Multiscale Modeling of Defect Cluster Evolution in Irradiated Structural Materials

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Irradiation effects impact structural materials performance in nuclear environments. Irradiation effects initiate from displacement cascades, and are ultimately determined by the diffusional transport and fate of point defects and their clusters, along with solute and impurity re-distribution. This presentation will introduce the inherently multiscale nature of irradiation effects in materials and describe a multiscale research paradigm based on close integration of materials modeling and experimental characterization. Results will be presented from in-situ ion irradiation studies of thin film materials in the transmission electron microscope of molybdenum and ferritic/martensitic steels. In this effort, spatially dependent cluster dynamics models, which are informed by lower length scale understanding of cascade dynamics and defect cluster behavior, are shown to be in good agreement with the fine details of the dose, dose rate and sample dimension dependencies of the defect density and size distribution observed experimentally for low dose irradiation.

Prediction of material behavior after irradiation for reactor pressure vessel steels and reactor internals : modeling aspects and implementation in the PERFORM platform

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In the frame of FP7 PERFORM60 European project [1,2], a main objective is to establish multi-scale models to predict the behavior of reactor pressure vessel steels and reactor internals. Once the models are proposed and validated, they are implemented in a numerical platform aiming at providing simple tools to both researchers and industrial users to conduct a wide range of calculations crossing length-scales from ab-initio to molecular dynamics, dynamics of dislocations, crystal plasticity and fracture mechanics. These different calculations will be briefly introduced, as well as the possible chaining between these calculations. The original methods introduced in the platform to take advantage of time consuming small scale simulations like molecular dynamics used to obtain atomic displacement cascades will be introduced, as well as specific and efficient methods like Object Monte-Carlo methods to predict longer term microstructure evolution once the cascade database is available. Larger scale simulations will be also discussed, and more specifically the introduction of irradiation in crystal plasticity laws and the calculation of the material fracture toughness and its change with irradiation.
A live demonstration of the platform will allow future potential users to realize the possibilities of use and appreciate the calculation efficiency.

[1] S. Leclercq et al., Journal of Nuclear Materials, **406**, 193-203 (2010).
[2] A. Al Mazouzi et al., Nuclear Engineering and Design, **241**, 3403-3415 (2011)

The necessary initial conditions for simulation of damage cascades under neutron irradiation

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A key requirement for the accurate prediction of the damage accumulation due to neutron irradiation is a full description of the nuclear events, resulting from interactions with neutrons (for example, generated in fusion plasmas or fission fuel rods), which produce the primary knock-on atoms (PKAs) that initiate the cascades. This is a more relevant transfer of information between complex calculations of neutron irradiation fields and the modelling and experiment of materials to investigate radiation damage, than the simple integrated quantities such as 'displacements per atom' (dpa) used normally. The need to use the data from neutron transport calculations more fully has become more urgent due to recent work that identified the large variation in neutron field and integrated dpa values as a function of position within the latest conceptual designs for the DEMO fusion reactor [1,2].

In this work, we have developed a computational methodology to enable the complete distribution of PKA energies for all atomic species (either pre-existing or those created by nuclear reactions) in a material subjected to a given neutron field. This involves taking data from the latest nuclear cross section data files to produce PKA cross section matrices, combining these with an appropriate neutron energy spectrum, and processing the results to produce output that is most applicable for subsequent statistical analysis of damage accumulation (perhaps using the results of extensive cascades simulations).

We present results for several important materials, with an emphasis on fusion materials, and, in particular, consider how the results vary with neutron field in DEMO. We find that the assumed PKA energies often used in displacement cascade simulations are in no-way representative of the complex picture observed in full PKA spectra. For instance, while the average PKA energy of Fe atoms in Fe may be of the order of a few 100s of keV, the full spectrum shows that recoils are produces with energies ranging from a few eV (where cascades would not be produced) right up to 1 MeV. Additionally, there are a significant proportion of recoils associated with transmutation nuclear reactions (such as neutron capture followed by proton or alpha-particle emission – producing, in Fe, Mn and Cr, respectively). While the probability of these reactions is lower than the dominant, simple scattering reactions, their PKA energies are generally higher (Cr atoms can be generated in Fe with recoil energies of more than 2 MeV). Furthermore, the energies of the secondary emitted (light) particles from these inelastic reactions can be much higher, with alpha-particles in Fe generated at 10 MeV or more. Such species are likely to produce markedly different damage compared to the lower-energy "standard" recoils of atoms identical to the host lattice.

[1] Gilbert *et al. J. Nucl. Mater.* **442** (2013) S755
[2] Gilbert *et al. Nucl. Fusion* **52** (2012) 083019

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Elastic trapping of dislocation loops in ion-irradiated tungsten foils

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We have investigated the role played by the elastic interaction between defects produced in radiation damage cascades with a joint experimental and modelling approach. Using in situ transmission electron microscopy, we have observed the nanoscale dislocation loops formed when ultra-high-purity tungsten foil is irradiated with a low fluence of self ions at a range of irradiation temperatures. The loops seen are predominantly of prismatic 1/2<111> type, of vacancy character, and formed close to the surface. Concurrently we have performed object kinetic Monte Carlo simulations of the evolution of self ion damage in tungsten foil. The simulations have been seeded with the best available cascade morphology statistics derived from molecular dynamics; with loops, clusters and vacancies treated entirely equivalently, and with no parameters tuned to improve the fit to experiment.

We find that the number density and size distribution of loops observed is reproduced only if the elastic interaction between loops is taken into account. Our simulations show how important residual defect statistics one nanosecond after a cascade are for understanding experimental observation one second later, and our analysis highlights the profound effect of elastic interaction between defects on the microstructural evolution of irradiated materials.

