UNIVERSITY OF BIRMINGHAM

BEAR Launch 2013

24th June 2013

Atomistic Simulation of Nuclear Materials

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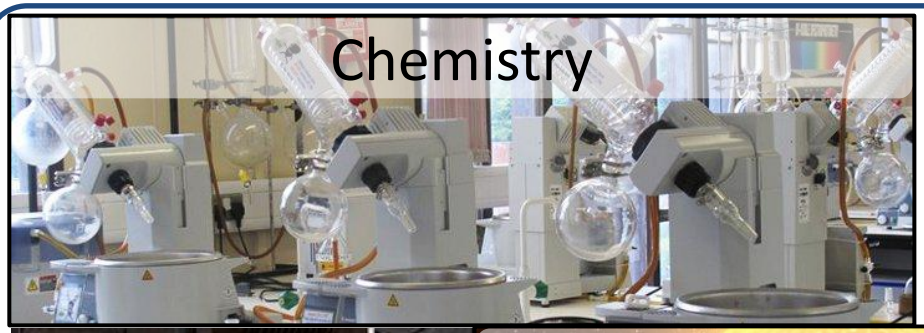
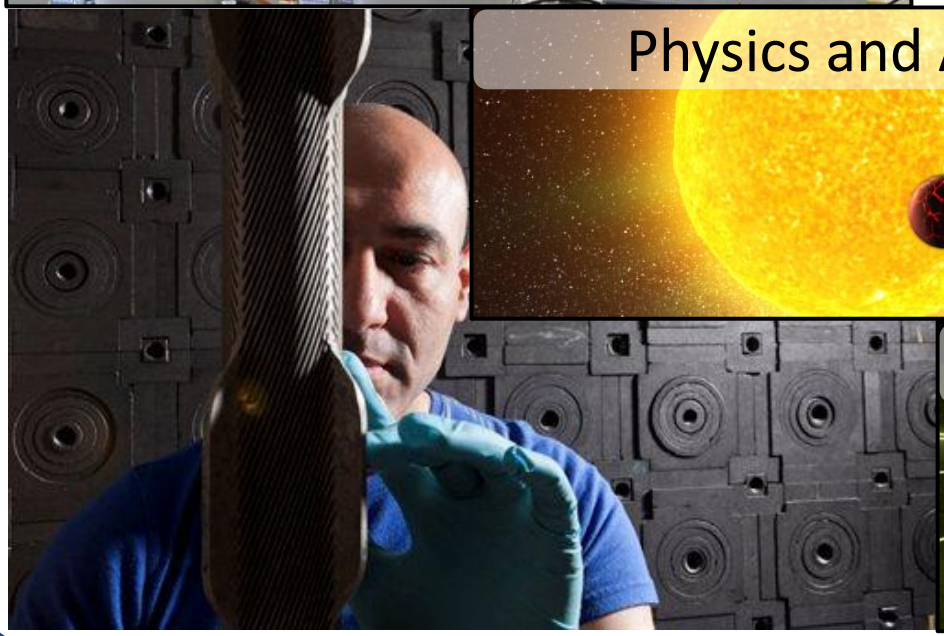
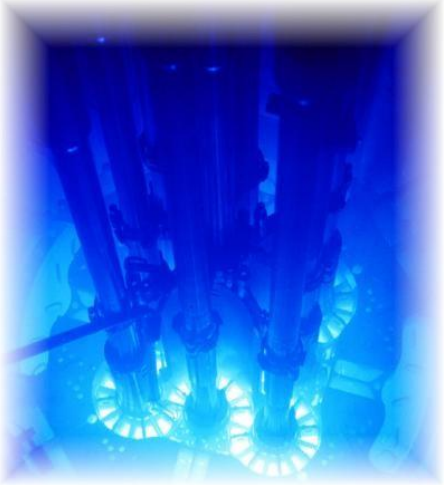


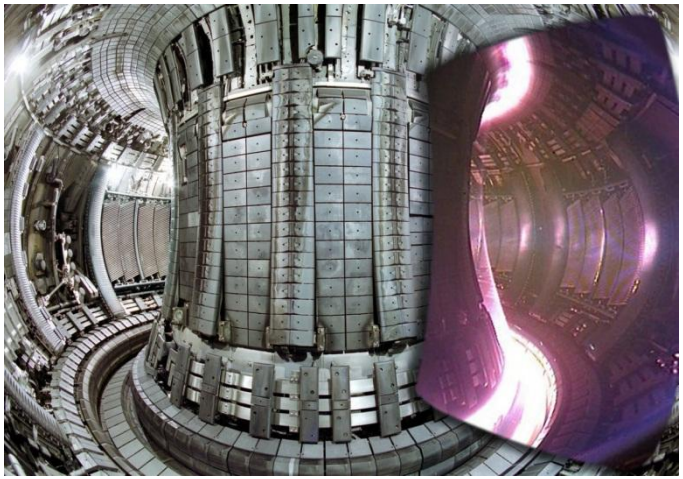


Modelling: Importance to Nuclear Industry

Atomistic Simulation: Materials Challenges



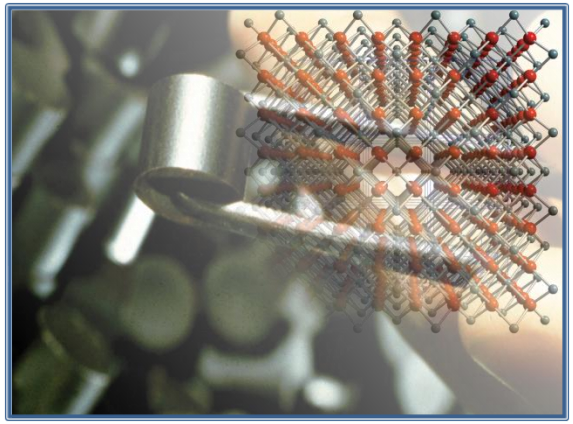
- **Current Fission Reactors:**
  - Nuclear Fuel
    - Performance and ageing phenomena prediction.
    - UO_2 / MOX / Thoria(?)
    - Radiation damage
  - High Level Waste Encapsulation and Immobilization
    - Predicting solution of interstitials and radioactive dopant ions.
    - Predicting stability 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



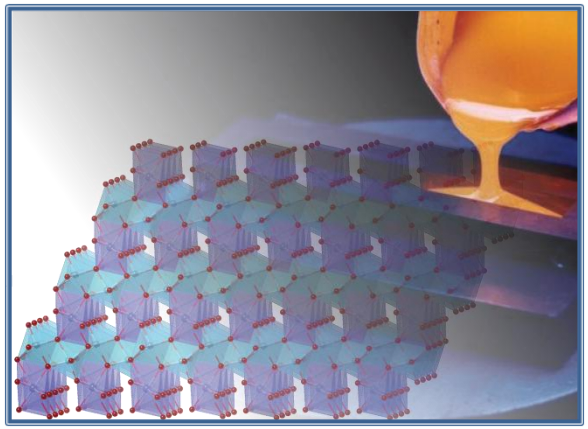
Behaviour under irradiation

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'

