Long term storage of spent nuclear fuels

The Problem

One key problem facing the nuclear industry is how to store spent nuclear fuel safely in the long term. Any deep geological repository will be built to last many thousands of years, and there is the very real potential that the stored spent fuel will come into contact with groundwater. 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 into highly oxidising species. These products then cause the dissolution of the fuel with the subsequent release of fission products into the environment.

The Challenge

The radioactivity and the complexity of real spent nuclear fuel make experimentation on nuclear materials particularly challenging. A smaller-scale, simplified approach represents one way forward, where understanding the role of each variable is essential to validate theoretical models; models that attempt to predict the likely behaviour of fuels in contact with ground water over long time scales. To support modelling efforts, high resolution, surface sensitive and time resolved experimental data were needed to monitor structural changes during the radiolytic dissolution process.

The Solution

Researchers from the University of Bristol have been using X-ray diffraction on beamline I07 to monitor structural changes in single crystal thin films (4-9 nm) of UO$_2$. These are model fuel surfaces. Not only is the mass of active material low enough to make experiments easy to perform, but they are especially sensitive to any changes in the surface structure. These films were exposed simultaneously to pure water and the high intensity incident X-ray beam. The X-rays in this case act as both source and probe; they are the source of radiolyzing 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 Benefits

The team at Bristol were able 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. 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.

“We are now able to use high intensity beams of X-rays as a simultaneous radiation source and probe of the UO$_2$/water interface to better understand the radiolytic dissolution behaviour at the surface of a model nuclear fuel.”

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