論文の内容の要旨

New Multi-Scale Tools for Modeling the Structural Evolution of the Reactor Pressure Vessel under Irradiation

(原子炉圧力容器鋼における照射下微細組織発達モデリングに関する
マルチスケールシミュレーションツールの開発)

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1 Introduction

1.1 Background

The reactor pressure vessel (RPV) of nuclear fission reactors is closely monitored for material changes through microscopic and macroscopic testing and observation, and predictions are supplemented with results from a range of computational tools. The focus of safety monitoring is the ability of the RPV's ability to withstand fracture at all normal operating temperatures, a property measured by the Charpy test. This test measures the energy absorbed during the fracture of identical specimens at a range of temperatures. Metals such as Alpha-Iron from which an RPV is constructed show two distinct regimes of fracture; The first is the ductile regime, where material stress is partially relieved by structural changes, and requiring the most energy to fracture. The second is brittle fracture in which the material cannot easily relieve stress and requires less energy to fail. The temperature at which the material makes the distinct transition from ductile to brittle fracture (the DBTT) is important to safety management and is itself strongly dependent on the irradiation history of the material. Thus safety management seeks to ensure that RPVs are never exposed to radiation such that the DBTT shifts to within normal operating temperature margins. Fig.1 shows the effect of radiation, annealing and re-irradiation on an RPV steel, demonstrating that whilst annealing (IARA) can largely heal the deleterious effects of radiation (IAR), a permanent shift nevertheless occurs (IARA - Unirradiated).

The distinctive DBTT shift shown in Fig.1 can be interpreted in a simplified manner as resulting from two processes. Firstly, radiation causes a high number density of precipitates to form, and then annealing forces coarsening of precipitates, lowering their number density. The effect on the Charpy energy is due to the precipitates acting as obstacles to the movement of dislocations across the matrix, preventing the material from relieving stress [11, 7]. Thus a high number density of precipitates results in hardening, which leads generally to a more brittle fracture. The microscopic phenomena relating to the formation, and later coarsening, of precipitates are dealt with in this study, although alternative non-hardening embrittlement exists, in which the possibility of fracture

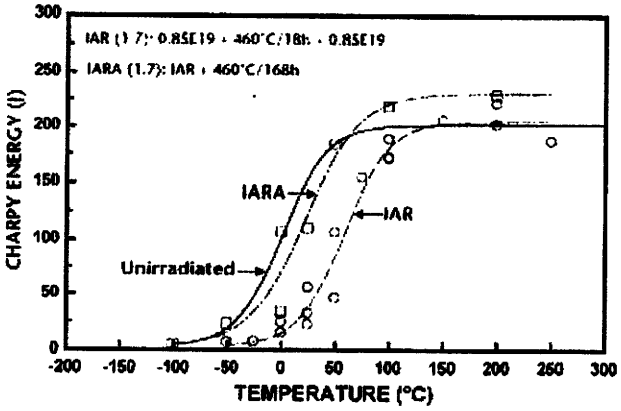


Figure 1: Energy absorbed in fracture of irradiated and reirradiated (IAR) and reirradiated and annealed (IARA) materials [4]

at grain boundaries is increased by radiation-related phenomena such as void growth and impurity segregation [6].

1.2 Motivation and purpose

This work focuses on multi-scale modeling, the general method employed by the safety management community for predicting macroscopic material behaviour from microscopic phenomena [15].

Multi-scale modeling relies upon useful and sufficient information being transferred from one stage to the other, (e.g. the results of a microscopic calculation being used as inputs for a larger scale calculation).It has become clear that there discontinuities currently exist in the chain, so small features that may be significant at the upper end of the process are not identified and communicated. This work contributes to multi-scale modeling by developing new computational methods that increase the scope of information accessible to certain stages, specifically concerning the formation and growth of impurity clusters and precipitates.

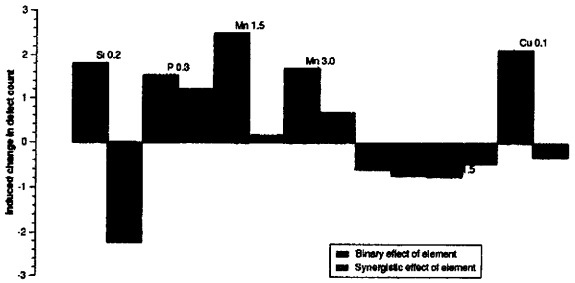


Figure 2: Binary vs. Synergistic effects

2 Cascade Simulation

The beginning of radiation induced precipitate nucleation is the cascade process, in which a fast neutron from a fission reaction interacts with the surrounding wall. The energy of the neutron is passed to a large number of atoms through a growing chain of elastic and inelastic interactions, giving many of them enough energy to leave their binding lattice sites and creating Frenkel pairs. Most of these recombine, but many will diffuse into structures such as clusters of vacancies, interstitial atoms and impurity atoms, , .