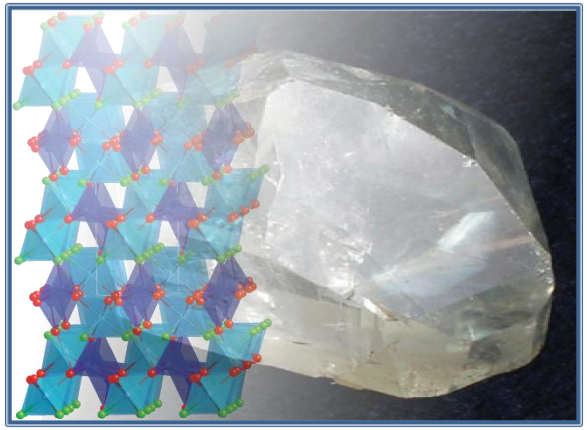
Nuclear Waste



'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

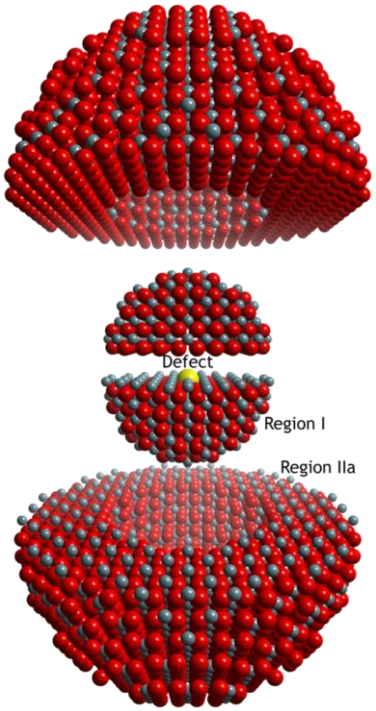
Relate to mode of operation of 'sensor'

Application and Relevance

Overview

Modelling augments experimental characterization NOT replace it!

Can be thought of as an additional analytical technique.

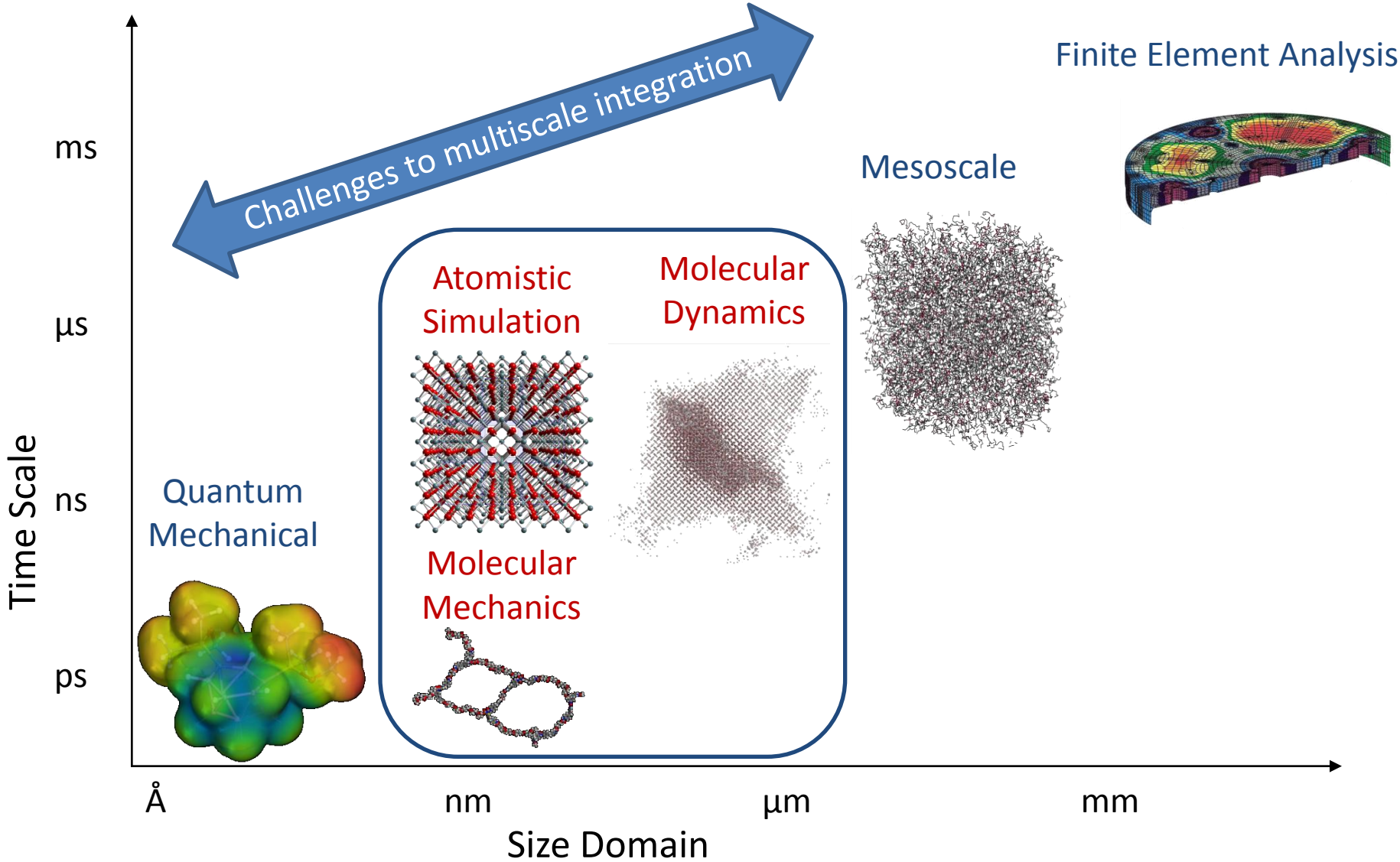


BRUKER D8

Mott Littleton !

