Analyzing interactions between radiation and matter for a long-term plutonium storage strategy

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Long-running research between the UK's National Nuclear Laboratory and The University of Manchester, UK, has probed the complex interplay between the surface features of plutonium dioxide and its surrounding environment. The new results provide even more confidence in a long-term storage strategy.

In many nations around the world—including the UK, United States, Japan, and France—plutonium is separated from used nuclear fuel and then stored as an oxide powder inside specially designed metal canisters. In the UK, the eventual fate of this powder hasn't yet been determined; it could be disposed of deep underground or it could be used to make new fuel for future nuclear energy.

Some of the material in the UK has been in storage for nearly 50 years, mainly on the Sellafield nuclear site in West Cumbria. Just like other radioactive elements, plutonium undergoes radioactive decay, so the conditions in the canisters gradually change over time.

Dr. Robin Orr, Senior Technology Manager at the National Nuclear Laboratory, has a wealth of experience in researching plutonium: "The safe and secure storage of plutonium is of national importance. All available evidence confirms our current solution is the right one. This new research forms part of this evidence by providing insight into the complex and dynamic interactions between plutonium and the environment inside the storage canisters."

"This insight allows us to better understand the fundamental science and engineering requirements in more depth so that we can inform Sellafield Limited's storage and conditioning solutions."

Plutonium dioxide has a strong affinity for water and will readily 'suck' it out of the atmosphere. Historically, laboratory experiments have shown that the energy from ionizing radiation is sufficient to break this water down, forming hydrogen gas. This rarely happens in the storage cans, suggesting that some extra process suppresses hydrogen generation.

A unique opportunity

New research led by the UK's National Nuclear Laboratory on behalf of the Nuclear Decommissioning Authority and Sellafield Limited focused on the historic, stored cans. Over a period of six years, dozens of samples of plutonium oxide powder with different characteristics were tested in the world's leading specialized laboratories at the National Nuclear Laboratory. Samples of different origins– and hence a different mix of plutonium isotopes and trace elements—were analyzed under atmospheres where both relative humidity and composition were varied in a controlled way.

The plutonium produced in the UK is unique, as it was produced from two different types of reactors. Magnox reactors were a creation of the UK's historic civil nuclear program. They produced plutonium isotopes less radioactive than the ones produced in the alternative Advanced Gas Reactors (AGR), which came in to use in the UK later. The isotopes were separated from the used fuel using different processes.

The AGR fuel was processed at the Thermal Oxide Reprocessing Plant, or THORP reprocessing plant, at Sellafield in Cumbria, UK. Once plutonium is separated from other elements in the used nuclear fuel, it is processed into an oxide powder, and the exact processing steps are different in the UK to those used by other nations.

The dynamic interactions inside the canisters are also unique. According to Dr. Orr, "You can't turn off the radiation coming from nuclear materials, so the plutonium and the environment inside the canister is changing even as you measure it. In the past, we've done controlled experiments on materials that are similar to plutonium oxide, which have provided some clues, but this isn't the same as working on plutonium itself. You really need to look at the real material."

Spotting trends

Since the environment inside the canister changes so quickly, the researchers looked for trends or patterns in the data to help narrow down what might be happening to suppress hydrogen production.

As the relative humidity decreased, so did the generation of hydrogen gas. Intuitively, this makes sense, as the amount of water will limit the amount of hydrogen that can be generated. At very low relative humidity, the water will be so strongly absorbed by the powder that it bonds to it, preventing it from escaping as a gas.

Samples that were in an atmosphere of normal air tended to produce more hydrogen than samples in an atmosphere of argon or nitrogen. This suggests that something about the mixture of oxygen, nitrogen, carbon dioxide, and trace gases in the air changes the chemical reactions caused by radiation.

Radiation, like the alpha particles given off by plutonium, can cause a cascade of reactions; even a simple molecule like water can undergo dozens of them. The energy from the radiation can be absorbed by matter and even transferred from one material to another.

The interface between the plutonium oxide powder and the water on its surface is very important. The powder has very tiny pores in it, small enough that molecules of water are more likely to bounce off the walls of the pores rather than collide with other water molecules. These interactions also affect the radiation chemistry.

Higher porosity means that the surface area of the powder is higher. Somewhat surprisingly, no clear correlation between surface area and hydrogen generation was seen, perhaps because of small differences in the composition of the powder affecting the surface chemistry.

Dr. Luke Jones worked on radiolysis of water during his prior research at The University of Manchester and is now a Senior Research Technologist for NNL. He explains the challenge of understanding the interface between the water and the powder, "There are many different variables to consider. Even when you use a non-radioactive surrogate for plutonium dioxide with an external radiation field that you can control, you still need to think about the purity of the sample as well as the microstructure of the powder."

"Small differences can have a big effect on the myriad of chemical reactions that happen when water is irradiated. When the water is confined in pores, it can affect how the water molecules and fragments created by radiation diffuse. If diffusion is restricted, then it can influence the rate of the reactions. Different chemical species sitting on the surface of the powder can also interact with the water fragments."

There was some variability in the data collected from the canisters. Samples derived from Magnox reactors (via Sellafield's Magnox Reprocessing Plant) tended to show less variability than samples that came from the THORP reprocessing route. Since the Magnox-derived plutonium contains isotopes that are less radioactive than those from the THORP processing route, it seems that the radiation dose rate influences the chemical reactions.

A lower dose means that the concentration of some products of the reactions would be lower, which in turn affects further reactions. This variability in the data suggests that the mechanisms affecting hydrogen production are incredibly sensitive to even small variations on the surface of the powder.

The trends also show that the type of atmosphere inside the canister also has a role to play in suppressing hydrogen generation; previously, it was thought that only the interaction at the surface of the powder was important. Dr. Orr explains the significance of these results, "Radiation chemistry depends on many factors. Our results show that even small changes to trace elements on the surface of the powder, the porosity of the powder, and the atmosphere in the canister all affect the myriad chemical reactions."

The results show that interim storage inside these specially designed canisters is the right choice; although there's a dynamic, changeable environment inside them, we can see that the changes are small.

Long-running research with international appeal

The UK has a proud nuclear heritage dating back to the 1950s when the world's first industrial scale nuclear power station was connected to the National Grid and started supplying the nation with carbon-neutral electricity. Radiation science has long been part of this heritage. The National Nuclear Laboratory's current program of research to uncover the dynamic process that takes place in stored plutonium began in 2011. The original work focused solely on gas production.

In collaboration with researchers in the United States, a significant body of information has been collected over the intervening years which demonstrates that the powder is safely stored. Since then, there has been a big shift in the research to uncover the fine details of the radiation chemistry. The data will be used to form theories and create specific experiments to test them.

The findings are published in the journal *Frontiers in Nuclear Engineering*.

More information: Kevin Webb et al, Effects of relative humidity, surface area and production route on hydrogen yields from water on the surface of plutonium dioxide, *Frontiers in Nuclear Engineering* (2023).

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