2.2.2 Defects in zirconium matrix

In hcp zirconium, different loops are observed experimentally and visible when they are larger than few nanometers. Atomic simulations based on DFT and empirical potentials for

small defects have been performed in order to characterize their stability and mobility. These simulations have revealed new structures:

Platelet interstitial clusters in the basal plane [7] (Fig 3. a).

Pyramidal vacancy defect clusters, which are seen as precursors of vacancy large c loops [8] (Fig 3. b).

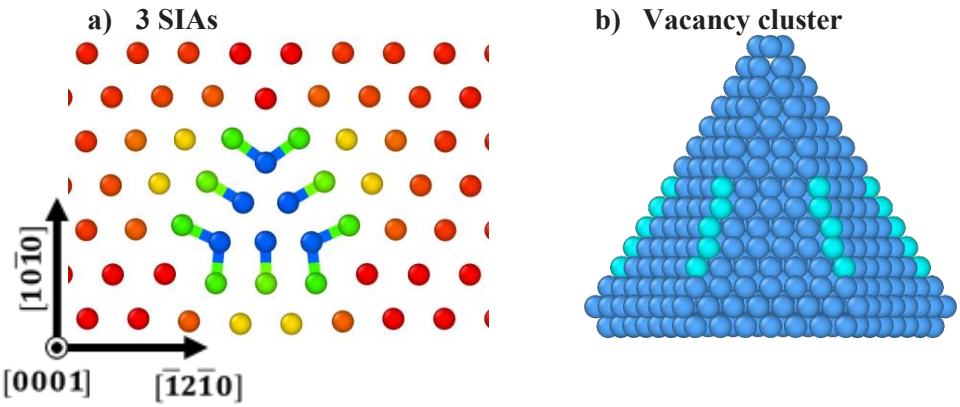

Fig. 3. a) planar 3 SIA cluster in the basal plane. b) pyramidal vacancy clusters

3 Kinetic Monte Carlo simulations of microstructure evolution

The simulations in this work have been performed with the Monte Carlo code LAKIMOCA developed at EDF [9] which mutualizes the AKMC and OKMC development presented here. These KMC simulations are quite long and our code in sequential, as parallelisation remains a challenge for general microstructure evolution simulation under irradiation, despite some parallelisation adapted to specific conditions [10][11][12]. In complementary to the study presented, when KMC is used to determine parameter input such as sink strength [13][14] or to optimized parameterization of large objects to fit some specific experimental results [15], a large number of independent KMC simulation can be run in parallel on HPC resources.

3.1 Model alloys of reactor pressure vessel steels

The complex alloy considered in this case is FeCuNiMnSiP, and to model its evolution under irradiation, an hybrid model combining both AKMC and OKMC has been developed [6]. Small defect clusters are treated explicitly in details by AKMC and large ones by OKMC.
AKMC simulations of isolated defect cluster complexes (point defect clusters with some solutes) allow to parameterize the object KMC model. This method provides a better understanding of the macro events (e.g. mobility of the cluster including solute drag, or emission of a point defect) involved in the simulations. The hybrid approach developed brings an acceleration of two orders of magnitude allowing to reach doses corresponding to 40 years of irradiation in service condition. The difficulty of the model to reproduce flux effect has been solved by adding an absorber that reduced the grain boundary sink strength. Traps have also been introduced to simulate the presence of impurities.

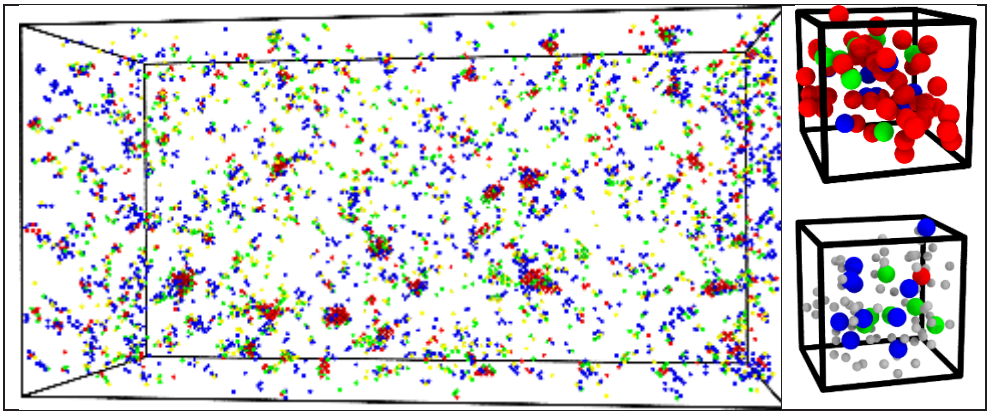

Fig. 4. Left: microstructure at 0.09 dpa for Fe-0.05Cu-1.38Mn-0.69Ni-0.47Si (at %) alloy at 600 K, simulation box 60×60×120 a_0, flux~10^{-5} dpa/s. Right: solute complex clusters formed (red: Cu, green: Si, blue: Ni, Grey: Mn)

3.2 Zirconium cladding materials

Based on atomic simulation results, a model including interstitial <a> loops and vacancy: pyramidal, <a> and <c> loops has been developed and optimized to predict irradiation growth by OKMC simulation [16][17][18]. The microstructure obtained (Fig. 4) is not homogenous, with the formation of alignment of prismatic interstitial loops in agreement with the corduroy patterns observed experimental by transmission electron microscope [19].

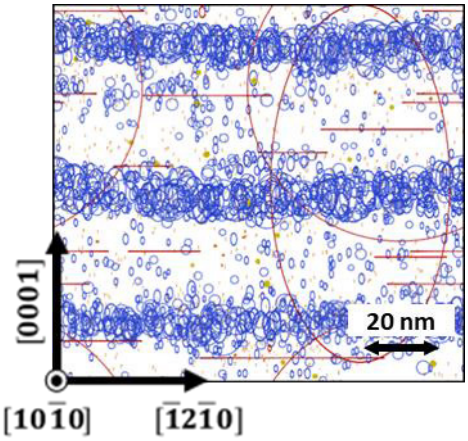

Fig. 4. One simulation irradiated zirconium microstructure (blue: SIA <a> loops, red: <a> and <c> vacancy loops calculated by OKMC.

4 Conclusion

Kinetic Monte Carlo simulations are one of the important methods capable of modelling microstructure evolution under irradiation. Atomic simulation on HPC is of paramount importance to get the physical mechanisms, inputs for the microstructure modelling.
For RPV, we have built an AKMC model as well as an hybrid AKMC-OKMC model of dilute Fe alloys representative of the PWR steels and simulated its behaviour under irradiation. The results obtained are in agreement with the experimental trends and indicate that the formation of solute clusters takes place via segregation mechanisms on the point defect clusters.
For zirconium modelling, we combined parametrized OKMC modelling using state-of-the-art atomic-scale calculations and experimental data. It leads us to conclude that the acceleration of macroscopic growth is the result of the formation of <c> vacancy loops and layers of <a> interstitial loops parallel to the basal planes that are directly connected to the vacancy diffusion anisotropy. The parametrization used allows the coexistence of <a> and <c> vacancy loops that were not taken into account in previous modelling efforts.
The complexity of the defect clusters associated to different type of solutes benefit also from advanced Monte Carlo techniques [3], as well as the use of machine learning techniques. The results of these advanced simulations on some limited irradiation conditions could be used to propose metamodels to predict more rapidly the microstructure in different irradiation conditions.

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