900 MHz, 21.2 T NMR

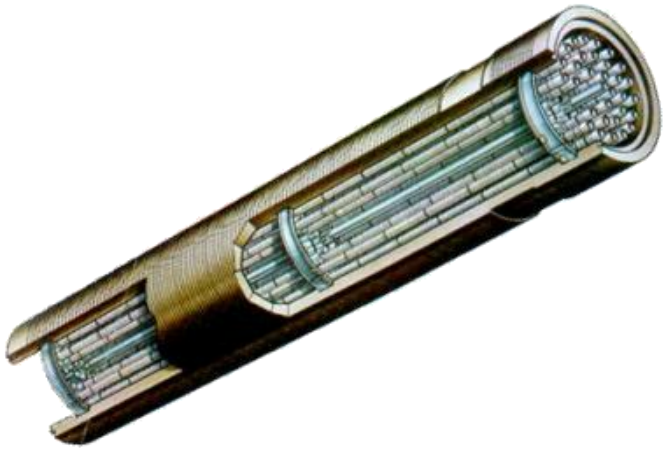
Modelling scales and regimes



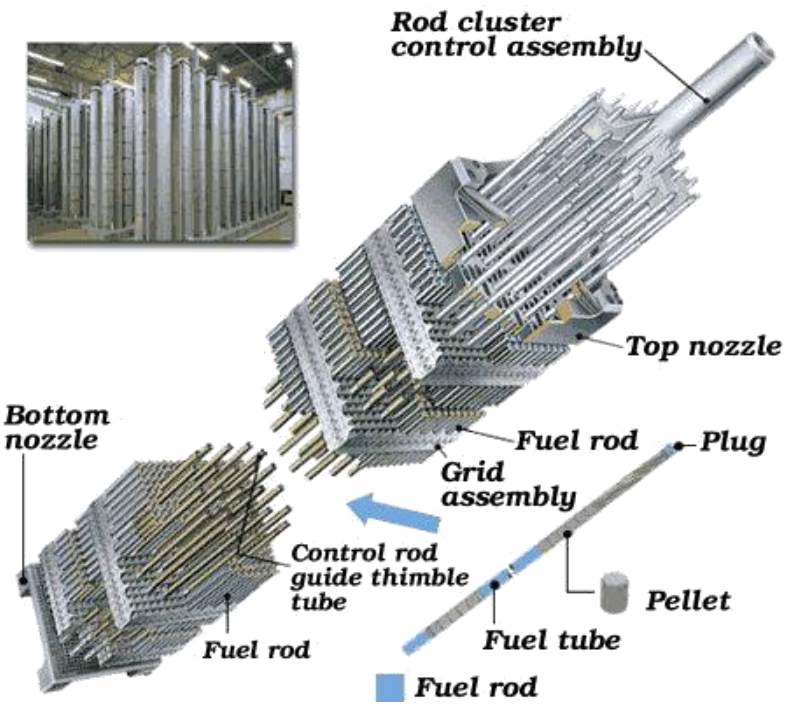
Nuclear Fuel

Ceramic Fuels

Advanced Gas Reactor (AGR)
Moderator: Graphite



Pressurized Water Reactor (PWR)
Moderator: light water – H_2O

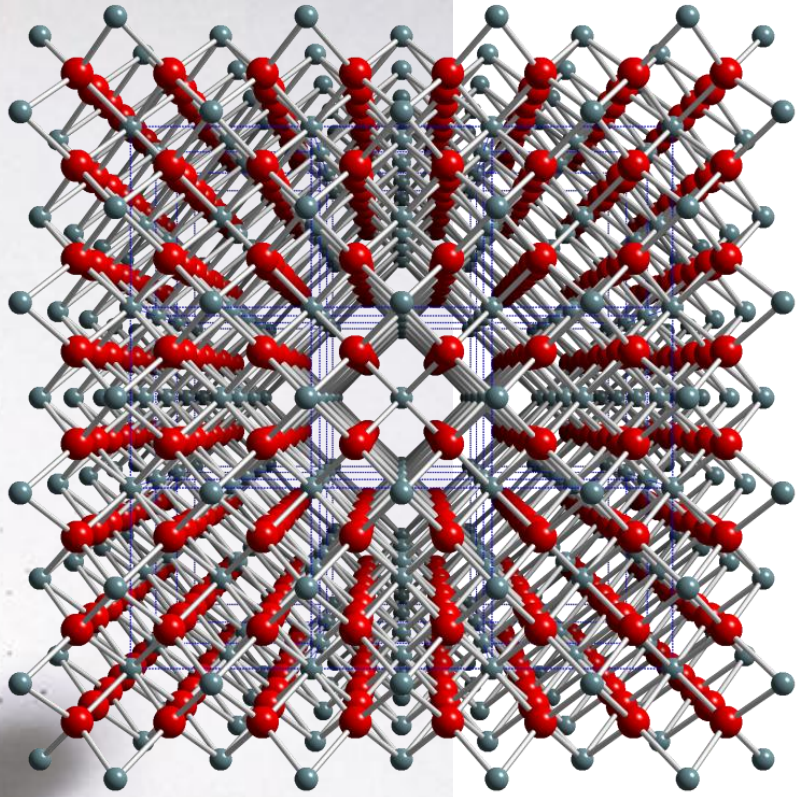


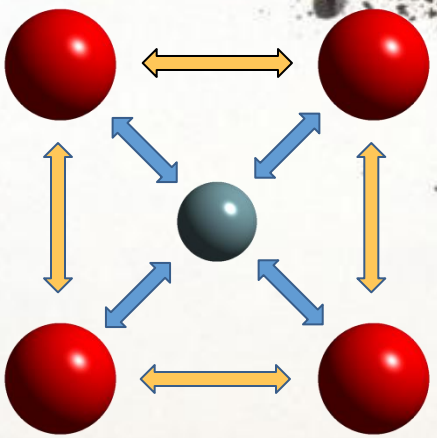
Fuel: UO_2 with 2.5% ^{235}U enrichment
Coolant: CO_2 (very weak moderator)

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:

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

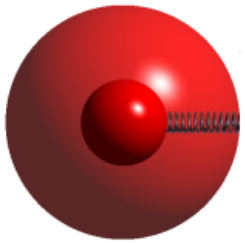
Coulombic Term

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

Classical Born model framework of ionic interactions:

A, ρ and C are variable parameters Empirically fitted to experiment

Shell Model (polarization)



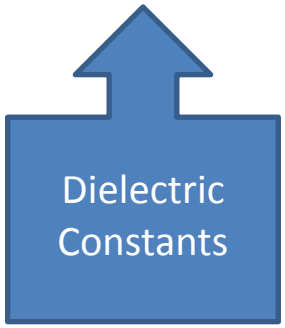
Empirical Fitting of U – O Potential

Employing GULP to Fit Parameters Simultaneously

Parameter Type	Parameter Original
Buckingham A	1518.920000
Buckingham rho	0.382080
Spring k 2	94.240000

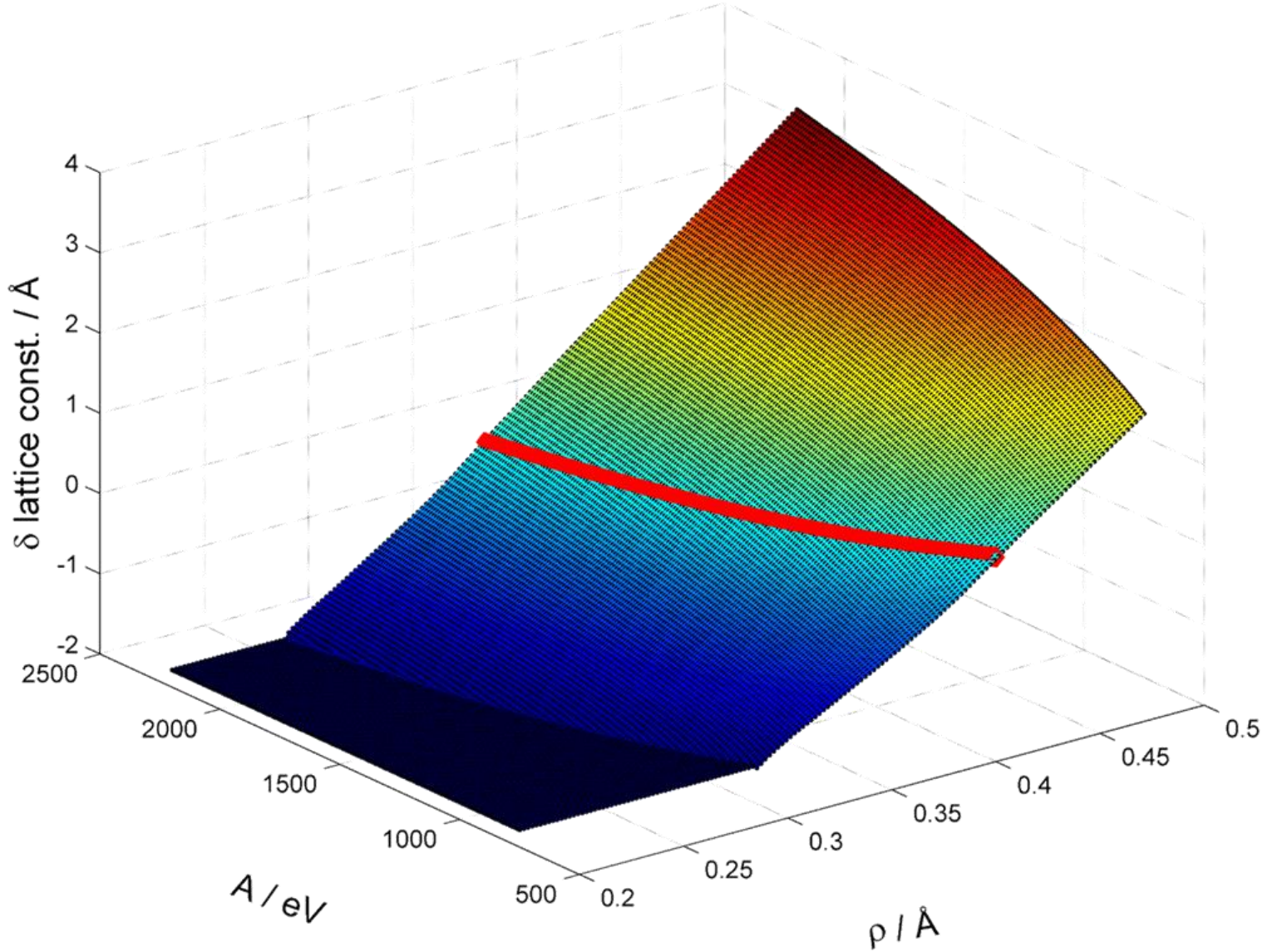
Parameter Type	Parameter Final
Buckingham A	1507.182133
Buckingham rho	0.373145
Spring k 2	65.627688


```
cell
   5.4682   5.4682   5.4682  90.0  90.0
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
```



