Changing a process leads to purer Pm-147, and more of it

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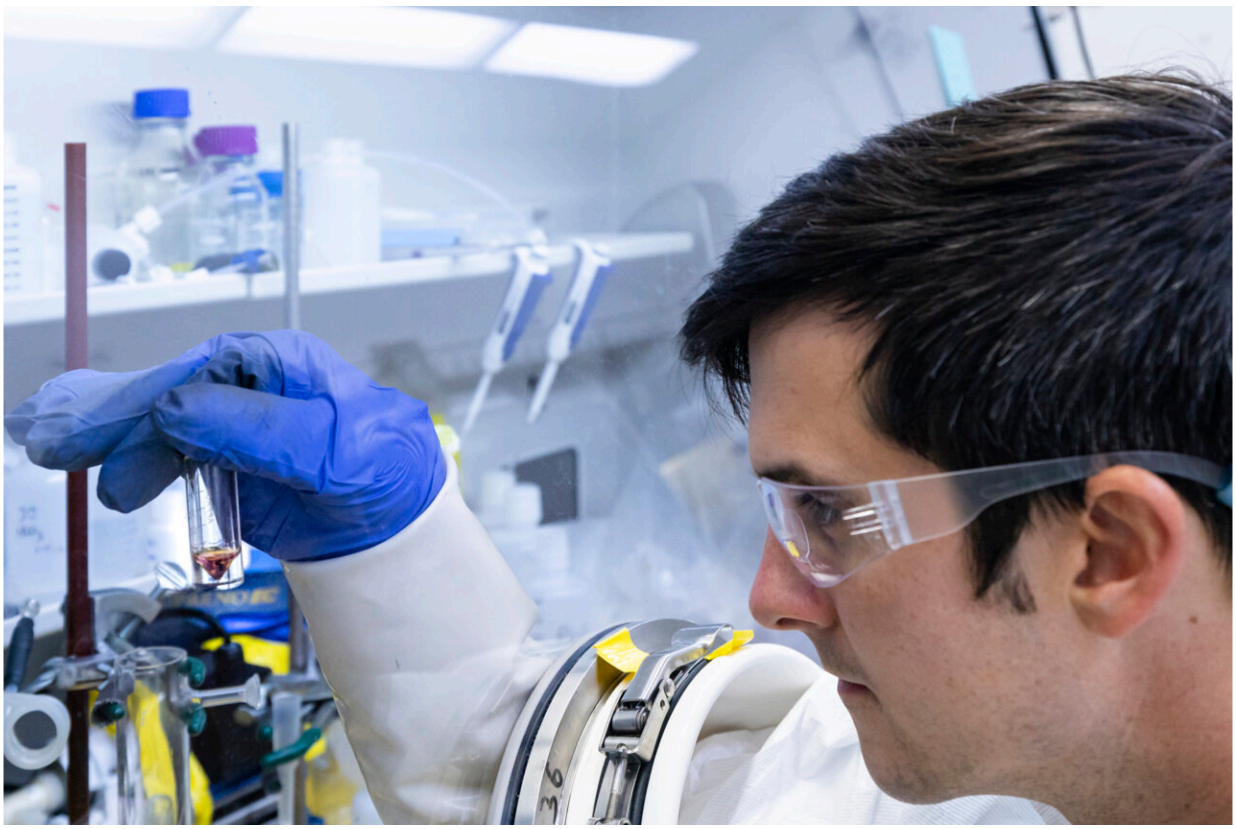

ORNL radiochemist Thomas Dyke works on the purification of promethium-147 in a glove box in the REDC. Credit: Carlos Jones/ORNL, U.S. Dept. of Energy

With larger, purer shipments on a more frequent basis, Oak Ridge

National Laboratory is moving closer to routine production of promethium-147.

That's thanks in part to the application of some specific research performed a decade ago for a completely different project.

ORNL's Pm-147 production began in 2020, with the Department of Energy's Isotope Program's effort to retrieve the valuable isotope from the waste stream of the NASA's plutonium-238 program.

ORNL produces Pu-238 to fuel deep space travel, and Pm-147—used for thin film measurements and nuclear batteries—is a fission byproduct when Pu-238 is separated from irradiated neptunium-237 targets. Its history at ORNL is long: Promethium, element 61, was discovered here in 1945 by scientists Jacob Marinsky, Lawrence Glendenin and Charles Coryell, who were working on uranium fuel irradiated in the graphite reactor during the Manhattan Project.

