Testing coatings to protect canisters against corrosion

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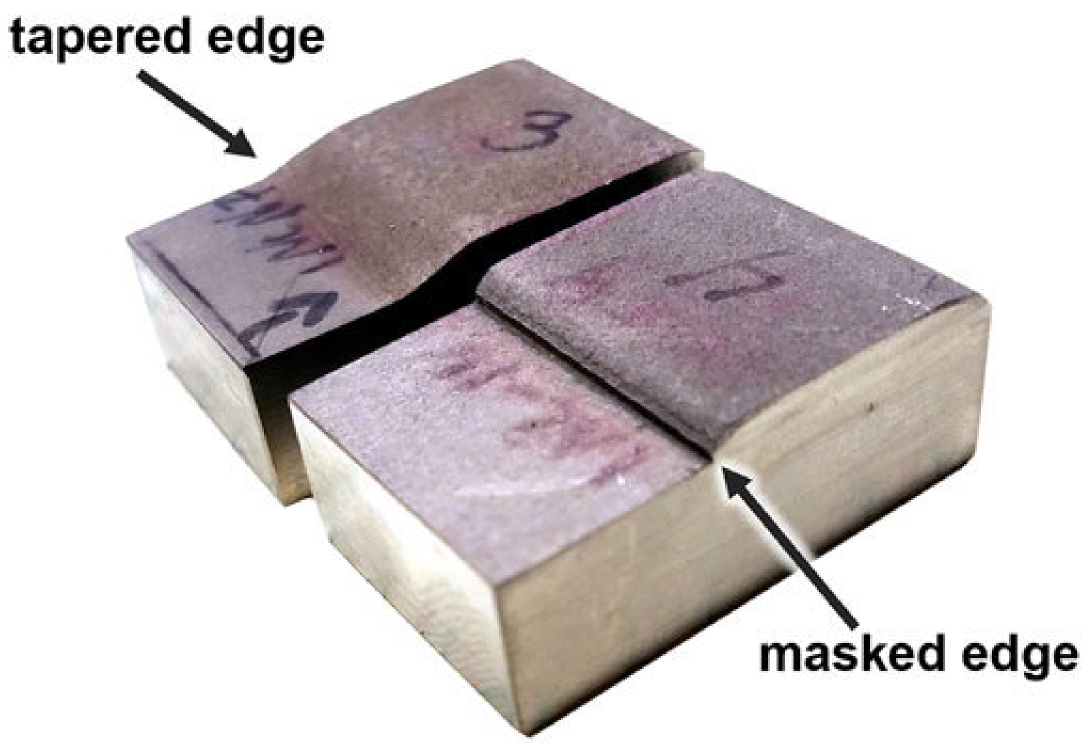

Cold spray samples with tapered and masked edges. Credit: *Frontiers in Metals and Alloys* (2022). DOI: 10.3389/ftmal.2022.1021000

As anyone who has lived near the ocean can attest, metal and sea mist are a recipe for corrosion. A nuisance of coastal life, the consequences of these common chemical reactions become far more serious when it is taking aim at the stainless-steel canisters that contain spent nuclear fuel.

To shield steel from the corrosive threats posed by sea air, Sandia National Laboratories researchers tested a variety of nickel mixtures as protective coatings on stainless steel. The researchers found that the specific material applied, and the specific application process used, impacted the properties of the coating, including how protective it was against corrosion. Their results were published recently in *Frontiers in Metals and Alloys*.

Spent nuclear fuel is stored in quite a few coastal areas, where sea breezes can buffet canisters and deposit corrosive chloride salts such as sodium chloride, or more commonly known as table salt. Given enough time, the brine formed by these salts can corrode and pit stainless-steel canisters.

"Through our research, it became clear that it would not be easy to completely eliminate the possibility of a type of corrosion known as stress corrosion cracking," said Charles Bryan, an expert on the storage of spent nuclear fuel and co-lead on the project.

"Stress corrosion cracking is likely to eventually occur at some interim storage sites. It might take hundreds of years, but it could happen, so people started thinking about mitigation and repair technologies. We started looking at cold spray, which is a technique industry is very interested in, and at corrosion-resistant polymer coatings."

Storing spent nuclear fuel

Nuclear fuel rods that no longer produce enough heat for a nuclear power plant—still the U.S.'s leading source of carbon-neutral electricity—are first transferred to a pool of water at the reactor site. After several years in the pool, the spent nuclear fuel, in a rack called a fuel assembly, is cool enough that it is removed from the storage pool and placed inside a stainless-steel canister with many other fuel assemblies.

Each fuel rod has solid uranium oxide pellets inside a zirconium-based tube. The solid zirconium can break down if exposed to oxygen and moist air, so the canisters are filled with helium, an inert gas, to protect the rods.

These dry storage canisters are highly radioactive, and often are surrounded by additional layers of concrete or steel called overpacks. These overpacks protect workers from radiation while allowing air to circulate around the canisters, cooling them off, Bryan said.