Type	Observable	Weight
Elastic Const	389.300000	0.0100
Elastic Const	118.700000	0.0100
Elastic Const	59.700000	0.0100
High Freq DiC	5.300000	1.0000
Static Di C	24.000000	1.0000

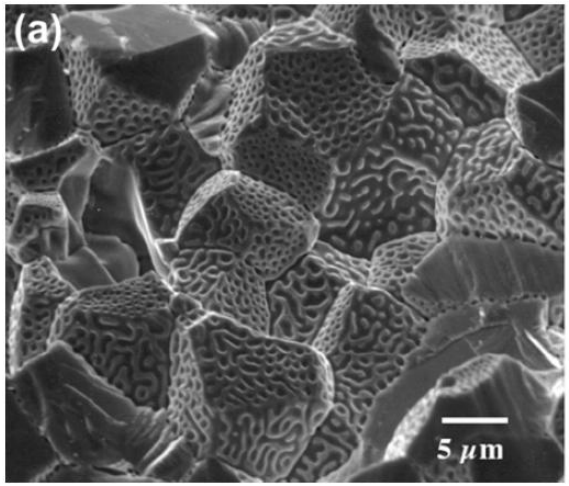
Parametric Study – U - O



C set to 0
A and r varied.

Solutions for
$\delta = 0$
identified by red diamonds.

