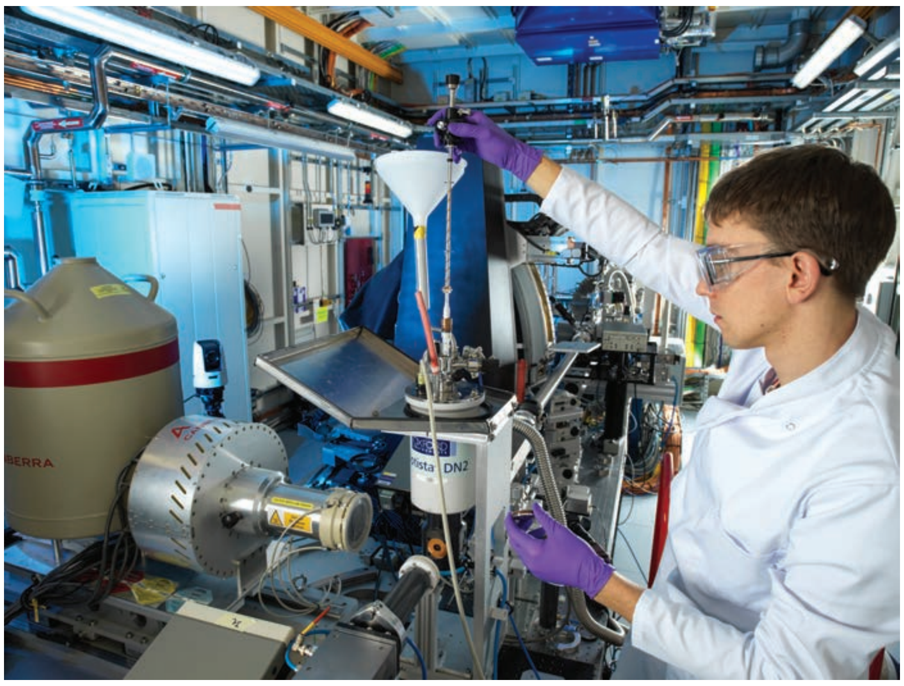
Uranium Complex Discovery offers possibilities for Nuclear Waste Management

In a collaborative study undertaken by the University of Manchester, Diamond Light Source and Radioactive Waste Management, researchers have been able to show for the first time how uranium forms a uranium-sulfur complex under conditions representative of a deep underground environment and how this complex then transforms further into highly immobile uranium oxide nanoparticles.[1]

The UK has a substantial nuclear legacy and this work further deepens the knowledge base and skills development needed to clean-up and manage radioactive wastes stored at nuclear sites or mines.

Professor Katherine Morris, Associate Dean for Research Facilities in the Faculty of Science and Engineering, University of Manchester and the Research Director for the BNFL Research Centre in Radwaste Disposal explained why recreating and studying these chemical complexes is highly relevant for understanding and dealing with radioactive waste. "To be able to predict the behaviour of the uranium during geological disposal, we need to take into account that it may have interacted with other processes taking place in the ground. These so-called biogeochemical reactions are often a complex set of interactions between dissolved chemical species, mineral surfaces, and microorganisms."

Using X-ray Absorption Spectroscopy (XAS) provided at the Diamond Light Source facility, the researchers studied uranium when it sits at the surface of the mineral ferrihydrite, a widespread mineral in the environment. The XAS data, in combination with computational modelling, showed that during the sulfidation reaction, a short-lived and novel U(VI)-persulfide complex formed during this biogeochemical process.

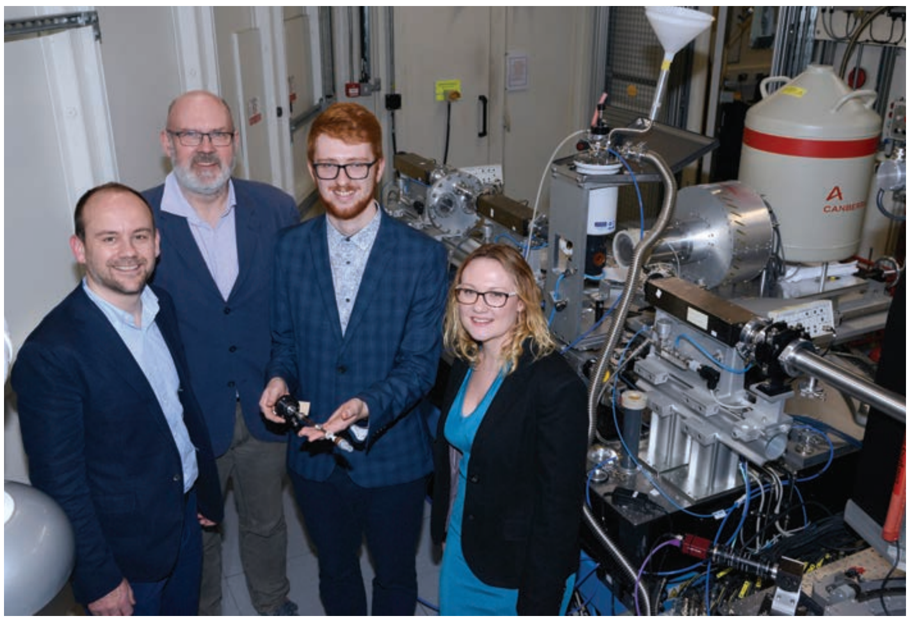


Callum Richardson, University of Manchester, PhD student working with the uranium samples on the 120 scanning beamline – Credit Diamond Light Source