Molecular dynamics simulations (MD) of this cascade, using the Yang embedded atom method (EAM) potential allows us to examine the effect of multiple impurity elements in a bcc iron cell during the cascade. The minor part of this is testing 3 PKA energies in pure iron and binary systems, with 10 repeats per composition. The major work is a 10KeV cascade in a volume of varying elemental composition of side 14.3 nanometers (250,000 atoms), lasting 5 Ps and evolved as an NVE ensemble. After cascading, the system is quenched and force-minimised and counts are made of sites without atoms, and atoms without substitutional sites [1]. This gives a good measure of the damage produced by the cascade, and up to 15 repeats are made at every compositional value. Data analysis reveals complex trends in synergistic effects between elements, and one key point of interest is the difference in behaviour between an elements binary effect and its synergistic effect, see Fig.2

3 Parallel Tempering

Following the dissipation of a PKA's energy by cascade interaction, in addition to the generation of many Frenkel pair, a thermal spike occurs. In the fission reactor environment, this is of magnitude of thousands of degrees, and lasts the order of picoseconds [14]. Simultaneously modeling atomistic behaviour under the dissipation of the thermal spike remains difficult, since physical properties are uncertain and temperature gradients of 10^4K/nm are present. Current Kinetic Monte-Carlo methods of analysing clustering during this period consist of counting defect number and type apx. 5 Ps after a cascade, and then modeling migration with a pre-calculated event table that relies on first principles calculations of binding and migration energy [16]. These current methods cannot explore phenomena outside the description of the event table, which is restricted to known and likely reactions. We present a model which allows greater range of atomistic interactions, including those that are only likely to occur at very high-temperature. This will provide greater insight into the structure and composition of clusters shortly after the dissipation of the thermal spike. In the simulation presented here, no prior calculation is necessary, meaning that all local environment variables are factored in to a diffusion event. Parallel Tempering (PT) works by simulating N exact replicas of a system at N different temperatures, where probability of swapping between neighbours increases as their potential energies become similar, whilst preserving the detailed balance criterion, guaranteeing reversibility [13, 2]:

$$A = min\{1, exp[(\frac{1}{K_B T_i} - \frac{1}{K_B T_{i+1}})(U(r_i^N) - U(r_{i+1}^N))]\}$$

Whilst PT could be used in multiple MD environments, it is particularly useful for cascade simulations since the higher temperature replicas can be seen as representing the possible events in the high temperature volume of the cascade. Achieving a reliable heat distribution is not possible in conventional simulations, so it is an advantage that PT can select stable configurations from a system undergoing high-temperature processes. Early results of cascade simulation followed by parallel tempering indicates some aggregation of impurities during this previously unexplored period, Tab.1.

Table 1: PT effect on cascade damage of a simulation cell composition (%at.); Si 0.2, P 0.3, Mn 0.0, Ni 0.8, Cu 0. Data reflects the percentage increase of number of 1NN and 2NN atoms containing an impurity solute following PT relative to the number of 1NN and 2NN atoms containing an impurity solute before PT.

	Si	***P***	***Mn***	***Fe***	***Ni***	***Cu***
Si	.0667	.0087	.0000	.9247	.0000	.0000
P	.0111	.0718	.0000	.9085	.0026	.0060
Mn	.0000	.0000	.0000	.0000	.0000	.0000
Fe	.0031	.0031	.0000	.9844	.0065	.0030
Ni	.0000	.0012	.0000	.5474	.0505	.0010
Cu	.0000	.0019	.0000	.9075	.0133	.0773

4 Optimisation of pair potentials

KLMC calculations require the calculation of the local configuration energy, typically employing empirical potential functions to do so. The requirements of a KLMC are quite basic, needing only a static single-time energy calculation, which can be reasonably handled by a pair potentials derived from chemical considerations [8]. A genetic algorithm optimisation code forwards sets of parameters to a KLMC simulation, where cluster counting program returns a value of goodness to each set of pa-

Table 2: Optimised pair potential parameters

T (K)	Fe-Fe	Fe-Cu	Cu-Cu	V-Fe	V-Cu
540	-0.609	-0.555	-0.601	-0.250	-0.240
560	-0.732	-0.601	-0.582	-0.234	-0.166
580	-0.686	-0.578	-0.614	-0.209	-0.179
600	-0.724	-0.600	-0.650	-0.295	-0.245
620	-0.714	-0.595	-0.619	-0.215	-0.175
640	-0.649	-0.576	-0.694	-0.207	-0.234
660	-0.766	-0.611	-0.635	-0.240	-0.162
680	-0.663	-0.550	-0.557	-0.218	-0.168

rameters. The goodness of the parameters is improved by observing biological rules of reproduction and mutation.

