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Investigations of Stress Corrosion Cracking of Spent Fuel Dry Storage Canisters Used for Long-Term Storage

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1. ABSTRACT

The United States (US) Department of Energy (DOE) and the US Nuclear Regulatory Commission are working to develop a better understanding about the environment to which the inventory of dry storage canisters is exposed and how that might contribute to the corrosion performance of the canisters over time. The canisters are mostly made of 304 stainless steel that is susceptible to stress corrosion cracking (SCC), and canister penetration by SCC due to deliquescence of salts in dust on the canister surfaces is considered to be the most likely mode of canister failure. The DOE and the Electric Power Research Institute have teamed to sample and characterize dust deposited on the surface of in-service spent nuclear fuel storage canisters at three sites. While all three sites were near salt water or brackish water, the deposits collected from the tops and sides of the canisters show considerable variability based on the geographic location of the storage site. Samples from the sites more distant from the open ocean contained chloride-poor salt assemblages typical of continental dusts, while samples from the near-ocean sites were enriched in chloride-rich marine salts. Future work will evaluate how the composition and abundance of salt species affects corrosion of 304SS over time. Other tests will be performed to characterize the weld and manufacturing residual stresses in the canister shell; high through-wall tensile stresses are an important criterion for canister penetration by SCC. This work will improve understanding of canister performance through time as a function of the geographic location and the chemical environment on the canister surface.

2. INTRODUCTION

In the United States (US), spent nuclear fuel (SNF) is likely to remain in interim dry storage until a permanent disposal solution has been developed and placed into operation. Current US policy dictates that a permanent disposal solution should be available in 2048 [1]. Until a permanent repository is opened, SNF will accumulate and remain in dry storage for longer than originally planned. There are currently about 70,000 metric tons of commercial used fuel in storage in the United States, of which 80% is in used fuel pools and about 20% is in dry cask storage systems. This amount increases by about 2000 metric tons annually [2]. As illustrated in Figure 1, at the end of 2013, the US had SNF stored in 1850 dry storage casks, located at over 60 different sites across the US [3]. Commercial spent nuclear fuel is stored throughout the US at both coastal and inland locations (Figure 1).

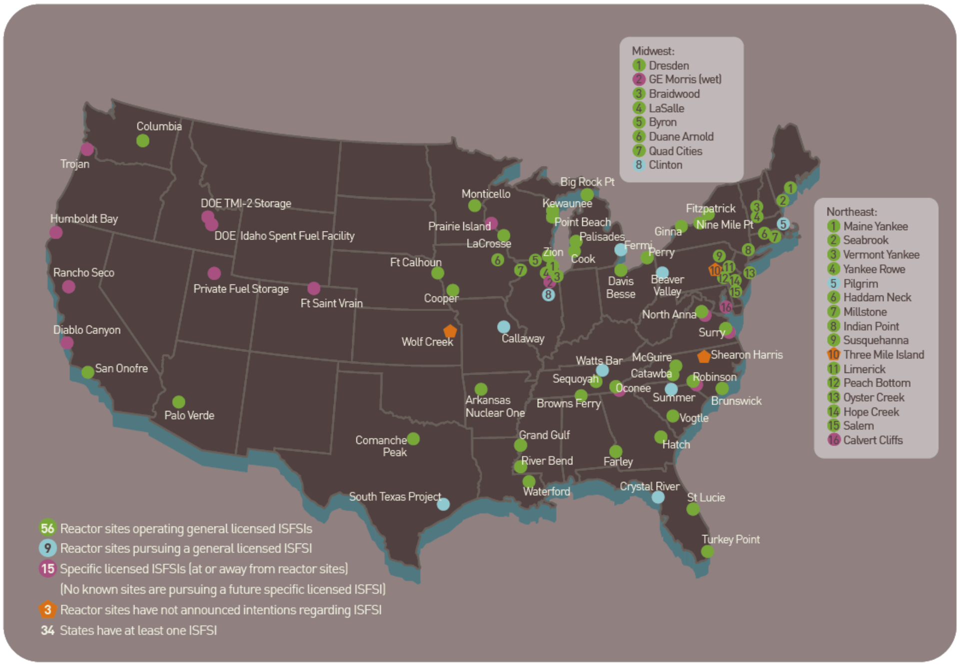

Figure 1. US Independent Spent Fuel Storage Installations.