The plan now is to reestablish domestic production of Pm-147, which is in short supply, while reducing the concentrations of radioactive elements in the Pu-238 waste stream, so that it can be disposed of safely in simpler, less expensive ways both now and in the future.

It's also a pilot, of sorts: If Pm-147 can successfully be mined from Pu-238 leftovers, so—eventually—might other valuable isotopes.

"We're looking at how to get more out of what we would traditionally call 'waste,' to look at what else is produced and whether we could decrease our waste burden by mining more useful radioisotopes from it," said Richard Mayes, the senior R&D staff member in the Isotope Applications Group who has led the effort.

But first, Mayes and the Pm-147 team—which includes Lætitia Delmau, Thomas Dyke, Tony Dyer and new team members Matt Silveira and April Miller—have been chipping away at the challenges involved with extracting the isotope from the rest of the fission products and purifying it.

Last year, the team began using a different method for removing the radioactive element curium from the mixed waste from which Pm-147 is extracted—a key step in production. Previously, curium was separated using the TALSPEAK (Trivalent Actinide Lanthanide Separation with Phosphorus-Reagent Extraction from Aqueous Komplexes) process developed at ORNL in the 1960s.

In looking at ways to increase purity and production, however, Delmau recalled a method she'd worked on more than a decade ago as part of Sigma Team for Advanced Actinide Recycle (STAAR). In 2010, while separating isotopes for that program, Delmau and ORNL chemist Kayron Rogers, then a student at Tennessee Technological University in Cookeville, Tenn., successfully used a new ligand—a molecule or ion that binds selectively to another ion, forming a chemical structure with unique properties—developed by an organic chemist with Tennessee Tech.

Delmau suggested using the ligand, camphor-BTP, for Pm-147 production. A small test was successful, and the team ordered a batch of the ligand from an outside lab and changed processes, dramatically improving the results.

"I knew how it behaved and what to ask for," Delmau said. "It allowed us to improve greatly on what we had."


APPROX

Purified Pm-147 has gone through several steps in ORNL's Radiochemical Engineering and Development Center. Credit: Carlos Jones/ORNL, U.S. Dept. of Energy

While the TALSPEAK method extracted the Pm-147, leaving the curium behind, the camphor-BTP method instead removed the curium from the mix, allowing for a more efficient series of separations afterward to further purify the Pm-147. The ligand separation method maintained high curium removal even under less-than-optimal conditions and resulted in less Pm-147 loss during separation. Delmau said the team will continue to tweak the process for even better results.

Mining an isotope from fission byproducts has unique challenges. There's a time element, for one: The longer the mixture sits, the more it will decay, leaving less to be extracted.

Also, the highly radioactive nature of the liquid waste solution from the Pu-238 campaign requires the initial separation—where the bulk of impurities present in the large volume of waste are left behind and only promethium and other elements with similar chemical properties are separated, collected, and concentrated—to be done in the heavily shielded hot cells at ORNL's Radiochemical Engineering Development Center, or REDC. Afterward, curium removal and other fine separations are done in the light-duty shielded cave at REDC, and then the Pm-147 is purified in a glove box.

"We have to utilize every capability at REDC, from the big hot cells to the cave to the glove boxes, to manage our dose for worker safety," Mayes said.

The team must schedule the Pm-147 production as close as possible after Pu-238 production wraps up, but without disrupting other hot cell operations, and work quickly so as not to strain other programs. Delmau said the addition of Silveira and Miller to the team last year has significantly helped grow production.

Another challenge is that the fission byproduct mix the team receives can vary in both size and composition, she said.

"We start with something that is everchanging," Delmau said, quipping, "What comes out of the hot cell is like a box of chocolates. You never know what you're going to get."

Much work now is going toward finding the best way to separate other metals from the mix after curium is removed. Still, the team is confident they're in the "early stages" of standardizing processes for Pm-147 production.

Already, improvements to processing have meant more Pm-147 per campaign. ORNL sends the finished product to the National Isotope Development Center, which then supplies it to customers. Right now, ORNL is the nation's only producer of Pm-147, for which there are currently three buyers: two that use it in research, and one that uses it in an instrument that measures the thickness of thin plastic films.

Last fall was the first time ORNL-produced Pm-147 was shipped to multiple customers, Mayes said.

"By supporting multiple customers and expanding our customer-base, we are providing stability for the program," he said.

Provided by Oak Ridge National Laboratory

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