Oxygen diffusion in relation to p-type doping in uranium dioxide

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Thermally or radiation induced transport properties impact practically all engineering aspects of nuclear oxide fuels, whether at the manufacturing stage, during in-reactor operation, or under long-term repository conditions. From a more fundamental standpoint, measuring transport properties is also a means of probing point or complex defects that are responsible for atomic migration. Although many studies relating to oxygen diffusion in UO_2 have been carried out in the past forty years, none has ever focussed on characterising this property as a function of all the physical variables which determine it, i.e. temperature and composition; the latter variable being determined, depending upon the temperature range investigated, by the equilibrium oxygen partial pressure and the concentration of bi- or tri-valent impurities inevitably present on the cation sublattice.

In this work we show how electrical conductivity and intrinsic oxygen diffusion coefficient measurements can be used in conjunction to further our understanding of oxygen related point defects in UO_2. From electrical conductivity measurements performed on two types of samples containing very different doping levels, we show the positive charge carrier concentrations to be determined by the impurity concentration in the temperature range studied. The gas-solid isotopic exchange method is then used to load the material with ^{18}O tracer atoms and ^{18}O concentration profiles are then characterised using SIMS. At the oxygen potential and temperature studied (750°C), the results point to oxygen migration proceeding via an interstitial mechanism and to the fact that impurities control point defect concentrations responsible for atomic migration.