Storage of Nuclear Fuel: Radiolytic Dissolution at the UO_2/H_2O Interface

Research area: Waste and Fuel Management

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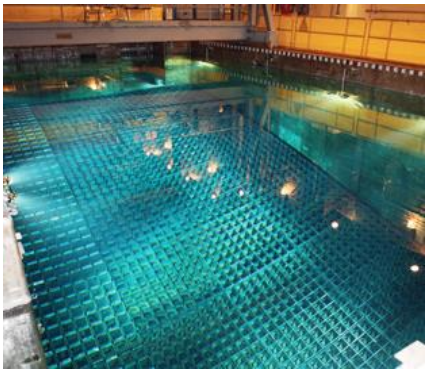
The Challenge

A key problem facing the nuclear industry is the safe and efficient long-term storage of nuclear fuel. The long-lived radiation fields that result from highly radioactive fission products require spent nuclear fuel to be stored over timescales of tens of thousands of years. This presents a very real possibility that the fuel will come into contact with water over the course of its storage lifetime.

The predominant component of nuclear fuel is uranium dioxide (UO_2), which is insoluble in water. However, the residual radioactivity of the fission daughter products, and of the fuel itself, cause the radiolytic splitting of water, yielding highly oxidising species that are able to transform UO_2 into the readily soluble UO_2^{2+} ion. In other words, this mechanism has the potential to corrode the spent fuel, causing the release of harmful radionuclides into the environment.

Figure 1



After being removed from the reactor, spent fuel is placed temporarily in large storage pools, allowing the radioactivity to decay, before it can be safely reprocessed.

The Solution

The radioactivity and the complexity of real spent nuclear fuel make experimentation on nuclear materials particularly challenging. To address this, thin films (~10 nm) of uranium dioxide have been grown at the University of Bristol. These are model fuel surfaces, of very low activity, that are extremely sensitive to any changes in the surface structure. The films were exposed simultaneously to pure water and the high intensity incident x-ray beam at both the ESRF and Diamond Light Source Synchrotron Facilities.

Figure 2

The European Synchrotron Radiation Facility (ESRF), Grenoble, France; where the first single crystal thin film dissolution experiments were performed at beamline BM28.

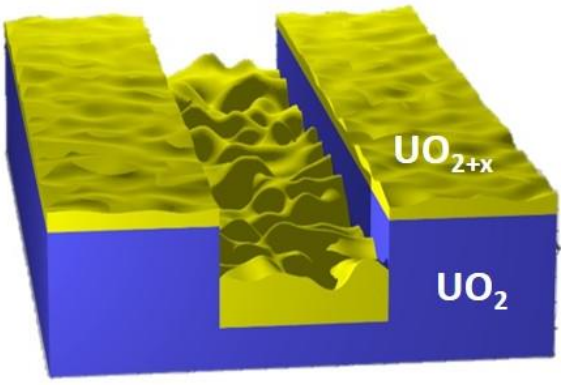
The x-rays in this case act as both source and probe; they are the source of radiolysing radiation, mimicking the radiation fields found in a spent fuel store, and they probe the surface morphology, oxide layer density and, ultimately, the dissolution of UO_2.

The Impact

Using this technique it was possible to observe the radiolytic dissolution of a model fuel in real time. Results from these measurements may help to refine predictive models of the spent fuel/water interface. Using engineered interfaces that are particularly sensitive to any change in surface structure, for instance, it is possible to investigate the role of individual variables.

These first experiments pave the way for a future research program, where further complexity can be added; investigating the effects of radiation lattice damage and fission product implantation, toward a more complete understanding of the corrosion behaviour of spent nuclear fuel. This information is important to the validation of theoretical models that attempt to predict the behaviour of fuels in contact with ground water over long timescales.

Figure 3



Along the X-ray beam path, the study found a loss of UO2 material (blue) accompanied by an increase in thickness of a hyper-stoichiometric surface layer (UO2+x) (yellow).