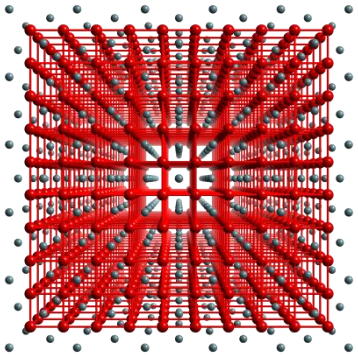
Programme Drivers and Strategy



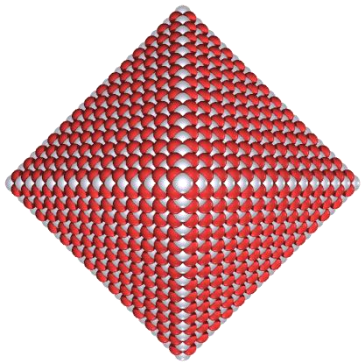
Robust Simulation of Actinide Oxide

Atomistic Regime

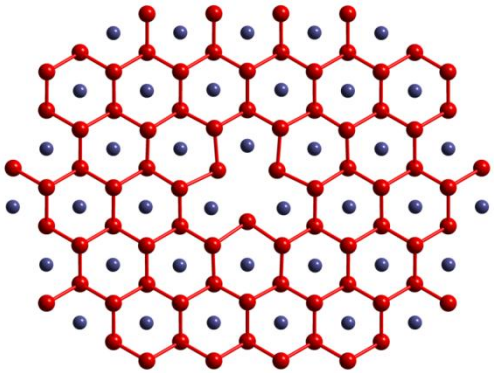
Robust description of Bulk Lattice



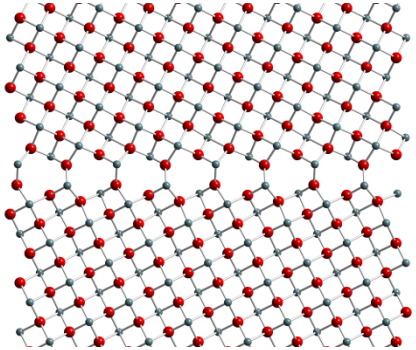
Surfaces Nanoparticles



Surface defects



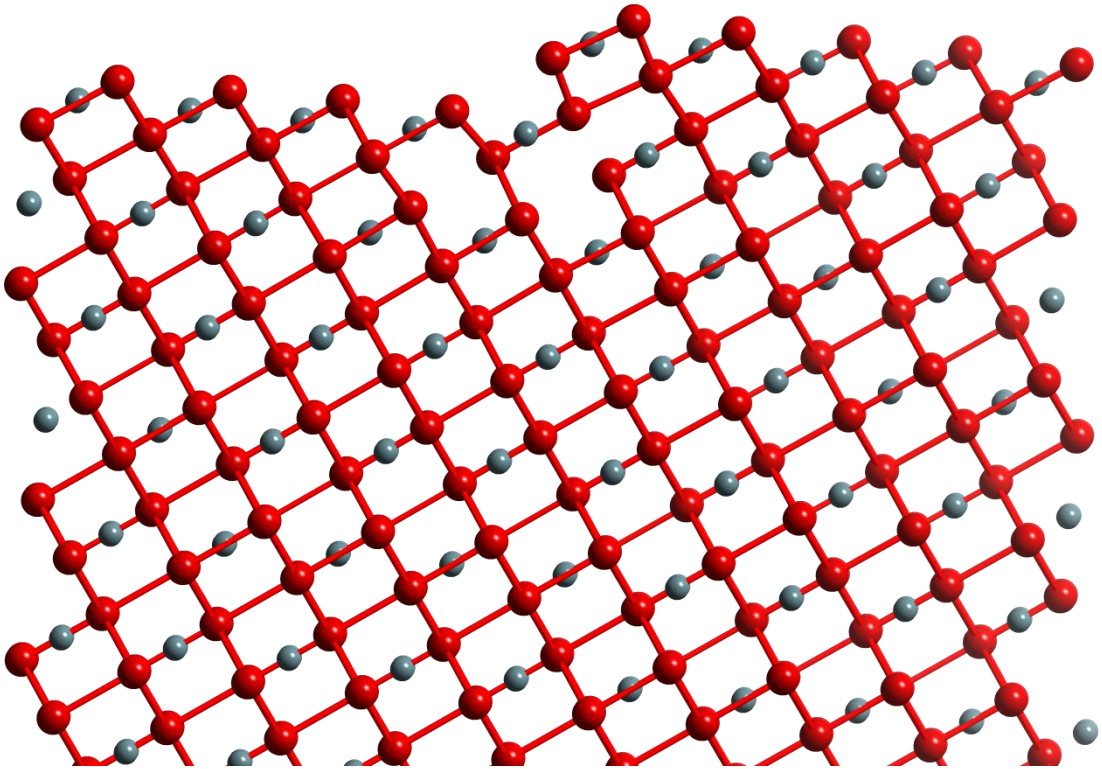
Extended defects



[a] P. Millet, Computational Materials Science 53 (2012) 31–36

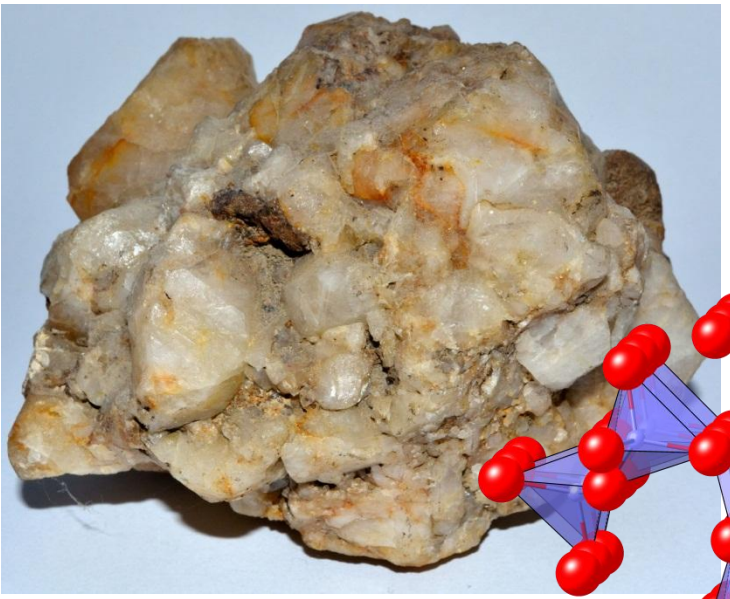
Higher Order Surfaces

<4-55>

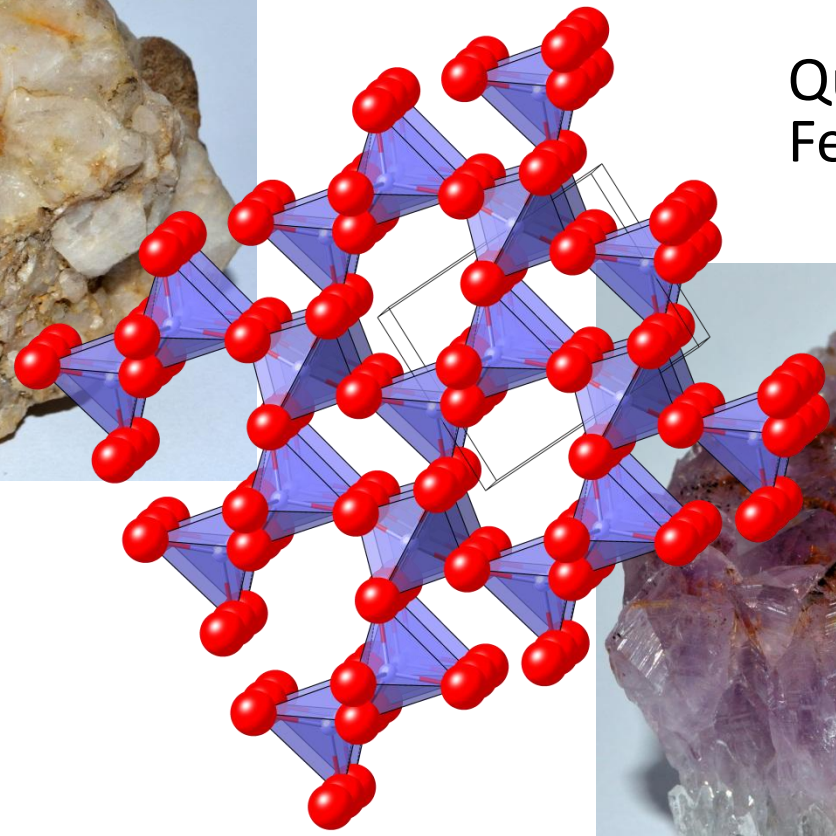


```
# Lowest energy shift per Miller Index
# index h k l Surface_Energy

Index   1  1  1  1.11561
Index   4 -5  5  1.19276
Index   2 -3  3  1.24479
Index   3 -5  5  1.27550
Index   6  5 -6  1.34829
Index   2 -5 -5  1.37917
Index   3  4  5  1.38527
Index  -6  6  5  1.38794
Index   6 -6  5  1.40633
Index  -6  5  6  1.40881
Index   1  3  3  1.41022
Index   3  5 -5  1.47694
Index   4 -4  3  1.49566
Index   1  5  5  1.50669
Index   3 -4  4  1.51483
Index   5  3 -5  1.60875
Index   1  1  0  1.62201
Index   1  4 -5  1.63955
Index   1 -4 -5  1.63956
Index   1  5  6  1.64315
Index   1 -2 -3  1.64449
Index   4  5 -5  1.65310
Index   1 -2  1  1.65846
Index   0  1 -1  1.67691
Index   2  3  5  1.69731
Index   1  6  6  1.70165
Index   1  4  6  1.70968
Index   1 -4 -4  1.71434
Index   5 -6  6  1.71905
Index   1  3  5  1.72907
Index   1  2  2  1.75407
Index   1  2  4  1.80652
Index   1  1  2  1.82508
Index   1 -4  0  1.86597
Index   1  2  5  1.87443
Index   1  5 -2  1.87493
Index   1  0  4  1.90954
Index   2  3  3  1.91579
Index   1  4  0  1.92097
Index   1  1  3  1.92818
Index   1 -3 -6  1.92842
Index   1  1 -5  1.97931
Index   0  1  4  1.99825
Index   1  1 -3  2.00320
Index   1  5  1  2.00526
Index   1  0 -6  2.03147
Index   1  1  6  2.03747
Index   1  5 -1  2.05252
Index   1  0 -4  2.05506
Index   1  2  6  2.07281
Index   0  1 -4  2.07560
Index   0 -6  1  2.07736
Index   0  0  1  2.09124
Index   4  0  1  2.09575
Index   0 -4  1  2.11127
Index  -6  1  1  2.11315
Index   1 -1 -4  2.16457
Index   1 -5  0  2.17623
Index   1 -1 -5  2.21188
Index   1  1  5  2.21805
Index   1  0 -5  2.22785
Index   0 -5  1  2.29404
Index  -5  0  1  2.31452
Index   1 -6  0  2.33259
Index   0  1  2  2.33554
Index   0  6  1  2.36212
Index  -5  1  0  2.36243
Index   0  1 -5  2.37786
Index   1  6  0  2.38650
Index  -6  0  1  2.38789
Index   1  0  6  2.39233
Index   0  1 -6  2.39504
Index  -6  1  0  2.40009
Index   0  1  3  2.40689
Index   0  1  6  2.41067
Index   6  0  1  2.43899
```