http://www.nrc.gov/waste/spent-fuel-storage.html. downloaded 3/18/2015

In the US, the predominant storage system consists of a welded austenitic stainless steel (typically 304) container. This container represents the primary confinement boundary for the spent fuel. The containers are stored either vertically or horizontally within an overpack, and their role is to provide neutron shielding, physical protection, and passive cooling for the spent fuel container. As seen in Figure 2 below, the canisters are housed in concrete or steel overpacks, which have space between the stainless steel canister and the overpack to allow for passive cooling by convective air flow. Air enters the overpack via inlet vents near the bottom and exits through outlet vents near the top. These vents allow for continuous convective cooling using ambient air to remove heat from the system. This fail-safe, zero energy, convective cooling has the disadvantage of allowing collection of dust, salts, and other air contaminants to contact and interact with the stainless steel canister.

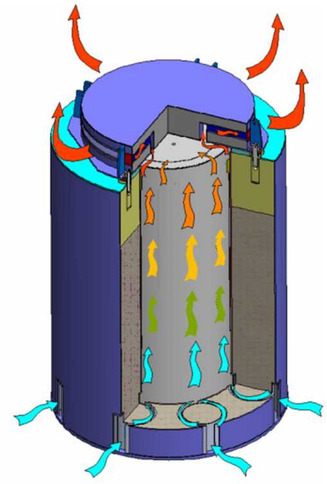

Figure 2. Diagram of typical horizontal (left) and vertical (right) dry storage canister systems. Printed with permission from Holtec International.

Prolonged exposure to the unfiltered air stream described above will result in the deposition of atmospheric dust on the container surface. A portion of this dust will likely be chloride rich salts, potentially creating an environment conducive to stress corrosion cracking of austenitic stainless steels. As a result, stress corrosion cracking (SCC) of interim storage containers has been indicated as a high priority data gap by DOE [4], [5], [6], [7]. The US Department of Energy (DOE) and the US Nuclear Regulatory Commission (NRC) are evaluating the risk for SCC in the US dry storage canister inventory. This paper summarizes work done to characterize the canister material properties that affect susceptibility to SCC (e.g., weld residual stresses), and to understand the diversity and composition of the dust deposited on the canisters at different geographic locations. This paper also summarizes future testing funded by the DOE in this area.

3. STRESS CORROSION CRACKING

The US DOE and NRC are evaluating potential failure mechanisms for stainless steel canisters during long term dry storage. Stress corrosion cracking is a plausible degradation mechanism for the container with the potential to result in a penetration, which typically initiates in areas of localized corrosion (e.g., pitting). Verification of the presence of SCC requires that a detailed inspection of the container surface be performed. To date, no such detailed inspections have been conducted. There is a series of factors that make implementing such inspections difficult. First, in-situ inspections of a canister within its overpack are difficult due to limited entry points and narrow annuli between the two. Second, high radiation levels exist at the surface of the container, hindering the extraction of the canister from the overpack followed by the implementation of traditional SCC inspection techniques. These factors, coupled with the inherent difficulty in detecting small stress corrosion cracks, have prevented the detailed surveillance of fielded spent fuel canisters for the presence of SCC. Because of these limitations and the lack of data, the US DOE Used Fuel Disposition (UFD) Program is currently funding work to understand the mechanisms for SCC and the anticipated environmental conditions on canisters in the current inventory.

Over time, the passive cooling system described above will result in the deposition of atmospheric dusts on the surface of the canister. A portion of these dusts will be salts which may deliquesce, potentially forming an aqueous brine on the canister surface. As austenitic stainless steels are particularly susceptible to SCC in chloride bearing brines, there is a risk of cracking in situations where this brine layer is chloride rich. Preliminary data indicates a difference in the depositions in “marine” and “near-marine” environments, which differ in terms of atmospheric salt compositions and concentrations. The dust deposited in marine environments will likely contain sea water (or brackish water) aerosols that are likely to be high in chloride.

