THE EFFECT OF CLUSTER RECONFIGURATION AND NON-STOICHIOMETRY ON URANIUM VACANCY MIGRATION IN UO$_2$

Michael WD Cooper, Los Alamos National Laboratory
cooper_m@lanl.gov
Simon C Middleburgh, Westinghouse Electric Sweden
Robin W Grimes, Imperial College London

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During reactor operation the release of fission gases from the fuel pellet is an important safety issue as it can lead to over-pressurization and failure of the fuel cladding. Uranium vacancy migration has been identified as the limiting step in the diffusion of fission gases through bulk UO$_2$. The uranium vacancy migration energy is, therefore, an important parameter in this phenomenon, as well as other atomic scale processes, such as recovery from radiation damage. Chemical changes under taken by the fuel during irradiation lead to deviations from stoichiometric UO$_2$ and the charge compensating defects that bind to the uranium vacancy also change. Therefore, we have examined the change in the migration energy for a uranium vacancy when bound to either two oxygen vacancies (Schottky defect) or to four U$^{5+}$ cations (hole defects) representing UO$_2$ and UO$_{2+x}$ respectively. By using empirical potentials within statics we were able to sample a large array of metastable cluster configurations to identify lower energy migration pathways that involve the reconfiguration of the cluster from the ground state configuration to metastable configurations (see UO$_{2+x}$ results in Figure 1). The work is published in ref [1].

 ![Figure 1](image)

**Figure 1 – The migration enthalpy for uranium vacancy migration in the V$_2$U+4U$^{5+}$ cluster that is associated with UO$_{2+x}$. All metastable cluster configurations are sampled within a 2x2x2 supercell and energies are reported with respect to the ground state configuration. Bound (green diamond), unbound (black square) and partially bound (remaining symbols) clusters are all shown. All data below the parity line represent lower energy pathways than the ground state configuration.**