Quartz

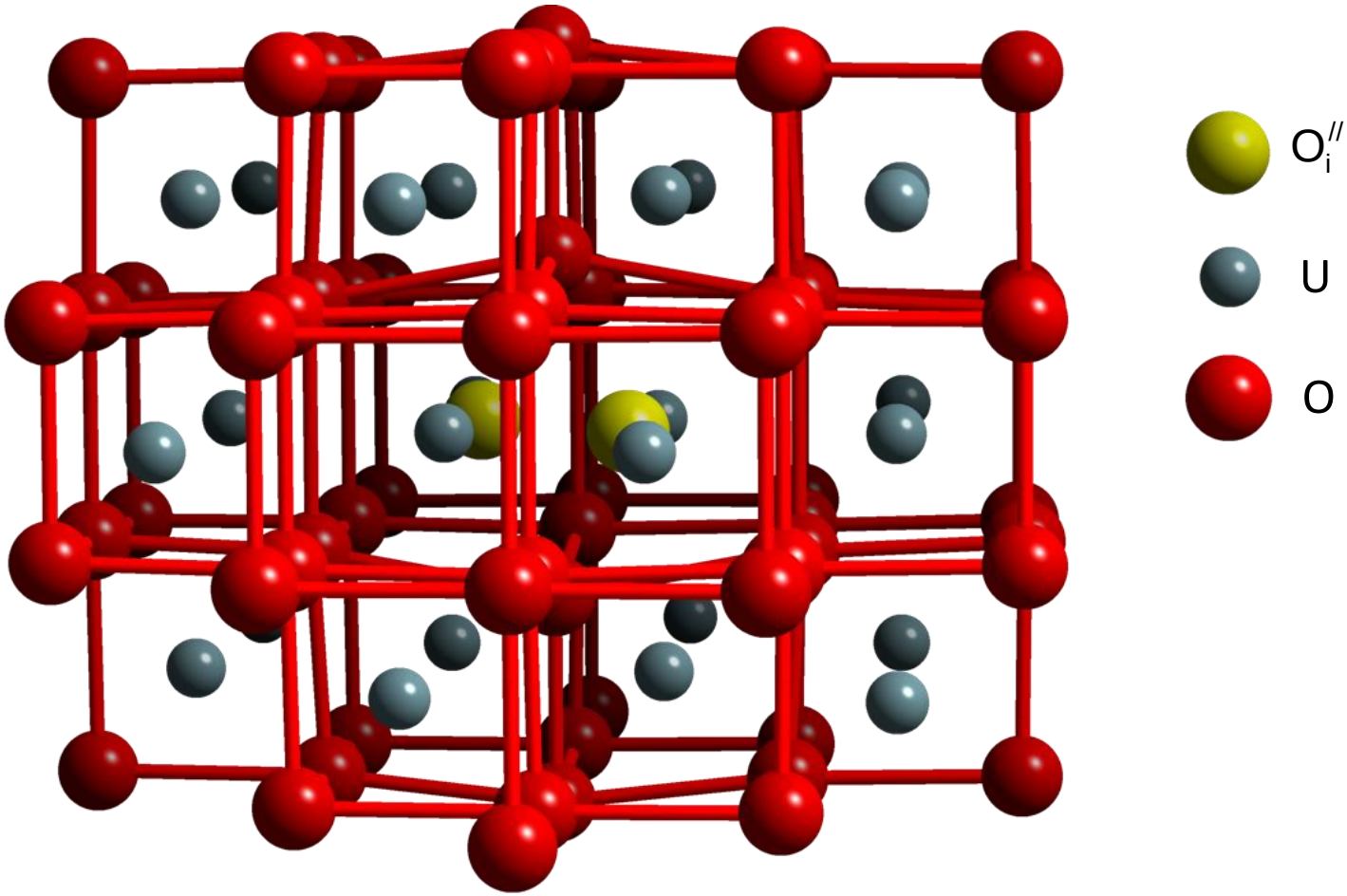


Amethyst

Quartz, SiO_2 doped with Fe^{3+} ions from Fe_2O_3.

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

Willis Cluster



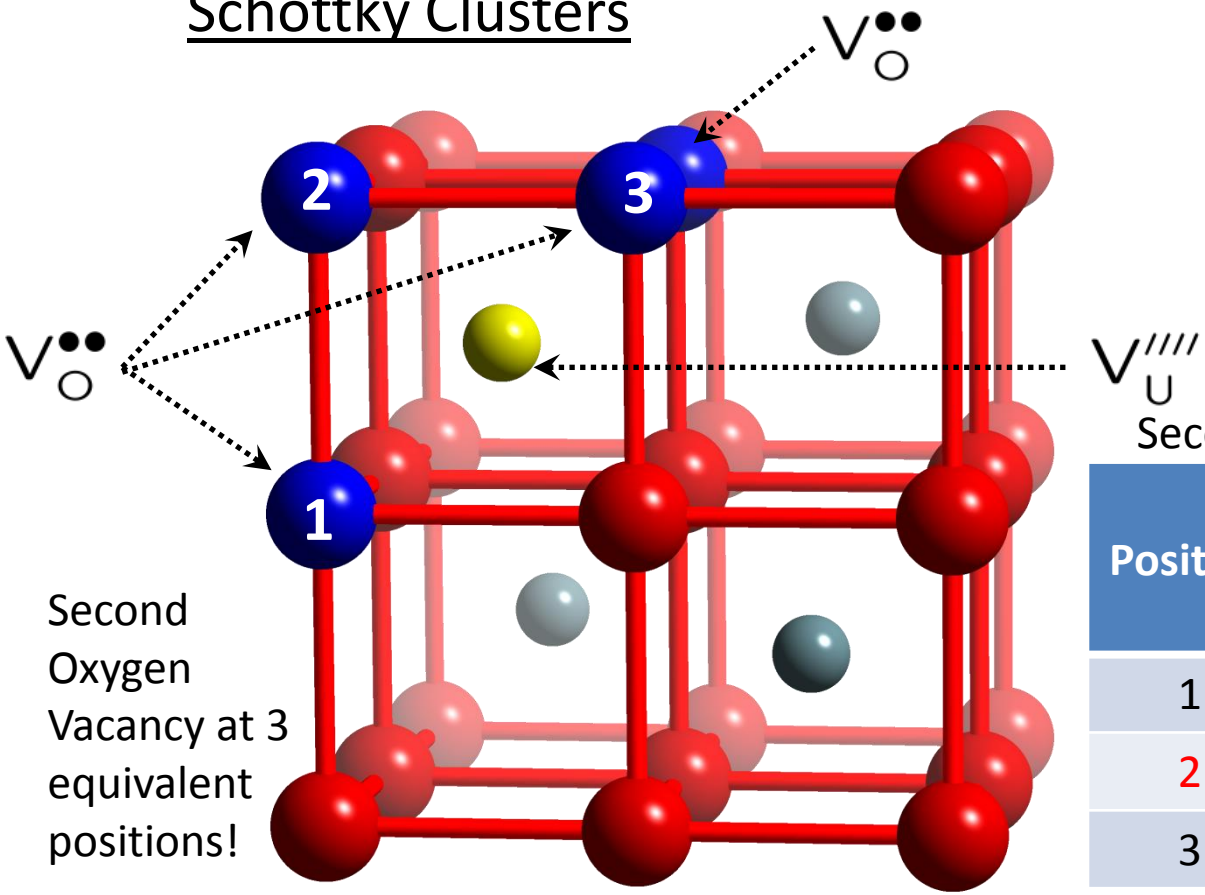
Cluster Energy = -23.21 eV

Binding Energy = -0.17 eV

Investigating the Defect Chemistry

Cluster Defects

Schottky Clusters



Second Oxygen Vacancy at 3 equivalent positions!