In order for stress corrosion cracking to occur, three conditions must be met, as shown in Figure 3. There must be

1. A susceptible material: Austenitic stainless steels such as 304SS, are highly susceptible to SCC in chloride bearing environments. This susceptibility can be exacerbated due to microstructural changes that accompany high heat input processes, such as welding. Results of some of these studies will be discussed in this paper.
2. An aggressive environment: Samples of the dust and salts from the surface of in-service canisters at marine and near-marine locations have been collected. In addition, canister surface temperatures have been collected. The results of these analyses will be discussed in this paper.
3. Sufficient tensile stress: A full- diameter scale cylindrical mock-up has been manufactured and residual stresses near the welds and heat affected zones will be measured. The plan for this testing will be summarized in this paper.

Figure 3. For stress corrosion cracking to occur, you must have the right material, environment, and stress.

3.1.Susceptible Material

As noted previously, there is ample evidence, both from industry and from experimental studies, that 304 SS is susceptible to SCC under atmospheric conditions if chloride-containing salts are present [10].

3.2.Aggressive Environment

Since it has been confirmed that SS304 is susceptible to localized corrosion and SCC, the next step was to investigate if currently in-service canisters could be exposed to a corrosive environment. Sandia National Laboratories and the Electric Power Research Institute (EPRI) have partnered to sample and analyze dust deposited on in-service storage canisters to increase the data set and knowledge of the environments on the dry storage canisters.

3.2.1. Storage Container Environmental Conditions

Because the canisters are shielded from the weather, the only mechanism for corrosive conditions to occur on the canister surface is by deliquescence of salts that settle on the metal canister from air convecting through the overpack. Deliquescence can occur when a critical combination of temperature and absolute humidity are achieved at the container surface, the magnitude of which is a function of the salts that are present. The relative humidity is controlled by the local temperature on the canister surface and the absolute humidity in the air entering the overpack, and the threshold value for deliquescence is a function of the salt composition present. Because of the importance of canister surface temperature, the US DOE has been funding work to better understand the surface temperatures of dry storage canisters with different fuel heat loadings and how those temperatures change with age, outside temperature fluctuations, and at different locations on the canister [9]. The temperature on the canister surface will vary with position and is a function of the thermal output of the fuel within the canister, the temperature of the air stream being pulled into the overpack to cool the canister, and the location of the surface within the overpack itself. This is demonstrated in Figure 4 which presents a model of the surface temperature on a horizontal storage canister located at the Calvert Cliffs Independent Spent Fuel Storage Installation (ISFSI) [9].

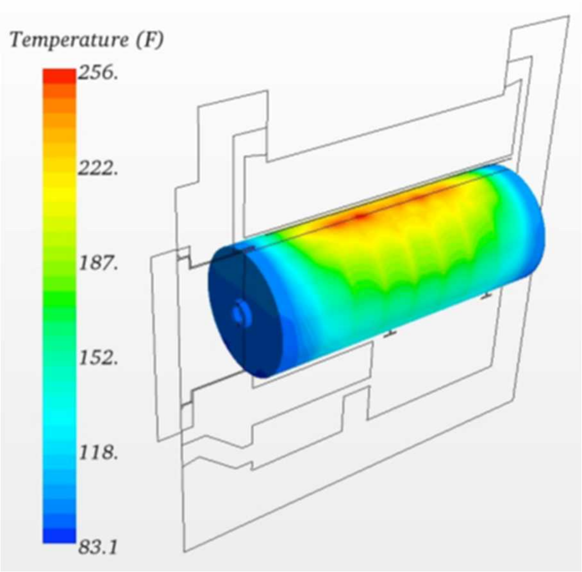

Figure 4. Example of Canister Temperature Model Results for a Vertical Canister Stored in a Concrete Housing [9].

