

Plutonium May Be Hitching a Ride on Colloids

If nuclear waste leaks out of its underground containers, how far is it likely to migrate? That's a critical question to answer, with hundreds of thousands of cubic meters of waste from nuclear power and nuclear weapons operations now awaiting permanent burial. It has long been thought that at least one of the more toxic contaminants—plutonium—would stay put. Because of its low solubility in water and its tenacious capacity to cling to mineral surfaces, some argued, plutonium would remain adsorbed on local rocks. That viewpoint has been challenged in the last 15 years by suggestions that plutonium can adhere to submicrometer-sized colloids and thereby be transported considerable distances by groundwater. This suggestion, however, has not been strongly supported by field studies. A recent study by researchers from Lawrence Livermore National Laboratory and Los Alamos National Laboratory¹ now provides the firmest evidence to date that, at least in one case, the plutonium has migrated 1.3 km from its source in 30 years—a speed that is consistent with the flow of groundwater in the area.

The Livermore–Los Alamos study implies, but stops short of proving, that plutonium traveled so far by adhering to colloids. (Typical colloids—clays and zeolites—found in the study region are pictured on this page.) The study reinforces the decision by the Department of Energy (DOE) to include, for the first time, the possibility of colloidal transport of plutonium in its latest study of possible future contamination at the proposed Yucca Mountain nuclear waste repository.²

Identifying the source

Since roughly the mid-1980s, concentrations of plutonium have been seen farther afield from their burial sites than was expected.³⁻⁵ However, the sources of the plutonium were never unambiguously identified, and there was always the possibility that the observed plutonium might have seeped into the ground from the surface (deposits on the surface could stem from plutonium still in the air as a legacy of atmospheric weapons tests). Furthermore, field studies indicated that colloids travel no more than a few tens of meters in groundwater.

In the recent study,¹ a Livermore–Los Alamos collaboration led by Annie Kersting of Livermore studied the transport of plutonium away from the sites of nuclear weapons tests in the Pahute Mesa region of the Nevada Test Site. The experimenters were able to

Measurements of isotopic ratios link plutonium measured in sampling wells at the Nevada Test Site to a particular underground weapons test site 1.3 km away.

establish definitively that plutonium had migrated in groundwater from one specific nuclear test site to a sampling well. They made this connection by measuring a signature of the plutonium from each weapons test: its ratio of ^{240}Pu to ^{239}Pu . This ratio has distinguishably different values at each of four sites of underground nuclear explosions in the study region. When Kersting and her colleagues measured the isotopic ratios of plutonium in groundwater pumped from two wells situated more than a kilometer from the four nuclear test sites, they found a match with only one of the four: the 1968 Benham test (which was the deepest of the four tests). Thus, the researchers showed that the distant plutonium stems from the Benham test and none other.

The quantities of plutonium found at the distant wells were small, on the order of 10^{-14} moles per liter, and represent only a very small fraction of the plutonium associated with the Benham test.

How did the plutonium travel so far? Most likely attached to colloids, concluded the members of the Livermore–Los Alamos collaboration. To explore this possibility, they filtered some of the groundwater pumped from the wells, using three different filter sizes to separate out the very small particles (colloids) ranging in size from about 7 nm to 1 μm . Filtering removed more than 99%

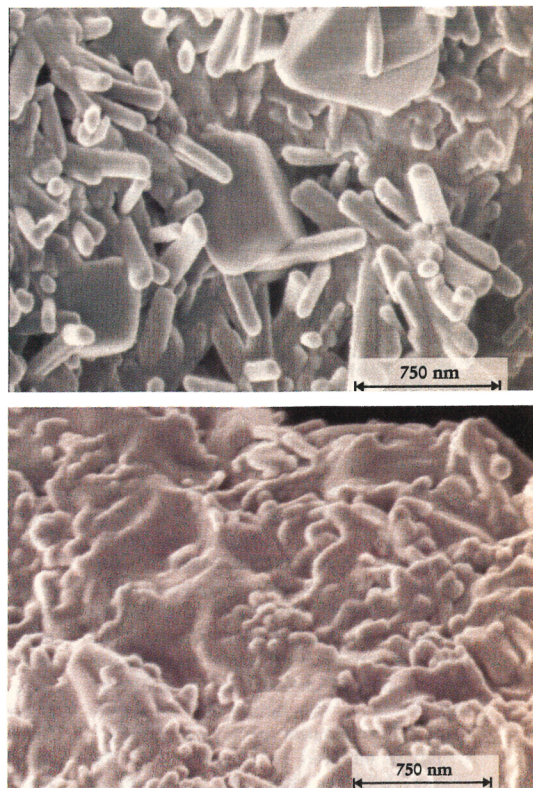
COLLOIDAL MATERIAL found in association with plutonium in groundwater taken from a Nevada Test Site sampling well, 1.3 km from the site of a 1968 underground nuclear test explosion. Based on these scanning electron microscope images, as well as on x-ray diffraction analyses, the experimenters from Lawrence Livermore and Los Alamos National Laboratories feel that the rodlike and rhombohedral structures (top) are most likely mordenites and clinoptilolites, both members of the zeolite family, and that platy materials (bottom) are clays.

of the plutonium and other radionuclides from the aqueous phase, showing that the measured plutonium was associated almost exclusively with the colloidal material.

Remaining questions

Although the new measurements are strong evidence for the role that colloids can play in transporting plutonium significant distances, the case is not fully proved. One alternate explanation, for example, is the scenario of “prompt injection” of the plutonium during the nuclear test: Perhaps the explosion opened fissures in the surrounding rock and the plutonium was transported in a gaseous phase through those fissures. Kersting and her colleagues argue that this scenario might explain plutonium migration on the order of tens to a few hundreds of meters but not at distances greater than a kilometer. It might be possible, however, for the plutonium to have been transported at least part of the observed distance by this mechanism.