Second Oxygen Vacancy at position:

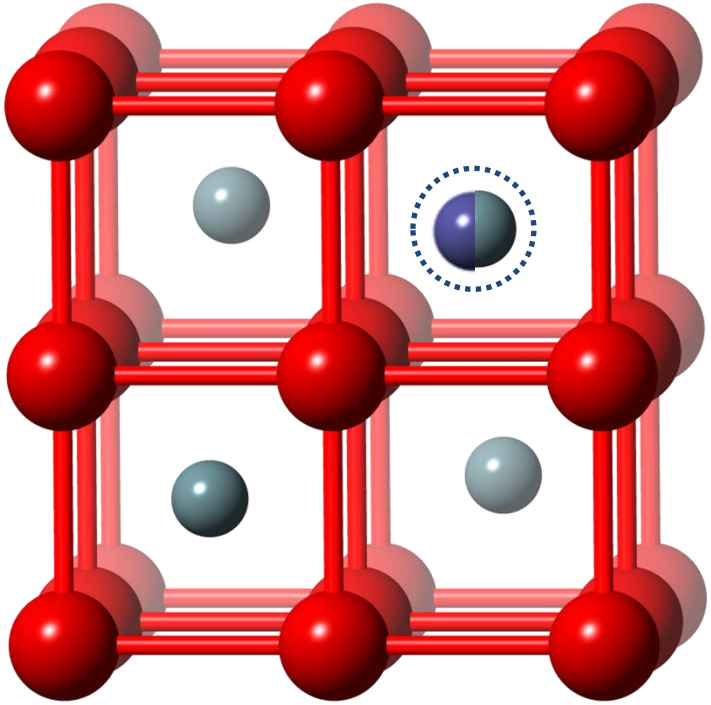
Position	Cluster Energy eV / Defect	Binding Energy eV / Defect
1	1.54	-1.08
2	1.50	-1.12
3	1.69	-0.93

$$\text{Binding Energy} = \{V_U^{////} : 2V_O^{\bullet\bullet}\}^{\times} - V_U^{////} + 2V_O^{\bullet\bullet}$$

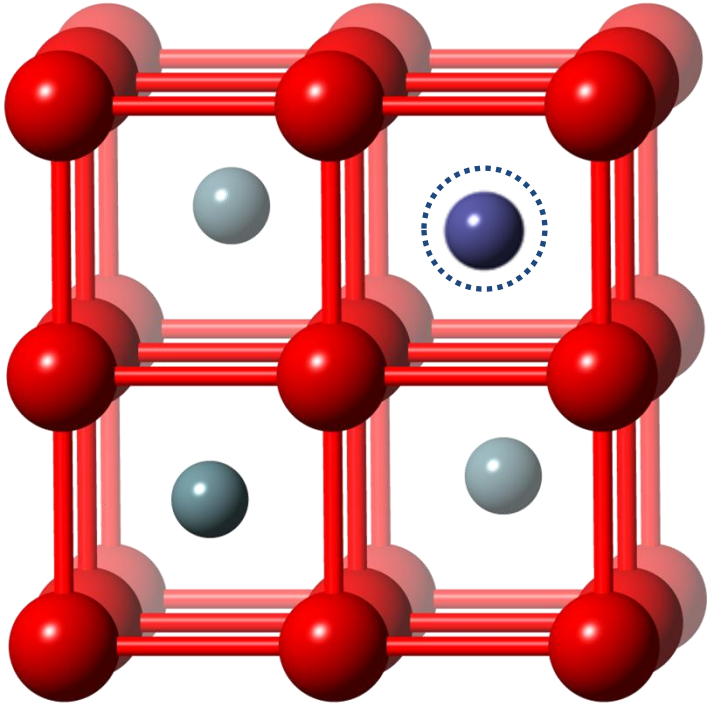
Simulating MOX fuel

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

'Mean Field Approach'