3.2.2. In-Service Samples

From 2012 to 2014, dust samples were taken from the tops and sides of canisters at three different ISFSI sites. The first was Calvert Cliffs, which is located in the state of Maryland on the Chesapeake Bay, a brackish water site with little wave action. This facility utilizes horizontal storage systems. The second site sampled was Hope Creek, which is located in the state of New Jersey, 0.25 miles from the tidal Delaware River. This is also a brackish water body with little wave action. The final site that has been sampled was Diablo Canyon, which is located 0.3 miles from the California coast near a rocky coastline with constantly breaking ocean waves. Both Hope Creek and Diablo Canyon use vertical dry storage systems. Both the design of the sampling systems and the acquisition of dust samples were performed by a combination of personnel from each reactor site along with a contingent from the vendor that supplied each storage system.

Dust samples were taken from two canisters at each of the three sites. At each location, two types of samples were collected. Dry samples of the dust were collected by using an abrasive Scotch-Brite™ pad; in the case of Calvert Cliffs, the pad was backed by filter paper and connected to a vacuum line (see Figure 5 for an illustration); while at the other two sites, the pad was simply backed with a steel coupon, and the dust collected via static attraction to the Scotch-Brite™ pad. Neither of the dry sampling techniques was characterized for their extraction efficiency.

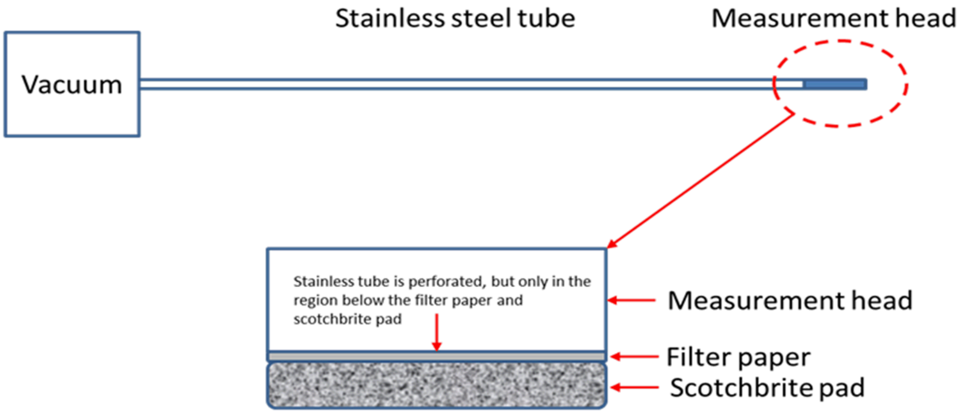

Figure 5. Schematic of the Dry Sampling tool used to collect dust on canister surfaces. (Diagram credit: D. Enos)

The second sampling method was a wet method, which used a SaltSmart™ sampling device to collect water-soluble surface contaminants, specifically soluble salts. The SaltSmart™ device quantitatively collects salts from a known surface area, allowing the chemical composition of the soluble salts to be measured. Following collection, the SaltSmart™ sensor was disassembled and the collected salts leached out for quantification. Figure 6 shows a disassembled Scotch-Brite™ sampling device. Sampling was done by hand, by inserting long probes through the open door of the overpack at Calvert Cliffs. At the Calvert Cliffs site, the door was removed and replaced with a shield plug that allowed access to the sides of the container. Unfortunately, there was not sufficient space to allow their tooling to deploy at the top of the container, hindering measurement. Therefore, only a single salt-smart measurement was obtained. Learning from this experience, at the other two sites, sampling tools were inserted through the outlet vents and directed down into the annulus between the canister and the overpack. Following insertion into the overpack, the samplers were pneumatically pushed onto the canister surface in order to ensure good contact. Although the SaltSmart™ sensors could quantitatively measure salt loads if better contact was made, obtaining constant contact was difficult. Moreover, the sensors did not operate as reliably at temperatures over 80ºC, limiting their use, especially at the Diablo Canyon site. Therefore, the Scotch-Brite™ pads were used to extract dust samples for mineralogical characterization, but could not be used estimate salt loads or surface concentrations, because the efficiency of collection is unknown, and because the actual area sampled is unknown.

