Atomistic Simulation of Nuclear Materials

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• **Current Fission Reactors:**
  – Nuclear Fuel
    • Performance and ageing phenomena prediction.
    • $\text{UO}_2$ / MOX / Thoria(?)
    • Radiation damage
  – High Level Waste Encapsulation and Immobilization
    • Predicting solution of interstitials and radioactive dopant ions.
    • Predicting stability of ions and migration energies to surfaces/grain boundaries and radiation damage of host matrix

• **Future Fusion Reactors:**
  – New materials
    • Able to withstand higher
      – temperature and neutron fluxes
Research Interests

**Fuel Performance**
- Behaviour under irradiation
- Safe, controlled and predictable ‘heat source’
- Calculate intrinsic/extrinsic defect energies to predict transport of species to grain boundaries and fuel-clad gap.
- Modelling of fission ‘gas bubbles’

**Nuclear Waste**
- ‘Safe and Secure’ immobilization of high level radioactive waste in a form suitable for final disposal
- Simulations predict properties of candidate ceramic compositions as host matrices, tolerance to radiation damage and radionuclide transport characteristics

**Sensors**
- Predicting formation energies of defects causing optical response
- Fundamental understanding of neutron radiation induced lattice effects
- Relate to mode of operation of ‘sensor’
Application and Relevance

Overview

Modelling *augments* experimental characterization *NOT* replace it!

Can be thought of as an *additional* analytical technique.

![Image of Bruker D8 NMR instrument](image1)

![Diagram of Mott Littleton](image2)

![Image of 900 MHz, 21.2 T NMR equipment](image3)
Modelling scales and regimes

Time Scale

ms
µs
ns
ps

Size Domain

Å
nm
µm
mm

Quantum Mechanical
Molecular Mechanics
Atomistic Simulation
Molecular Dynamics
Finite Element Analysis

Challenges to multiscale integration
Nuclear Fuel

Ceramic Fuels

Advanced Gas Reactor (AGR)
Moderator: Graphite

Pressurized Water Reactor (PWR)
Moderator: light water – $\text{H}_2\text{O}$

Fuel: $\text{UO}_2$ with 2.5\% $^{235}\text{U}$ enrichment
Coolant: $\text{CO}_2$ (very weak moderator)

A second-generation UK-designed nuclear reactor employing vertical fuel channel design
Simulation of Uranium Dioxide

Simulation of the bulk lattice

Need to describe interatomic forces
Simulation Methodology

Potential Models

Atomistic approach to modelling crystal structure and properties involves:

Interatomic potential functions simulating the forces acting between ions.

Interatomic pair potentials can be written as:

\[ U(r) = \sum_{ij} \left( -\frac{q_i q_j}{\varepsilon r_{ij}} + A_{ij} \exp \left(-\frac{r_{ij}}{\rho_{ij}}\right) - \frac{C_{ij}}{r_{ij}^6} \right) \]

**Coulombic Term**

**Short range interactions** attributed to the repulsion between electron charge clouds, van der Waals attraction

Classical Born model framework of ionic interactions:

\[ A, \rho \text{ and } C \] are variable parameters Empirically fitted to experiment

Shell Model (polarization)
Empirical Fitting of U – O Potential

Employing GULP to Fit Parameters Simultaneously

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<th>Original</th>
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cell
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90.0
fractional
U core 0.00 0.00 0.00 -2.54 0 0 0
O core 0.25 0.25 0.25 2.40 0 0 0
U shel 0.00 0.00 0.00 6.54 0 0 0
O shel 0.25 0.25 0.25 -4.40 0 0 0
space
225
Parametric Study – U - O

C set to 0
A and r varied.

Solutions for $\delta = 0$
identified by red diamonds.
Programme Drivers and Strategy

Robust Simulation of Actinide Oxide

Atomistic Regime

Robust description of Bulk Lattice

Surfaces Nanoparticles

Surface defects

Extended defects

Higher Order Surfaces

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<tr>
<th>Index 1</th>
<th>Index 2</th>
<th>Index 3</th>
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# Lowest energy shift per Miller Index

- Index h k l Surface_Energy
Quartz, $\text{SiO}_2$ doped with $\text{Fe}^{3+}$ ions from $\text{Fe}_2\text{O}_3$.

The value of the quartz is drastically increased by the presence of a relative small number of $\text{Fe}^{3+}$ ions!
Willis Cluster

Cluster Energy = -23.21 eV
Binding Energy = -0.17 eV
Investigating the Defect Chemistry

Cluster Defects

Schottky Clusters

Second Oxygen Vacancy at position:

<table>
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<tr>
<th>Position</th>
<th>Cluster Energy eV / Defect</th>
<th>Binding Energy eV / Defect</th>
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<tr>
<td>1</td>
<td>1.54</td>
<td>-1.08</td>
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<td>3</td>
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Binding Energy $= \left\{ V_U''' : 2V_O^\bullet \right\}^x - V_U''' + 2V_O^\bullet$
Simulating MOX fuel

MOX 25% ($U_{0.75}Pu_{0.25}O_2$)

‘Mean Field Approach’

Ionic radius = 0.97 Å

Pu$^{4+}$

Supercell

Ionic radius = 0.93 Å

U$^{4+}$
Intrinsic disorder

Cerium brannerite

Schottky Vacancy

Unstable Pu nucleus within MOX fuel decays principally by $\alpha$ decay

- 86 keV Recoil nucleus
- U range 12 nm
- $\alpha$ range 10 $\mu$m
- 5 MeV $\alpha$ particle
- Cascade size 0.8 nm
- 256 Frenkel Pairs

Cascade size 7.5 nm
- 2550 Frenkel Pairs
The theoretical peak performance of the compute nodes is calculated as follows:

\[ \text{Theoretical peak performance} = 848 \text{ (cores)} \times 2.2 \text{ (GHz)} \times 8 \text{ (floating point operations/cycle)} = 15 \text{ TFlop/s} \]

The HPC service was completely replaced in December 2012 and is currently based on an IBM iDataplex HPC cluster with 800 Sandy Bridge based compute cores and other facilities such as large memory servers and a GPU-assisted compute node.
A centre of excellence for computational science, engineering and mathematics

Easy access to e-Infrastructure, research and collaboration for business

MidPlus is a collaborative partnership between four of the UK’s leading universities:

- University of Warwick
- University of Birmingham
- University of Nottingham
- Queen Mary, University of London.

Partnership brings together academic expertise and leading-edge facilities for computing capability and capacity, with an aim to facilitate the rapid realisation of modern computational research methods for business and industry.

This regional HPC centre complements the BEAR facilities and adds additional resources for demands that cannot be met locally.
Acknowledgements

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Prof. Richard Catlow FRS

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