Supercell

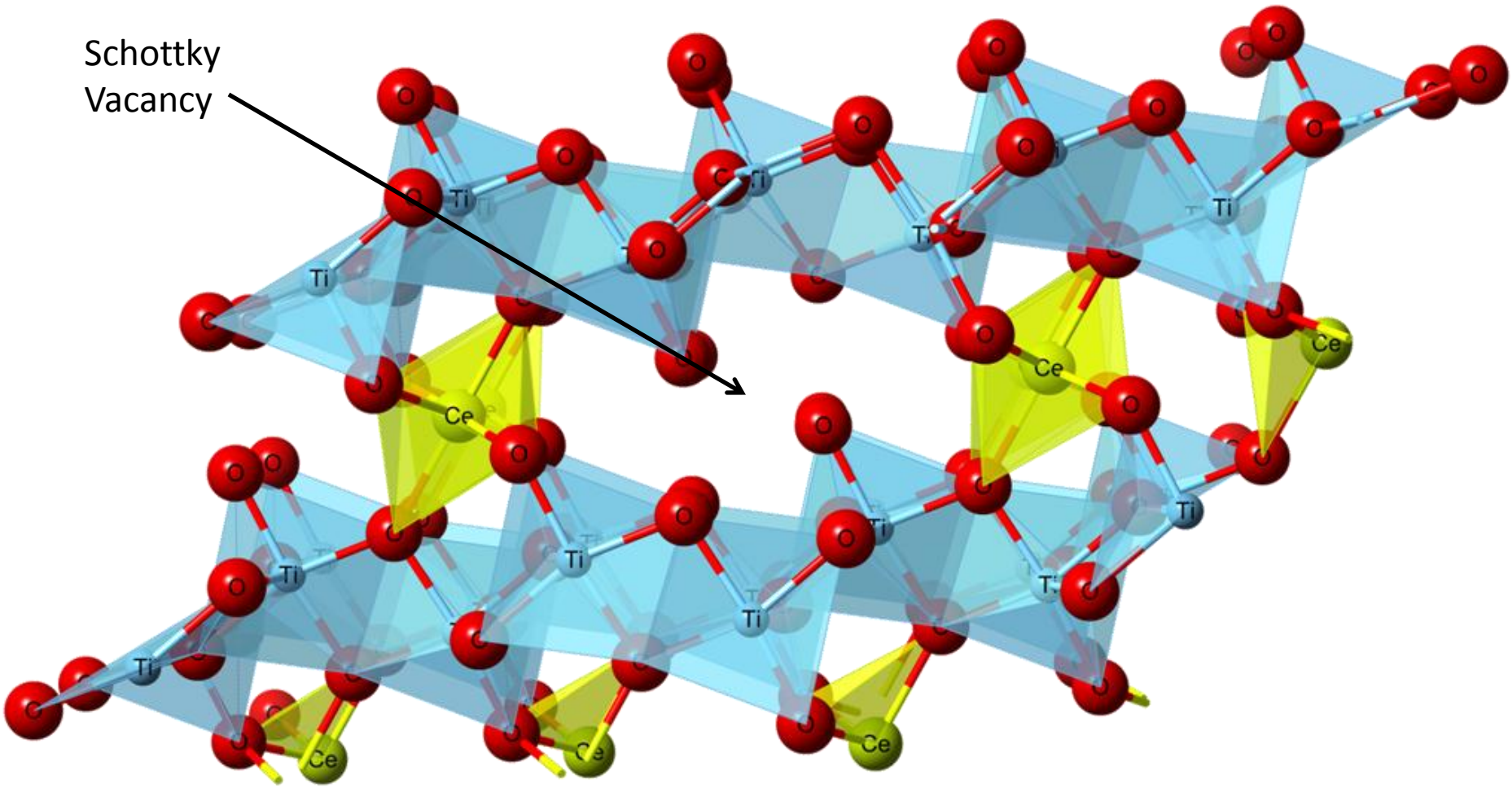


U^{4+} Ionic radius = 0.97 Å

Pu^{4+} Ionic radius = 0.93 Å

Intrinsic disorder

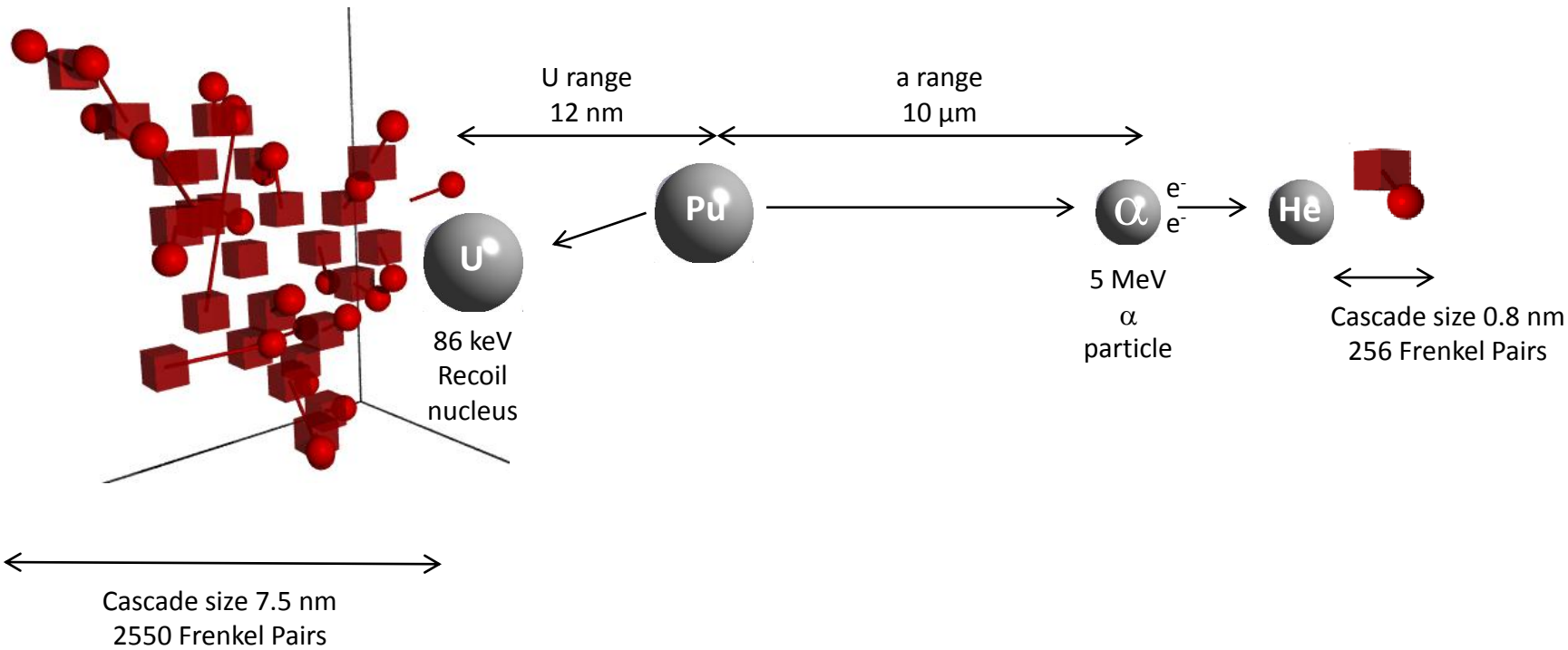
Cerium brannerite



M.C. Stennett, C.L. Freeman, A.S. Gandy, N.C. Hyatt, Journal of Solid State Chemistry 192 (2012) 172–178

Modelling Radiation Damage

Unstable Pu nucleus within MOX fuel decays principally by α decay



HPC Resource

Birmingham Environment for Academic Research

Blue BEAR
Theoretical peak performance of the compute nodes =

848 (cores) * 2.2 (GHz) * 8 (floating point operations/cycle) = 15 TFlop/s

150 TB of user disc space with a sophisticated cluster filesystem.

HPC service was completely replaced in December 2012. currently based an 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

HPC Resource

MidPlus Regional HPC Centre

A centre of excellence for computational science, engineering and mathematics

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

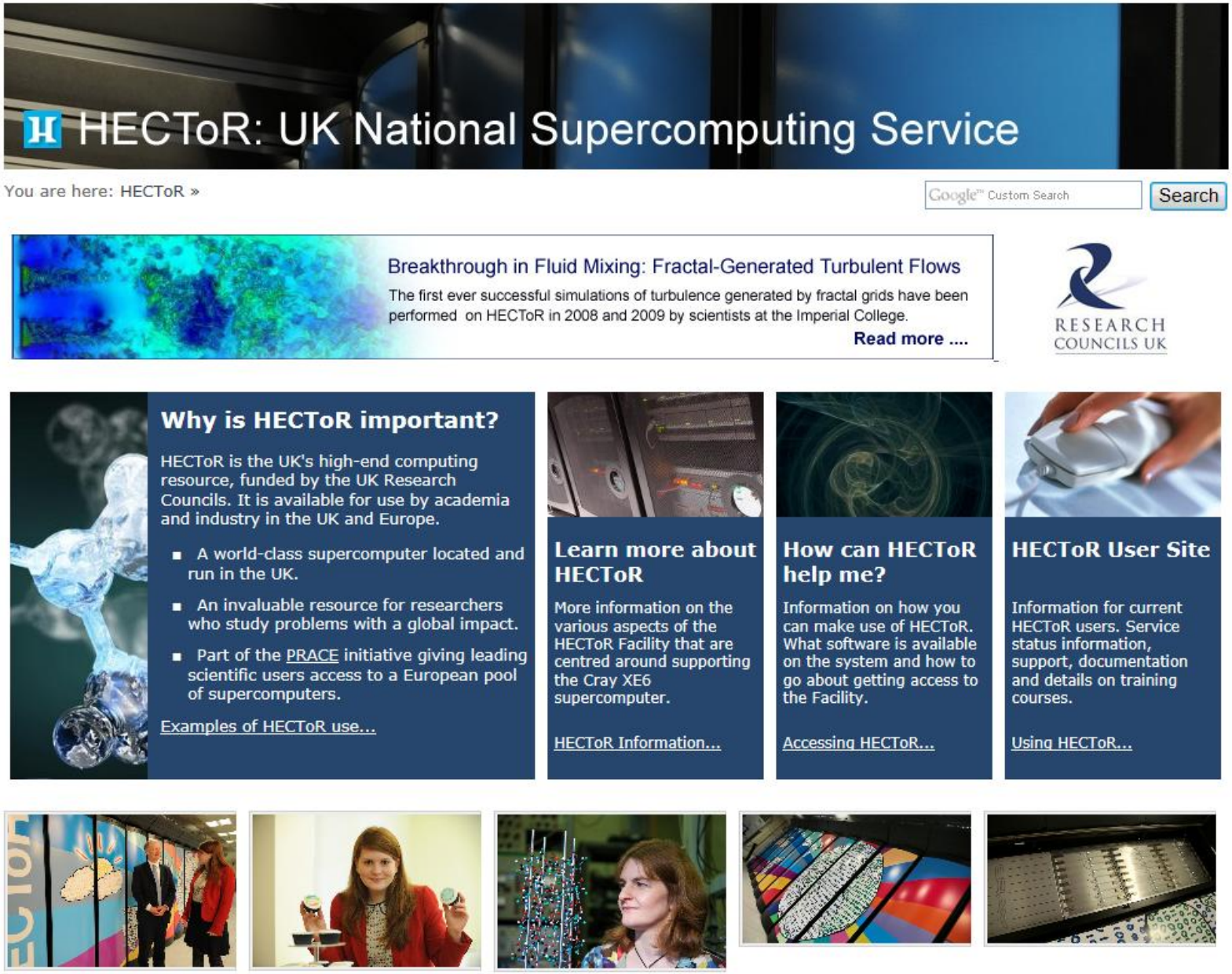
MidPlus is a collaborative partnership between four of the UK's leading universities:

- 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.

HPC Resource



Acknowledgements

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UCL

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The Midlands Energy Consortium

UNIVERSITY OF BIRMINGHAM

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The University of Nottingham