The Livermore–Los Alamos team is still studying the types of colloids found in the ground water pumped from the sampling wells to determine those colloids that selectively absorbed the plutonium. The experimenters would also like to explore whether the plutonium



formed its own colloids, which it can do under some circumstances. Some of the colloids present are typical of those that can, in the presence of water, spill off the waste glass formed by a nuclear explosion.

Kersting told us that there is some concern that the pumping of groundwater out of the well may have increased the proportion of colloids in the water. This summer, her group plans to study the water flow through fractures without pumping to understand the natural concentration of colloids in different groundwaters.

There are many other questions that need answering. How reversible is plutonium sorption onto various colloids? How far can colloids travel in groundwater? Does the low concentration of plutonium observed at the Nevada Test Site sampling wells reflect the long distance traveled, a low concentration of plutonium incorporated into the waste glass at the explosion site or a low concentration of plutonium absorbed onto naturally occurring colloids in the underground flows? Did the nuclear explosions at the Nevada Test Site create fractures that facilitate groundwater flow, or did they merely amplify the effect of fractures already occurring naturally at the site?

Applicability to other sites?

Of course, one of the largest questions is whether the finding at the Nevada Test Site is applicable to other types of waste and to other disposal sites, where the geology and hydrology may be considerably different. Of particular concern are the Hanford Nuclear Reservation near Richland, Washington, where, for many years, plutonium-containing waste has been buried in trenches or stored in leaky million-liter storage tanks, and Yucca Mountain in Nevada, where DOE is exploring the establishment of a permanent repository for spent fuel rods from nuclear power plants and nuclear waste from defense operations. (See the special issue on radioactive waste, PHYSICS TODAY, June 1997.) Although so far there are few data for assessing the possibility of colloidal transport at the Yucca Mountain site, planners there are including all possibilities. Just last December, the DOE released its *Viability Assessment of a Repository at Yucca Mountain*—a report mandated by Congress—which was the first such analysis to include potential plutonium transport by colloids in estimating the possible future radiologic contamination.² Abe van Luik of the Yucca Mountain Project, who heads the total system performance analysis (TSPA), which is covered in chapter 3 of the report, told us that the inclusion of

colloidal transport made a difference; in TSPA models, plutonium made a major contribution to the radiation dose at a point 20 km from the repository, but only some hundreds of thousands of years after burial.

Like the analysts at Yucca Mountain, researchers at other DOE sites where nuclear waste is stored are studying aspects of colloidal transport. The concern is not limited to transport of plutonium. For example, quite a bit of cesium and other radionuclides has moved unexpected distances from storage sites where containment has failed. Preliminary work suggests that transport was facilitated by the makeup of the solutions in which the contaminants were disposed, but colloidal contributions have not been ruled out. Van Luik cautions that “care must be taken in using the data or insights from

one location, waste stream or radionuclide to make assumptions about phenomena at other locations.” Nevertheless, he adds, researchers working on various aspects of radionuclide transport can learn from one another.

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Low-Energy Electron Beams Modify Semiconductor Surfaces

It is a truth universally acknowledged—at least in quantum mechanics—that you can’t observe something without changing it. But, until recently, it’s been widely assumed that the low-energy electron beams that form the basis of low-energy electron diffraction (LEED) and Auger electron spectroscopy (AES) do not significantly alter clean semiconductor surfaces.

Testing this conventional wisdom, the University of Minnesota’s Koji Nakayama and John Weaver found that, in fact, electron beams do create defects on silicon and gallium arsenide surfaces.¹ Their results not only emphasize that LEED and AES should be used with care, but also hint that electron beams could be used instead of chemicals to etch semiconductor surfaces.

Electrons as surface probes

In LEED, electrons are fired perpendicularly at a surface to probe its symmetry and structure. The electrons diffract because the energies they’re accelerated to—typically 5–500 eV—confer wavelengths that are about the same size as the atomic separation at the surface.

To bounce back from a surface, electrons must interact strongly with it. In doing so, they careen off more than one surface atom—with or without losing energy. Determining surface structure from a LEED diffraction pattern, therefore, is tricky. Indeed, from Clinton Davisson and Lester Germer’s original 1927 demonstration of electron diffraction, it took 40 years for theorists to

Recent experiments suggest that electron beams could be used to pattern semiconductor chips.

forge and hone the requisite mathematical tools.

Nowadays, thanks largely to this theoretical investment, LEED is one of the most successful techniques for determining quantitatively how atoms are arranged on a surface.

Like LEED, AES also exploits electron beams, but in a different way. Electrons are fired at a surface to provoke the ejection of atomic electrons through the Auger process. Analyzing the resulting electron spectrum reveals the identity and number of the atoms on the surface. If you want to know what impurities are covering a surface, AES is your tool.

Nakayama and Weaver did not set out to examine the limitations of LEED and AES. Rather, the focus of their investigation was etching.

In dry etching, halogen atoms are wafted onto a semiconductor surface, where, like lions preying on a herd of zebras, they separate and seize the most weakly attached surface atoms. Electron beams aren’t generally used as etchants, but, as observed first by Paul Redhead² and Dietrich Menzel and Robert Gomer,³ they can pry loose gases adsorbed on metallic surfaces.

To find out whether electrons would aid and abet halogen etchants, Nakayama (who has just moved to Tokyo University) decided first to as-