

THE EFFECT OF CLUSTER RECONFIGURATION AND NON-STOICHIOMETRY ON URANIUM VACANCY MIGRATION IN UO_2

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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].

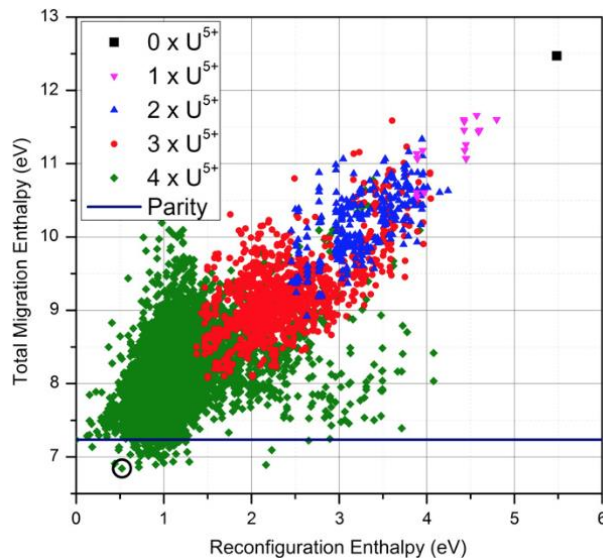


Figure 1 – The migration enthalpy for uranium vacancy migration in the $V_U+4\text{U}^{5+}$ cluster that is associated with UO_{2+x} . All metastable cluster configurations are sampled within a $2 \times 2 \times 2$ supercell and energies are reported with respect to the ground state configuration. Bound (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.