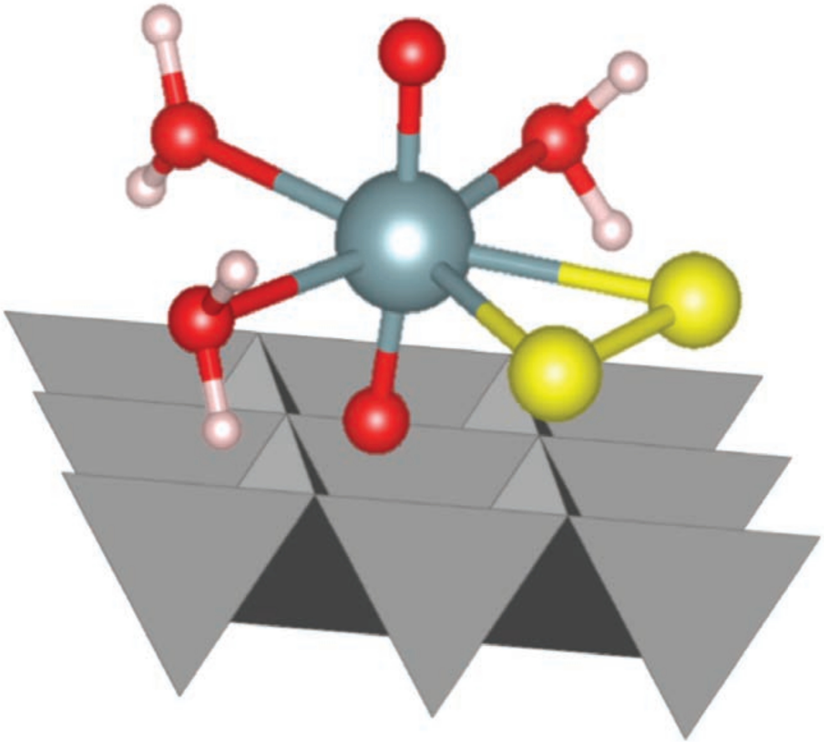
Professor Sam Shaw, Co-Investigator and Professor of Environmental Mineralogy at the University of Manchester said: "Shining the synchrotron beam onto the sample causes the uranium within to emit X-rays. By analysing the X-ray signal from the samples our team were able to determine the chemical form of uranium and to which other elements it is bound. To further validate the theory on the formation pathway of the uranium-sulfur complexes, our team also made computer simulations to conclude which type of complex is more likely to form. This is the first observation of this form of uranium under aqueous conditions and provides new insight into how uranium behaves in environments where sulfide is present. This work demonstrates the deep understanding we can develop of these complex systems and this knowledge will help underpin efforts to manage radioactive wastes in a geological disposal facility."

Demonstration of a sample containing uranium being studied and carefully loaded onto Diamond's I20-scanning beamline – Credit Diamond Light Source

Dr Luke Townsend, Postdoctoral Fellow in Environmental Radiochemistry at The University of Manchester, who undertook this research as part of his PhD further added:



From left to right: In Diamond's I20-scanning beamline: Professor Sam Shaw, Co-Investigator and Professor of Environmental Mineralogy at the University of Manchester, Fred Mosselmans, Principal Beamline Scientist at beamline I20 at Diamond Light Source, Dr Luke Townsend, Postdoctoral Fellow in Environmental Radiochemistry at The University of Manchester holding a sample, who undertook this research as part of his PhD and Dr. Rosemary Hibberd, Senior Research Manager, Radioactive Waste Management – Credit Diamond Light Source.



The uranium-persulfide complex associated with the transforming mineral surface

"When trying to mimic environmental processes in the laboratory, it's a challenge to produce accurate, high quality, reproducible science with such complex experiments, whilst also maintaining relevance to the geodisposal environment. However, obtaining exciting results such as these makes all the hard work and commitment to the project from myself and the group, both in our labs in Manchester and on the beamlines at Diamond, completely worthwhile."

The XAS measurements were performed on beamlines I20 and B18 by the researchers who used highly controlled sulfidation experiments that mimic biogeochemical processes in the deep underground environment. This was combined with geochemical analyses and computational modelling to track and understand uranium behaviour.

Physical Science Director at Diamond, Laurent Chapon concluded; "This is another example of how Diamond's state of the art analytical tools are enabling scientists to follow complex processes and help them to tackle 21st century challenges. In this instance, our beamlines allowed the users to gain real insight into the environmental relevance of this new uranium-sulfur complex, which feeds into our understanding of geological disposal."

1 http://dx.doi.org/10.1021/acs.est.9b03180
Published in Environmental Science & Technology, the paper is called "*Formation of a U(VI)-persulfide complex during environmentally relevant sulfidation of iron (oxyhydr) oxides*"