New thermodynamic model predicts plutonium solubility with iron

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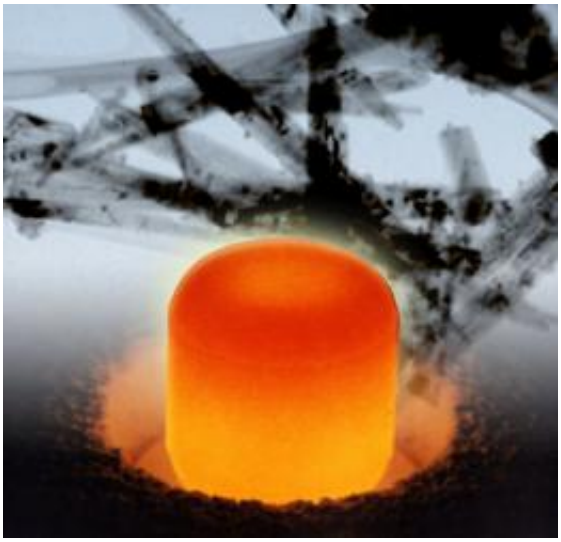

New research demonstrated the effect of goethite on reducing less-soluble plutonium to a more-soluble form.

A hard-to-detect but stable form of iron helps convert subsurface plutonium from barely to very soluble, according to scientists at Pacific Northwest National Laboratory and Rai Enviro-Chem, LLC. Plutonium resides underground at weapons sites around the world. In one form, abbreviated Pu(IV), it essentially stays put. But when soluble iron and a stable iron are present the plutonium becomes soluble, easily mixing with water. Soluble plutonium is a concern because it could mix with groundwater and flow into rivers and streams.

Underneath weapons sites, plutonium and other radioactive remnants are present and become difficult to isolate and remove. Complete removal of these materials can be costly and complex. Discovering how to keep radioactive material in the soil and out of the groundwater, whether it is in the United States, Russia, or elsewhere, could protect people and the environment without incurring extreme expenses.

The team's strategy was to move beyond bulk studies of plutonium/iron interactions. They explored the microscopic and molecular processes involved in the reaction. This research has not been tried before because of the samples' radioactivity, which requires specialized facilities and equipment.

The team coupled measurements of plutonium concentrations in water with scanning and transmission electron microscopy analysis of the reaction. The measurements showed that in the presence of soluble iron and a more stable phase of iron, known as goethite, the reaction is enhanced. The two forms of iron speed the formation of soluble plutonium. With this data, the scientists developed the first thermodynamic theory of such an effect.

"This work is a major step forward in understanding the molecular mechanisms that direct the changes in the chemical forms of plutonium," said Dr. Andrew Felmy, a geochemist and lead author of the study.

Because of continued interest in plutonium's behavior in the environment, from both environmental and security angles, the researchers will continue to perform research directed at identifying and understanding subsurface reactions involving iron and plutonium. New capabilities in EMSL, a national scientific user facility, will play a critical role in this research.

More information: Felmy AR, et al. 2011. "Heterogeneous Reduction of PuO2 with Fe(II): Importance of the Fe(III) Reaction Product." *Environmental Science & Technology* 45:3952-3958. DOI: 10.1021/es104212g