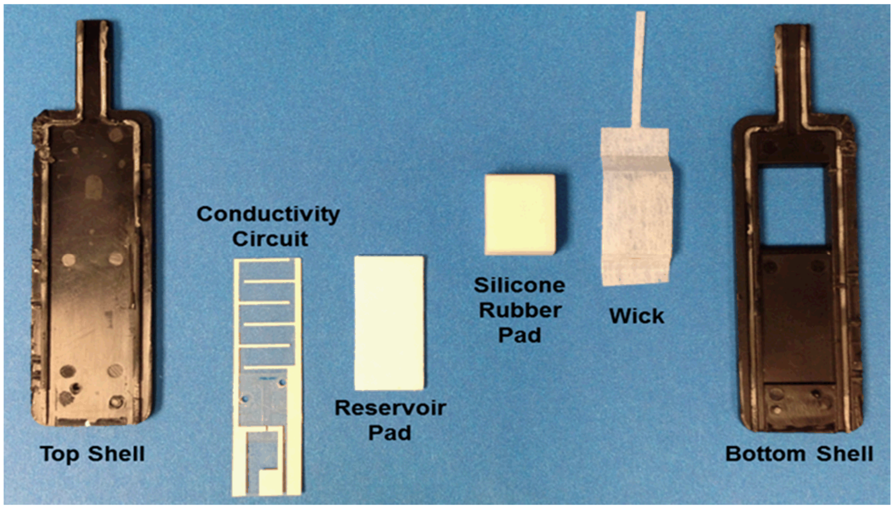

Figure 6. Wet sampling device taken apart to show the inner workings. This device is approximately the size of a pack of gum.

Samples were analyzed using several techniques. Dry pad samples were characterized using Scanning Electron Microscopy to identify organic matter and dust and salt mineralogy, and by X-ray fluorescence analysis to obtain semi-quantitative chemical analysis and element ratios for chemicals heavier than sodium. Finally, leachate from the Scotch-Brite™ and SaltSmart™ pads was analyzed to determine the chemical composition of the soluble salts present.

3.2.3. In-service Dust Analysis Results

At all sites, there was a greater concentration of dust on the tops of the canisters and less on the sides. Dust was collected from all sampled locations on the canister surfaces and consisted largely of aluminosilicates and quartz, and metal particles (stainless steel and iron) which are presumed to be generated from the canister manufacturing process. The soluble salts present on the canisters differed from storage site to storage site, and the compositions can mostly be differentiated on the basis of marine and near-marine locations.

3.2.4. Near-Marine Sites: Calvert Cliffs and Hope Creek

These sites were adjacent to brackish, calm water with prevailing inland winds. Dust on the vertical sides of the canisters was very sparse, with the top surfaces having a much heavier dust load. The composition of the dust was similar at the two sites; consisting mostly of quartz, feldspar and clays. Sulfates and nitrates dominated the soluble salts, with $CaSO_4$ being especially common. At both sites, NaCl was the only chloride salt present, and was observed as rare, isolated particles

The dust and salt analyses from Calvert Cliffs also revealed little in the way of chloride salts (low Na^+ and Cl^-, high Ca^2 and SO^2_4) [8]. Insoluble dust components were largely detrital quartz and aluminosilicates (feldspars and clays). The Hope Creek soluble salts were mostly sulfates and nitrates, mostly of calcium. As with Hope Creek, there was very little chloride found [10].

3.2.5. Marine Sites: Diablo Canyon

Diablo Canyon is located on the Pacific coast, and the dry storage site is one-half kilometer from breaking waves. At Diablo Canyon, sea-salt aggregates consisting mostly of NaCl and $Mg\text{-}SO_4$ with a little potassium and calcium, were common dust components. The observed dust and salt loads on the dry pads from the vertical canister sides were light, and the SaltSmart™ sensors measured low levels of chloride; however, high canister surface temperatures may have affected the performance of the SaltSmart™ devices, and the results may be questionable. Heavier salt loads were observed on the dry pads from the tops of the Diablo Canyon canisters, but because the tops were very hot (above 100ºC), SaltSmart™ measurements were not taken, and salt loads cannot be estimated. Detailed sampling results are documented in [10]. The abundance of chloride-rich sea-salts in the dust indicates that upon deliquescence, an environment that will support chloride-induced stress corrosion cracking will be present.

