Plutonium Decontamination Agent Characterized at Berkeley Advanced Light Source

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In an on-going effort to design and synthesize chemical substances that can safely and effectively remove plutonium and other radioactive materials from the human body or from the environment, scientists at the U.S. Department of Energy's Lawrence Berkeley National Laboratory have made an important advance. Using the exceptionally bright and intense x-ray beams of Berkeley Lab's Advanced Light Source (ALS), they have determined the crystal structure of a molecular complex that has shown promise as a sequestering agent for plutonium and other members of the actinide family of elements.

"This is the first plutonium complex that has been characterized using single crystal x-ray diffraction with a synchrotron radiation source like the ALS," says Anne Gorden, the Glenn T. Seaborg Center Postdoctoral Fellow with Berkeley Lab's Chemical Sciences Division (CSD), and one of the co-authors of a paper on this work which appears in an upcoming issue of Chemistry, a European Journal.

"Taking advantage of our ability to do x-ray diffraction at the ALS, we would like to generate a library of complexes with plutonium or other actinides that will aid in the design of novel sequestering systems, and also provide benchmarks for additional structural studies with actinides," Gorden says.

Collaborating with Gorden on this research, and co-authoring the paper

in Chemistry were David Shuh, a staff scientist at the ALS, plus Bryan Tiedemann, Richard Wilson, Jide Xu and Kenneth Raymond, all of whom hold appointments with Berkeley Lab's Seaborg Center and/or the Chemistry Department of the University of California at Berkeley.

Actinides – the radioactive chemical elements that span from actinium to lawrencium on the periodic table – have generated a great deal of interest in recent years. Their use in nuclear power plants, as well as their military applications, has raised a variety of environmental and health concerns, that have been further compounded by their potential use in "dirty bomb" terrorist weapons. Concerns are especially high over plutonium because of its alpha radioactivity and the complexes it forms with bone and tissue once a person has ingested or inhaled it.

"The need for agents that will bind with plutonium and other actinides, and immobilize them for removal from living tissue, or allow them to be sequestered from the environment is more urgent than ever," says Gorden. "However, scientists first need to develop a better understanding of basic actinide chemistry."

One of the best ways scientists have of obtaining detailed information on the atomic structure and chemical bonding of molecular complexes is a technique called single-crystal x-ray diffraction. A beam of x-rays sent through a crystallized sample of the complex is scattered (diffracted) by the atoms in the crystal creating a pattern that can be imaged. This diffraction pattern can then be translated by computer into highly detailed, graphic 3-D images of the complex.

"Because it is very difficult to crystallize plutonium and other actinide complexes, being able to do the diffraction work on a synchrotron like the ALS is a huge advantage," says Gorden. "We can work with crystals as small as 15 microns to a side, and collect data in about 45 minutes that would take 10 hours or more using a conventional x-ray source."

The ALS is an electron synchrotron and storage ring, designed to accelerate electrons to energies of nearly 2.0 billion electron volts (GeV) and extract from them – using either bending, wiggler, or undulator magnetic devices – premier beams of ultraviolet and low energy or "soft" x-ray light. Light at the ALS undulator beamlines shines up to a hundred million times brighter than the light from the most powerful x-ray tubes and can be focused to a very small spot, making it ideal for diffraction crystallography studies.

Gorden and her colleagues studied their crystals with a state-of-the-art diffractometer at the ALS' Small-Crystal Crystallographic Beamline (11.3.1), which uses a bending magnet to provide highly intense beams of monochromatic x-rays in the energy range of 7 to 17 kilo electron volts (keV). This experimental setup was developed by UC Berkeley crystallographers Allen Oliver and Fred Hollander.

The plutonium crystals that Gorden and her colleagues investigated were ether-bridged hydroxypyridonate ligands nicknamed HOPO, which are based on the chelating unit found in siderophores, small molecules that are secreted by bacteria to extract and solubilize iron. For the past two decades, collaborator Ken Raymond has been leading investigations into the development of HOPO and other complexing agents that would encapsulate plutonium into tightly bound cage-like complexes.

In tests on mice, HOPO complexes have stood out as being far more effective at removing plutonium and other actinides from the body than the current approved treatments. Also, HOPO complexes could be administered orally via a pill, and experiments have demonstrated that it may even remove plutonium that has been deposited in bone.

"We expected the plutonium HOPO complex to have the same crystal structure as a cerium HOPO complex but found they were quite different, even though crystals of both were grown in water," says

Gorden.

Cerium, a member of the non-radioactive lanthanide family of elements, has been used as a safe stand-in for plutonium in structural studies. Although the two elements have different chemistry, the Ce(IV) and Pu(IV) ions have a similar size. The Berkeley scientists plan to continue their studies with lanthanides and actinides to determine if the differences in these complexes are due to solvent effects or the interactions between the ligands and these heavy metals.

“Our work with the plutonium HOPO complex indicates that a library of actual actinide structures will be necessary for developing effective actinide decorporation or sequestering agents,” Gorden says.

Source: Berkeley Lab

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