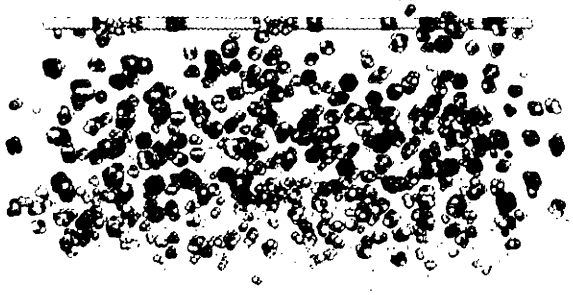
This method succeeded in optimising the pair potentials for a binary alloy against an EAM potential standard. In the future, this could be exchanged for an experimental standard since the detection efficiency of Atom Probe Tomography [4] has increased steadily to the point where the chemical composition of clusters is clarified. The method established here would enable the experimental atom map to be used as the standard to which the pair potentials could be optimised, thus linking one stage of the multi-scale modeling process to experiment.

5 Distorted lattice monte-carlo

Exposure to radiation will have an embrittling effect on RPV materials, but this can be mostly recovered by an annealing process, as seen in Figure 1. The annealing process allows for coarsening of precipitates, increasing their average size but decreasing their average number density. During this process, the main agent for transport of large impurity elements is vacancy diffusion. Since the jump of a vacancy is a rare yet energetically well understood event, the movement of a vacancy across a crystalline matrix can be simulated with a Kinetic Lattice Mont–Carlo program. [5, 17] As the vacancy moves it drags impurity elements through the matrix which can, depending on the type of element, lead to clustering and later precipitation.

Recent observations with one of the most accurate techniques for determining chemical compositions, atom probe tomography, reveals clustering to favour lattice defects for nucleation. This is of special importance when the defect is a dislocation, since other studies show that the stress required for a dislocation to pass through the precipitate (CRSS) depends upon the relative location. To clarify this situation, a Kinetic distorted-Lattice Monte-Carlo (DLMC) program is introduced, in which the atomic positions are non-homogeneous, and represent atomic positions near to a defect. The vacancy, and in turn the elements it drags, are sensitive to the differing atomic radii causes by the distorted lattice. In all real-world crystalline materials, there are many dislocations in every grain, thus the possibility that dislocations act as nucleation sites must be covered. Furthermore, contemporary MD simulations regard precipitates as homogenously formed across the lattice, and of regular shape and distribution (see e.g. [9, 12]). The DLMC simulations conducted here provide strong evidence that this is a misleading way to begin a simulation, since the impurity atoms tend to cluster in the highly stressed regions inside the dislocation core, where they can have the most significant effect on hardening, see Fig.3.

Figurenn 3: Compressive stress cylinder effect on copper clustering. The dislocation core is at the top of the figure, demarked by a broken line of gray atoms in the z direction. The simulation volume has been quartered in x and y directions and all iron atoms that are not part of the dislocation core have been erased.



6 Conclusion

This work represents contributions to the multi-scale modeling community in the study of fission reactor pressure vessels.

The effect of impurity elements such as found in Japanese RPVs on cascade damage processes is modeled, being the first such simulation to include a range of 5 impurity elements described by an Embedded Atom Model potential formulation.

Parallel Tempering is demonstrated for the first time as a potential addition to the range of multi-scale modeling techniques. Describing the behaviour of the RPV matrix at times and spaces very close to the cascade, without the need for an event list, enables more realistic evolution of the system. Results show the tendency of certain impurity elements to aggregate together very shortly after the cascade, although the method of diffusion is unclear.

Linking computational Monte-Carlo techniques and molecular dynamics methods is shown to enable more realistic evolution, in this case by forcing diffusion of individual atoms (MC), and allowing the system to respond (MD). This allows the simulation to respond to two distinct time and length scales, giving new results about the nucleation and growth of clusters. A technique of optimising pair potentials for use in a Kinetic Lattice Monte -Carlo expands the utility of such simulations, and paves the way for a new link between multi-scale modeling and experiment.

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