Originally the spent nuclear fuel dry storage canisters were licensed to store spent nuclear fuel for up to 20 years before being transported to a deep geologic repository for permanent disposal, Bryan said.

However, development of such a repository in the U.S. has proved politically challenging. Since the spent nuclear fuel dry storage canisters are remaining in use for longer than expected, in 2012 industry scientists and regulators began to explore potential issues facing aging canisters. They identified one type of corrosion called stress corrosion cracking as posing a potential risk, Bryan said.

Decoding stress corrosion cracking

Ten years ago, Sandia collaborators at the Electric Power Research Institute, working with nuclear power plant operators, began collecting samples of dust from the outside of dry storage canisters at a variety of sites, Bryan said. Sandia analyzed these samples and found chloride salts in every sample. Canisters stored near an ocean had a lot more salt, but even inland canisters had a bit of chloride from road de-icing salts and sodium chloride used to soften water for steam-based power generation, such as nuclear power plants.

Sandia researchers also carried out a large experiment to ascertain if the welds used to manufacture the dry storage canisters produced enough stress to allow stress corrosion cracking to occur, Bryan said. They found that the welds do produce enough stress.

Additionally, Sandia researchers were involved in developing computer models of the canister temperature, ambient humidity, and the chemical composition of the dust that impact formation and growth of corrosion pits in the steel. They also studied the process by which the pit becomes a crack and how long it takes for a crack to grow large enough to pose a risk. Refinement of these models is still ongoing, he added.

Over the past ten years, researchers at Sandia and other institutions have not been able to completely rule out stress corrosion cracking of dry storage canisters over extended canister lifetimes; however, canister inspections have never detected a crack, Bryan said.

Sandia engineer Sam Durbin is studying the possibility of radioactive materials coming out of potential stress corrosion cracks, which is key for evaluating the possible radioactive exposure risk to the general public, Bryan said. Instead, the primary concern with canister stress corrosion cracking is degradation of the fuel rods, and possibly exposing workers to radioactive material if they repackage the spent fuel for permanent geologic disposal, he added.

Studying cold spray protections

Researchers at Sandia and Pacific Northwest National Laboratory started exploring crack mitigation and repair technologies three years ago. These researchers, including engineer Erin Karasz, who started the project as a postdoctoral appointee and is now staff, have tested a variety of cold spray coatings to see if they could protect 1/2-inch-thick pieces of stainless steel from chloride corrosion.

"We found that you have to be very cognizant of the kind of material you are spraying onto what other kind of material," said Karasz, who is the lead author of the Frontiers in Metals and Alloys paper. "I was surprised at how much the porosity determined the behavior when corrosion got going in between the cold spray coating and the steel. There seems to be a specific level of porosity, below which the cold spray has enhanced corrosion resistance."

Cold spray is a process of taking small metal particles, about as wide as half of a human hair, and spraying them onto a surface using gas hotter than a commercial pizza oven, but much cooler than the temperature needed to melt metal, Karasz said. The inert gas "splats" the small metal particles onto the stainless steel using pressures 10 to 20 times higher than the pressure of a car tire.

For this paper, the team tested three different nickel-based metal mixtures, two with known anti-corrosive properties and pure nickel as a comparison. They tried two different gases, nitrogen and helium. And they tested the effect of tapering off the coating on the metal or leaving a sharper edge between the coated area and the uncoated area.

They found that the gas used to spray on the metal particles had a strong impact on how porous, or spongy, the coating was. The porosity of the coating greatly impacted the corrosion behavior of the coating, Karasz said.

To test the corrosion protection of the cold spray coating, Karasz soaked the small pieces of cold-spray-coated stainless steel in a very corrosive ferric chloride bath for three days. This is a standard method to speed up chloride corrosion, but it isn't a perfect model of what would occur to spent nuclear fuel canisters over hundreds of years, said Rebecca Schaller, a materials scientist and co-lead on the project. They found corrosion on all the samples, but the location and shape of the corrosion differed, suggesting further refinement of the coatings is needed.

Now the team is moving on to testing stainless steel with cold spray as well as other polymer coatings under more relevant atmospheric conditions, Schaller said. Eventually, the goal is to test the coatings on welded stainless steel to test them with stress and corrosion, she added.

"When we put anything on these canisters, first and foremost we're trying to make sure it's not doing any harm," Schaller said. "What Erin was trying to establish is where we need to look at in these studies to optimize the coatings and ensure that they're not going to create more problems for us in the future. Cold spray is a newer technique and very few people have looked at it under atmospheric corrosive conditions, let alone its corrosion performance over hundreds of years."

More information: Erin K. Karasz et al, Accelerated corrosion testing of cold spray coatings on 304L in chloride environments, *Frontiers in Metals and Alloys* (2022). DOI: 10.3389/ftmal.2022.1021000

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