

# Chloride-Induced Stress Corrosion Cracking of Austenitic Stainless Steel for Dry Storage of Spent Nuclear Fuel

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The NRC staff views expressed herein are preliminary and do not constitute a final judgment or determination of the matters addressed or of the acceptability of any licensing action that may be under consideration at the NRC.

This document is prepared, partly based on the recent presentation by the authors at the Extended Storage Collaboration Program (ESCP) Meeting.\*

Ahn, T., S. DePaula, J. Solis, T. Mintz, H. Jung, R. Pabalan, K. Lee and R. Einziger, "An Evaluation of Stress Corrosion Cracking (SCC) of Stainless Steel Canister in Marine Environment for Long-term Dry Storage of Spent Nuclear Fuel," The U.S. Nuclear Regulatory Commission (NRC) ADAMS, www.nrc.gov/reading-rm/adams.html - ML113350299, 2011



# Outline

- Background
- Dry Cask Storage System for Spent Nuclear Fuel
- Failure Mechanism Parameters
- Current Understanding of Canister Surface Conditions
  for SCC Initiation
- Potential Uncertainties Associated with Canister Surface Conditions
  - Salt Deposition Rate
  - Localized Humidity and Aqueous Chemistry
  - Salt Composition and Water Volume
  - Gamma Radiolysis
  - Test Method

#### Summary



### Background





- Various industries have recognized that austenitic stainless steels (304/304L or 316/316L) are susceptible to chlorideinduced stress corrosion cracking (SCC) under salt deposit conditions in the coastal area.
- The SNF dry casks have been stored at Independent Spent Fuel Storage Installations (ISFSIs) for roughly 20 years in the U.S.
- As passively cooled systems, some dry storage cask designs utilize a welded austenitic stainless steel canister (typically 304/304L or 316/316L) surrounded by a concrete shielding structure. The concrete structures have vents open to the outside atmosphere for cooling.
- Literature review identifies the primary parameters affecting potential SCC of the canister are temperature, RH, amount of salt deposits, and tensile stresses, at the canister surface.



## Dry Cask Storage System (DCSS) for Spent Nuclear Fuel



• Several vendor designs for DCSS in use, including vertically or horizontally oriented



### **Failure Mechanism Parameters**

- This presentation is intended to elicit expert feedback on parameters that may indicate susceptibility to this failure mechanism:
  - <u>Temperature and Relative Humidity (RH)</u>: Under what environmental conditions has chloride-induced SCC of austenitic stainless steel been observed? Temperature decreases and RH increases with time as radioactivity decays.
  - <u>Salt deposition</u>: What locations (i.e. distance from coast, highway, agricultural area) and sources of chlorides are of concern?
  - <u>Stress State associated with Fabrication/Weld</u>: How sensitive is this failure mechanism to the magnitude of canister tensile stress?</u>
  - <u>Potential Uncertainties</u>: Salt deposition rate, localized humidity and aqueous chemistry on the canister surface, salt composition, water volume, radiation effects, laboratory test methods.



# Current Understanding of Canister Surface Conditions for SCC Initiation



## **SCC Initiation Testing**

• Key Parameters: temperature, amount of salts deposited, relative humidity (RH), and residual stress, on the canister surface

• For SCC initiation, aqueous chloride concentration and stress need to be sufficient, generally with lower temperature, higher RH, more salts deposited, and higher stress. They are interrelated.

• Differences in testing methodologies make results difficult to compare.



# **SCC Initiation Testing (continued)**

100 -E-10 g/m3 Na<sub>2</sub>SO<sub>4</sub> Deliquescence 90 -<del>C 15</del> g/m3 -<u>A</u>-20 g/m3 <del>\*</del> 25 g/m3 80 NaCl Deliquescence 📥 30 g/m3 CaCl2.6H2O DRH 70 CaCl2.4H2O DRH 35 °C RH Range Relative Humidty, percent CaCl2 2H2O DRH 60 MaCl2.6H2O DRH NaCI DRH Na2SO4.10H20 DRH 50 Na2SO4 DRH 40 45 °C RH Range 30 VIgCI, Deliq 20 CaCL Delig 10 0 0 20 40 60 80 100 120 Temperature, °C

**NRC** Tests

• Type 304 SS U-bend specimens; as-received, sensitized, welded conditions; crack initiation at 0.1 g/m<sup>2</sup> ( $3.3x10^{-2}$  oz/ft<sup>3</sup>) surface salt concentration at 45 °C (113 °F)

(Mintz, T., L. Caseres, X. He, J. Dante, G. Oberson, D. Dunn, and T. Ahn, "Atmospheric Salt Fog Testing to Evaluate Chloride-Induced Stress Corrosion Cracking of Type 304 Stainless Steel," CORROSION 2012 Salt Lake City, Utah, March 11 – 15, 2012) **Japanese Tests** 



Yield stress was applied on specimens.