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Defect Interaction and Evolution in Iron and Its-based Alloys

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The evolution of defects governs the microstructure and properties of materials for various energy applications. Since defect evolution usually involves processes at very different time scale, *e.g.,* the defect production in cascades occurs over picoseconds while defect accumulation in reactors is up to forty years, the span of more than twenty orders of magnitude creates tremendous challenges for both experiments and simulations and there is a significant gap in the accessible time scale between them. To bridge this gap and to computationally model and predict the defect evolution over a long period while maintaining the desired fidelity, an accelerated kinetic Monte Carlo (KMC) approach, self-evolving atomistic KMC (SEAKMC), has been developed for simulating complex defect structures. In contrast to the conventional KMC models that require all reactions to be predetermined, this method incorporates on-the-fly determinations of transition states with a new scheme for defining active volumes in an off-lattice (relaxed) system and any physically relevant motion or reaction may occur. Applications of SEAKMC for simulating defect evolution are demonstrated through three selected problems for BCC iron: (i) <100>-loop formation, (ii) annealing of cascade damage and (iii) long-term evolution vacancy clusters and their growth. The formation mechanism of <100> loop in BCC iron is revealed using SEAKMC. For cascade annealing, a comparative study using SEAKMC and object KMC is performed. For the multi-vacancy case, the results are compared with those obtained using the autonomous basin climbing method, kinetic activation-relaxation technique, and molecular dynamics (MD). It is found that SEAKMC possesses the atomistic fidelity that is similar to the MD but on a much longer time scale. The differences between SEAKMC and other methods are discussed. In addition, the unique predictive capabilities and the limitations of SEAKMC as well as its potential applications to a wide range of problems are elaborated.

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Diffuse interface modeling of void growth in irradiated materials. Mathematical, thermodynamic and atomistic perspectives

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We present an assessment of the diffuse interface models of void growth in irradiated materials. Since the void surface is inherently sharp, diffuse interface models for void growth must be constructed in a way to make them consistent with the sharp-interface description of the problem. Therefore, we first present the sharp-interface description of the void growth problem and deduce the equation of motion for the void surface. We also compare two existing phase field models to determine which one corresponds to the sharp-interface analysis. It was shown that a phase field model of type C, which couples Cahn-Hilliard and Allen-Cahn equations, is the most adequate since this type of model can take into account the reaction of point defects at the void surface via an Allen-Cahn equation. Fixing the model parameters in the diffuse interface model is discussed from the points of view of asymptotic matching. Sample results for void growth in a single component metal based on sharp and diffuse interface models are presented. Finally, a perspective on the use of atomistic modeling in both constitutive and nucleation modeling within the phase field approach for void formation in irradiated materials is presented.

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Constrained ab initio method for non-collinear magnetic excitations

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Magnetic excitations determine the finite temperature phase stability of the most important class of structural nuclear materials, such as iron alloys and steels. They influence production, migration and evolution of defects in such materials. *Ab initio* calculations are a powerful tool for the investigation of defects on the atomic scale. However, it is intrinsically incapable of treating excited states, including spin excitations. In order to consider the effect of non-collinear magnetic excitations in ferromagnetic iron, we develop a constrained method involving the use of Lagrange multipliers. Some of the methods developed earlier involve various sophisticated procedures for calculating vector Lagrange multipliers iteratively, and all of them do not guarantee numerical convergence.

We propose a new constrained method for the *ab initio* treatment of non-collinear magnetism. The method involves using a fixed scalar Lagrange multiplier λ and does not require re-calculating Lagrange parameters on the fly. We prove analytically that the method guarantees convergence if a sufficiently large value of λ is used. The penalty energy or the penalty potential are inversely proportional to λ. The method is simple and straightforward, and can be readily implemented in a DFT code, including VASP. Both the atomic and magnetic configuration can be relaxed using our method.

In this work, we describe applications of the new method. Firstly, we study a 2 atom configuration in BCC Fe using constrained orientations of magnetic moments. The energy change as a function of orientations compares well with literature data. Variation of energy for the 2 atoms case with magnetic moments at 90 degrees confirms the predicted relation between the penalty energy and the choice of λ. Secondly, we study a 54 atom amorphous system with magnetic moments pointing in random directions, showing that the method applies also to large magnetically disordered configurations. Lastly, we investigate the case of a <110> dumbbell SIA defect that shows how magnetic excitations induce extra forces acting on atoms. We find that the local volume in the core of a dumbbell increases when the magnetic configuration changes from antiferromagnetic to ferromagnetic.

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Ab initio study of threshold displacement energies in tungsten

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The most fundamental parameter in phenomenological radiation damage theories, such as the Kinchin-Pease- or the NRT models predict a damage dose that depends on the average threshold displacement threshold of the material. We here present an *ab initio* molecular dynamics study of the the angular dependent threshold displacement threshold surface in the refractory metal tungsten. W is the strongest candidate material for the plasma divertor in nuclear fusion tokamaks. We present a detailed study of the anisotropy of the energy surface and an average value that can be used as a more reliable input in damage dose assessments. We compare our results to predictions made using semi-empirical interaction potentials, as well as available experiments for tungsten. We also compare to other bcc metals such as iron, where a similar study has been recently performed [1]. In order to improve the short range part of stiffened interatomic potentials, we also predict the energy loss in replacement collision sequences and discuss the implications of the here performed quantum mechanical simulations.

[1] P. Olsson, C. Domain, Submitted to Phys. Rev. Lett (2014).