3.3.Residual Stress

The third requirement for the initiation and propagation of a stress corrosion crack is the presence of a sufficiently large tensile stress. Based upon modeling, such stresses are believed to be present through the canister wall at welded regions. Stress distributions in actual storage canisters have never been measured, so to obtain data on weld residual stresses, a full-diameter canister mock-up has been made using the same materials, welding procedures, and manufacturer as a frequently-used commercial storage canister. The canister has an outer diameter of 1.7 meters, a wall thickness of 1.58 cm, and an overall length of 3.66 meters. The parent material is 304SS and the weld filler material is 308SS. Because we were more interested in residual stresses in the longitudinal and circumferential welds (where penetration could result in local penetration of the cask wall), a lid and closure welds were not fabricated. The base plate was also excluded, for cost and logistical reasons. The residual stresses will be measured at the circumferential and longitudinal welds and weld-repairs, heat affected zones, and in the base metal of the canister remote from the welds. Techniques that will be used include incremental deep hole drilling, incremental center-hole drilling, contour mapping, and X-ray diffraction techniques. All data will be shared with the NRC and EPRI and published in a US DOE UFD report. After residual stress testing, the mock-up will be cut into pieces and shared with partner organizations for further stress and corrosion experiments and testing. The end goal is to understand what areas of the canister SNF might be most susceptible to chloride-induced stress corrosion cracking, and to characterize stresses and material properties in base metal, weld, and heat-affected zones. This information will be used to inform decisions for inspection intervals, to identify locations on the canister that might be of greatest risk, to provide realistic sample properties and tensile stresses for corrosion testing, and to validate existing models for weld residual stress estimation.

4. CONCLUSION AND NEXT STEPS

Chloride induced stress corrosion cracking resulting from the deliquescence of atmospherically deposited salts could potentially result in the local penetration of the canister wall in existing SNF dry storage systems. Therefore, the US NRC is developing protocols for inspections of individual canisters while in storage. The research and development being performed by the DOE UFD will be used to help determine the need for on-site canister inspections, potential inspection start times and inspection intervals, the geographic locations around the country at highest risk for SCC, and specific locations on the canister that might be more susceptible. This work will provide a greater understanding of how canisters will age over the current licensing times and beyond to better inform regulators about the performance of long-term interim storage and subsequent aging management.

The DOE UFD program continues to identify research needs and to develop testing programs to evaluate the risk of SNF storage canister failure by SCC. Current experimental work evaluates the effect of limited chloride surface loads on the rate and extent of localized corrosion (pitting and crevice corrosion), which is a precursor for SCC. Additional testing is planned that will evaluate pit and stress corrosion crack growth rates as a function of temperature, relative humidity, chloride surface loading, and crack depth.

Testing is also being carried out to understand the environment on storage canister surfaces. These tests evaluate the stability of different salts and brines on the heated surfaces of the storage canisters. Initial testing has been done with ammonium salts, which are the dominant components of inland salts, and which degas rapidly both in the solid state (prior to deliquescence), and after deliquescence. Additional testing will assess the stability of ammonium-bearing salts and brines containing NaCl or $MgCl_2$, a conditions which might occur if road salts aerosols were incidentally deposited on canisters at inland sites. Finally, the long-term stability of heated sea-salt assemblages will be evaluated. These tests will increase our understanding of how different salts are retained on the canister surface, how the salt/brine chemistry changes with time, and how particle-gas conversion reactions, acid degassing, and decomposition processes can potentially affect the corrosiveness of deliquescent brines on the metal surface.

Lastly, to obtain data on weld residual stresses, a full-diameter canister mock-up has been made and the residual stresses will be measured at the circumferential and longitudinal welds and weld-repairs, heat affected zones, and in the base metal of the canister remote from the welds.

5. ACKNOWLEDGEMENTS

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