- Fracture mechanics specimens
- Three different types of stainless steel including type 304
- Measured as chloride amount rather than total salt amount

K. Shirai, J. Tani, T. Arai, M. Wataru, H. Takeda, and T. Saegusa, "SCC Evaluation of Multi-Purpose Canister," Proceedings of 2011 International Radioactive Waste Management Conference (IHLRWMC), Albuquerque, New Mexico, April 10-14, Paper No. 3333, 2011; copy right, August 31, 2012, by the American Nuclear Society, La Grange Park, Illinois



# Potential Uncertainties Associated with Canister Surface Conditions

- Many of these uncertainties associated with the key parameters are tied to the lack of observations on in-service canister and the difficulty in making those observations/measurements.
- The lack of field data applies to all four of the key parameters temperature, RH, stress and salt amount.
- Main challenge is not whether SCC can occur, but identifying if or when SCC parameters exist to initiate canister SCC.



# **Salt Deposition Rate**

• Rate of salt deposition on canister surfaces may depend on their orientation and proximity to predominant air flow paths.



A Schematic Deposition Rate for Various Air Flow Configurations. Vertical flow deposits more salts than horizontal flow.

(Wataru, M., K. Shirai, J. Tani, H. Takeda and T. Saegusa, "Sea Salt Deposition on the Canister Surface of Concrete Cask," ISSF2010, Central Research Institute of <sup>11</sup> Electric Power Industry [CRIEPI], November 16, 2010); copy right permission by Wataru



# Localized Humidity and Aqueous Chemistry

- Laboratory SCC initiation tests have used closed test chambers.
- In-service canisters may experience flowing air, creating non-equilibrium (dynamic) boundary layer on the canister surface as shown in Hot Plate of slide 11.
  - At the boundary there will be mass balance between convective flow and diffusion of water molecules in air toward and away from the canister surface.
  - The convective flow could be dominant (e.g., Wataru, et al.; Takeda, et al., 2008). This will potentially alter temperature and humidity inside the boundary, compared with static conditions assumed in most studies.
- Water ingress into the DCSS and the moisture content of the overpack concrete on the canister surface conditions are not well understood. Species from concrete degradation or other air species may modify aqueous chemistry related to the passivity.
- Other industry experiences highlight the importance of understanding local humidity effects in the observed SCC of austenitic type 304/316 stainless steel (Faraj, 2002; Barber, et al., 2001; Bush, et al., 1997)

(Wataru, M., H. Takeda, K. Shirai and T. Saegusa, "Thermal Hydraulic Analysis Compared with Tests of Full-Scale Concrete Casks," Nuclear Engineering and Design, Vol. 238, pp. 1213 – 1219, 2008);

(Takeda, et al., 2008)

(Barber, S. H., N. W. Sachs, and W. J. Salot, "Long-Term Leak Monitoring of Two Large process Vessels," Materials Performance, pp. 60 – 63, December, 2001) (Bush, S. H. and R. L. Dillon, "Stress Corrosion in Nuclear Systems," Unieux Firminy, France, June 12-16 1973, NACE, p 61, 1977) (Faraj, A. H., "External SCC Phenomena of Austenitic Stainless Steel in Urea Plants," Proceedings of CORROSION2002 Research Topical Symposium, Paper

No. 02439, Houston, Texas: NACE International, 2002)



# Salt Composition and Water Volume

• Equilibrium chloride concentration in solution decreases with increasing RH (i.e., more water volume); pitting (as SCC precursor) tendency decreases (Tsutsumi, et al., 2007).

• It is uncertain whether chloride dilution at high RH (more at lower temperature at longer time) could affect the evolution of SCC on dry storage canisters.





# **Gamma Radiolysis**

- A gamma radiation field may cause decomposition of water by radiolysis and nitric acid formation in air.
- Radiolysis rates are calculated, using data on gamma ray strength, amount of adsorbed water, and number of water molecules dissociated per 100 eV.
- Water evaporation rates are obtained from the measured rates in the pool.
- Evaporation rate and air flow rate appear to be faster than radiolysis rate: vapor replenishment on the salts will not allow accumulation of radiolysis products.

(Radulescu, G., "Radiation Transport Evaluations for Repository Science," ORNL/LTR-2011/294, Oak Ridge National Laboratory, 2011; dose rate values for a cross section through the center of the cask) (Arkhipov, O. P., A. O. Verkhovskaya, S. A. Kabakchi and A. N. Ermakov, "Development and Verification of a Mathematical Model of the Radiolysis of Water Vapor," Atomic Energy, Vol. 103, No. 5, pp. 870 – 874, 2007) (ThermExcel, http://www.thermexcel.com/english/propgram/pool.htm)



### **Test Method**

- Do laboratory conditions match field conditions? It is difficult to judge unless we know field conditions.
- Test methods among various options need to be carefully selected to simulate actual storage conditions of SNF canisters.
  - Heating Methods: RH on the sample surfaces vary.
    - (i) sample heating; chamber heating
    - (ii) static air; dynamic (flow) air; water or radiolysis present
  - Cracking Methods: strain applied may be different. U bend; fracture mechanics; un-notched tensile
  - Crack Initiation:

pit; flaw; others (e.g., intergranular attack)

- Deliquescence RH: difficult to determine experimentally
- Time Effect: longer time induces more cracks



### Summary

- Dominating parameters for SCC initiation obtained from national and international tests are:
  - Temperature and Relative Humidity
  - Salt deposition
  - Fabrication/weld residual stress
- Potential uncertainties include: salt deposition rate, localized humidity and aqueous chemistry on the canister surface, salt composition, water volume, radiation effect, and laboratory test methods.
- Optimum values of the primary parameters for SCC initiation appear to be interdependent