

## Plutonium Under Glass

Wherever the U.S. decides to put its high-level nuclear waste, planners must also decide how to store a particularly worrisome component: plutonium. There are an estimated 50 tons of the radioactive metal in the U.S. alone, left over from the production of nuclear weapons and their subsequent dismantling under disarmament treaties. Policy-makers are considering converting some of this stockpile into fuel for commercial nuclear reactors. The rest will have to be stored. Now researchers have come up with a new scheme for putting plutonium safely under glass.

Glass has long been considered a good storage medium for nuclear waste because it can be engineered to be stable over thousands of years, resist corrosion, and can easily encapsulate other types of contaminated waste such as incinerator ash. The type of glass traditionally used to entrap nuclear waste has one big drawback, however: It wasn't designed to hold more than a trace amount of plutonium. But at the Materials Research Society meeting in Boston 2 weeks ago, researchers from Argonne National Laboratory reported that a new glass formulation has the potential to incorporate up to 10% plutonium by weight.

Although the new glass, known as alkali-tin-silicate (ATS) glass, must still prove itself under long-term testing, other researchers say these results are welcome news. "If you can't get [plutonium] off the face of the Earth, putting it in that form may be the best option," says Mal McKibben, a chemist at the Department of Energy's Defense Waste Processing Facility in Aiken, South Carolina. Other potential storage materials, such as ceramics, are more expensive and harder to produce. Moreover, ATS glass can be spiked with highly radioactive cesium to deter terrorists from stealing the glass to extract the plutonium for nuclear weapons fuel.

ATS is, of course, far from pure silica glass. Plutonium can't be stored in pure silica glass because the metal atoms tend to congregate—a dangerous prospect, because a large enough cluster could form a critical mass. To prevent this, the Argonne chem-

ists—led by Adam Ellison and John Bates—loaded ATS glass with alkali metals, such as sodium, lithium, and potassium. These positively charged atoms bind to negatively charged oxygens in the glass, creating compounds called nonbridging oxygens (NBOs), which still harbor a slight negative charge. Plutonium

atoms carry a strong positive charge, so when they are mixed in the glass each one binds to several NBOs. The NBOs thus form layers around each plutonium atom, dispersing them throughout the glass network.

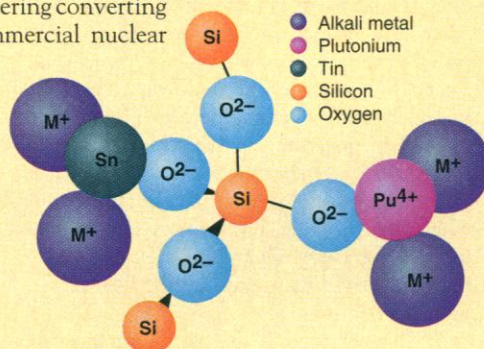
In addition to scattering plutonium atoms, the alkalis perform other useful roles, such as lowering the melting temperature of the glass to 1150 degrees Celsius. Unlike "lanthanide-rich glass"—an alternative formulation for high-plutonium-content glass that melts at a higher temperature—that's low enough to incorporate cesium, which vaporizes at around 1200°C, into the matrix.

Loading ATS glass with alkalis does create a problem, however. Alkalis are fickle: Unless they are stabilized by plutonium or a metal with a similar electronic behavior, they will drop their connections to oxygen and leach out of the glass. That would then speed the glass's decay and free the plutonium atoms to assemble in dangerous congregations.

Adding even more plutonium as a stabilizer was ruled out—more than 10% in the glass could make the mixture potentially explosive during processing. So instead, the researchers chose other positively charged metals, including tin and zirconium.

Thus far, the recipe seems a good one. After subjecting an ATS sample loaded with 7% plutonium to 56 days of standard durability tests—including continuous exposure to high temperatures and water vapor—the sample showed no evidence of corrosion or signs that the plutonium was congregating in the material. Says Bates: "The rate of reaction is similar to the natural glass obsidian," which survives intact for millions of years.

—Robert F. Service



**Glass trap.** Alkali metals help bind and disperse plutonium safely throughout a new glass formulation.

SOURCE: J. BATES AND A. ELLISON

that water won't drip back in. This would be impossible with closely spaced tunnels, because the zones would overlap to an even greater degree, eliminating drainage routes. Finally, an insulating blanket of gravel and sand around the containers would help keep the temperature at the canister surfaces higher—and corrosive humidity lower—than in the rock. Even after the waste has cooled for 100,000 years, Buscheck says, these measures will keep the packages dryer than ambient conditions.

But critics such as hydrogeologist Nick Stellevato, Nye County's technical representative at Yucca Mountain, warn that subjecting the mountain's highly fractured interior to above-boiling temperatures for many years could alter its strength and permeability to water. "With a high thermal load, you are going to have localized effects on stability around the tunnels," Stellevato says. Scott

Tyler, a hydrologist at the Desert Research Institute in Reno, Nevada, adds that "water has the potential to run back down the fractures," picking up corrosive mineral solutes along the way.

Buscheck agrees that his group's calculations need to be verified, and says his group is planning to do just that by placing large electric heaters in tunnels and measuring temperature and vapor movement behind the tunnel walls. But he points out that under the localized dryout plan, less rock would have to be heated to achieve the same humidity reductions in the tunnels. Even if ground water inundated the tunnels, the temperature difference created by the gravel blanket would keep the waste packages drier than the surrounding rock. And because the blanketing effect strongly reduces humidity at the waste package surface far into the future no matter what the repository's overall

thermal output, he adds, officials could take measures such as adjusting the waste packing density, or ventilating excess heat from the tunnels, to limit the period of boiling to as little as 100 years—within the repository's planned observation period.

Still, this and other aspects of the Yucca Mountain plan have a long way to go before the repository gets a green light. A recent NAS report, for example, found deficiencies in DOE's analysis of the potential for surface erosion at the site. The project is a magnet for criticism, and chemist John Bates, head of the nuclear waste office at Argonne National Laboratory, says anything pertaining to safety is relevant: "If you don't have a convincing technical basis for safety, you won't get [Nuclear Regulatory Commission] approval, and I wouldn't expect public approval either."

—Wade Roush