Modelling high-energy radiation damage in nuclear power and fusion applications

Kostya Trachenko, Eva Zarkadoula
Queen Mary University of London

Martin Dove, Cambridge and Queen Mary

Ilian Todorov, Daresbury Laboratory
Radiation damage effects in ceramics

- Motivation: pressing need to safely encapsulate radioactive nuclear waste that comes from power plants and surplus plutonium

- HLW accounts for 95% of the total radioactivity produced during nuclear electricity generation. The amount of HLW worldwide is increasing by 12,000 metric tons a year, equivalent to 100 double-decker buses

- Pu stockpile in the UK: ~100 metric tons (reprocessed from waste and from decommissioned weapons), of which 20 tons has been declared as waste (Am)

- Ceramics are proposed to be used for encapsulation of high-level waste
Effect of radiation-induced amorphization on diffusion

Actinides are long-lived. A waste form should be stable during millions of years. Traditional glasses are not an option. UK NDA and NNL want to use ceramics.

Case study: zircon ZrSiO$_4$ found minerals are $\sim$1 billion years old, completely amorphous yet intact

Absorbs large ions like Pu on Zr site
Molecular dynamics simulation of radiation damage

Details:

1. Empirical potentials and short-range ZBL potential at short $\sim 1$ Å distances

2. Almost perfectly scalable MD code based on domain decomposition strategy (DL_POLY 3, 4)

3. Parallel computers
   Cambridge HPC, HPCx, HECToR
   (time through Materials Chemistry Consortium)

4. Adapted MD code to handle out-of-equilibrium conditions (variable time step, boundary scaling, inhomogeneous density) and to analyze radiation damage on the fly
Channels of low density appearing along the track

This gives channels of increased diffusion and explains percolation-type increases of transport at $p=0.3$
Resistant vs amorphizable materials

Gd$_2$Ti$_2$O$_7$ – “official” US Department of Energy waste form. Amorphizes easily under irradiation

Gd$_2$Zr$_2$O$_7$ – does not amorphize even at very large radiation doses!

What is the nature of the process of resistance to amorphization by radiation damage?
Modelling resistance to amorphization by radiation damage

- 100 keV in rutile TiO$_2$, ~5-10 mln atoms, MD box size is ~500 Å
- 512-1024 HPCx parallel processors
Look at the process in detail
Rutile TiO$_2$
Two types of relaxation:


2. Relaxation and recovery of the true structural damage

Both happen on the few picosecond timescale.
Resistance to amorphization

50 keV U recoil

SiO$_2$  GeO$_2$  TiO$_2$  MgO  Al$_2$O$_3$

Poor recovery  Intermediate recovery  Good and perfect recovery

$t=1$ ps

$t=5, 50$ ps


MD simulations reproduce experimental behaviour of resistance to amorphization (activation barriers correlate with the curvature of the potential at equilibrium)
Resistance to amorphization

- Time scales of damage recovery: several ps

- Correlate the details of interatomic potentials with damage recovery: damage increases with the stiffness of O-O interaction
Resistance to amorphization

MD simulations reproduce experimental behaviour of resistance to amorphization.

MD simulations can be used to predict highly resistant materials (e.g. ZrO$_2$, Gd$_2$Zr$_2$O$_7$) where resistance to amorphization operates on the time scale of picoseconds.
Current work and future plans: fusion!

Simulate radiation damage of energies relevant to fusion reactors: high radiation fluxes (200 dpa and 500 keV-1 Mev Fe recoil atoms from 14 MeV neutrons)

- No experiment possible other than fusion reactor. MD simulations are therefore important.

- These energies were not studied before, yet are important to simulate

- Need **up to 1 billion atoms** in a MD box. Doable with 16,382 HECToR processors. The size of this system is about **0.2 micrometers**! (2\textsuperscript{nd} in the world after Lawrence Livermore people)

- Study the effects of temperature, pressure, important mechanical properties, deformation, elasticity etc

- Possibly exciting new effects, as new energy and length scales (µm) are approached
Current and future work on fusion

Simulate radiation damage of energies relevant to fusion reactors:
500 keV – 1 Mev Fe recoil atoms from 14 MeV neutrons

- One configuration of 250 mln atoms is about 100 Gb, 1000-frame history file is 100 Tb!
  We will be facing and solving new interesting challenges:
  writing speed and storage (parallel writing, analyze on the fly what we can)

- Common to all future MD simulations of very large sizes. The appetite for large systems approaching µm is growing: shock, fracture, initiation of micro-cracks, micro-structural changes and interfacial effects, macromolecules, biological systems of wide ranges
Current and future work on fusion

Simulate radiation damage of energies relevant to fusion reactors:
500 keV – 1 Mev Fe recoil atoms from 14 MeV neutrons

- Include electronic energy loss in the simulation - in collaboration with Ilian Todorov (Daresbury) and Dorothy Duffy (UCL).
- Exciting new effects, as new energy and length scales (µm) are approached

Ongoing work, supported by EPSRC grants:

1. “Development of high-performance software”
2. Impact QM: collaboration with the UK National Nuclear Laboratory
3. “Pathways to Impact”: collaboration with the Culham Centre for Fusion Energy
First attempt:

~ 100 million -1 billion atoms

~2048-8192 parallel HECToR processors

250 keV Fe recoil in iron

System size ~ 1000 Angstrom
Thank you