

# Spent Fuel and Waste Science and Technology

SAND2018-5875C

## Research into Stress Corrosion Cracking of Spent Nuclear Fuel Dry Storage Canisters

**Charles Bryan**  
**Sandia National Laboratories**

***National Transportation Stakeholders Forum***  
***Omaha, Nebraska***

**June 6, 2018**

# Spent Fuel and Waste Science and Technology

## Outline

---

- **Background: Long term interim storage of SNF**
- **Dry storage system designs**
- **Criteria for stress corrosion cracking (SCC)**
  - Susceptible material
  - High tensile stresses
  - Aggressive chemical environment
- **Evaluating the risk: A timeline for possible canister failure by SCC**
  - Corrosion incubation
  - Pitting and pit-to-crack transition
  - Crack growth and canister penetration

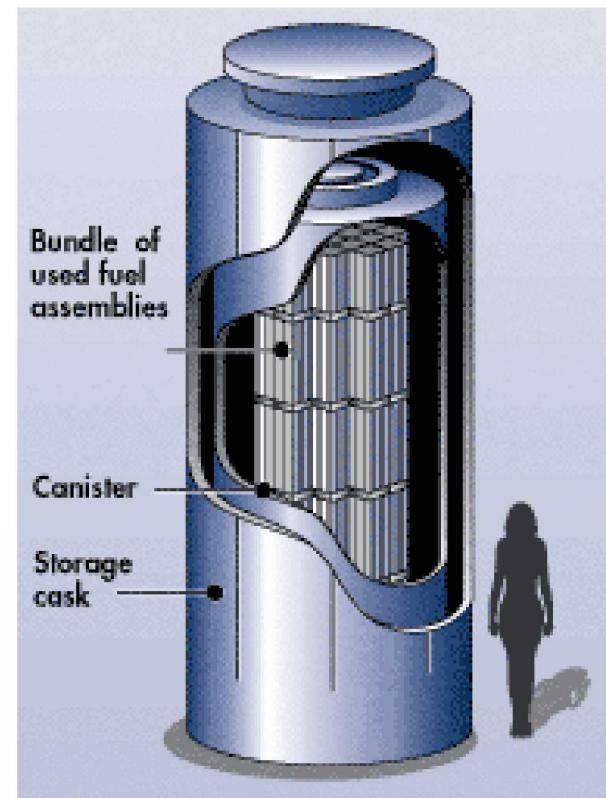
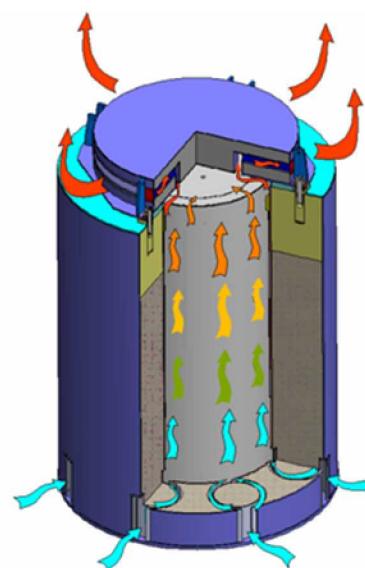
# Spent Fuel and Waste Science and Technology Background

---

- United States currently has over 80,000 metric tons of Spent Nuclear Fuel (SNF), about 30% in dry storage systems (about 2700 canisters total).
- The dry storage systems are intended as interim storage until a permanent disposal site is developed. Systems were licensed for 20-40 years with an additional 20 to 40 year renewal.
- However, the United States currently does not have a disposal pathway for SNF. It is likely that some SNF will be in storage for decades beyond the original storage system specifications.
- In most systems, SNF is stored in stainless steel (304 SS) canisters, which, given longer-than-intended storage times, could potentially undergo Stress Corrosion Cracking (SCC) due to deliquescence of chloride-rich salts in dust on the canister surface.
- Efforts to better understand the potential effects of SCC, and to predict timing and conditions of occurrence, are being pursued by the DOE and others.

# Canistered SNF Dry Storage Systems—Two Standard Designs

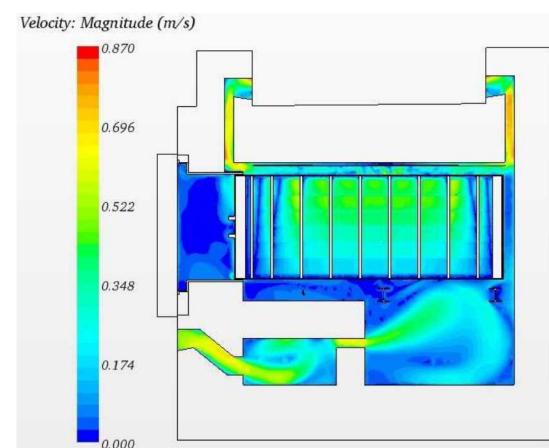
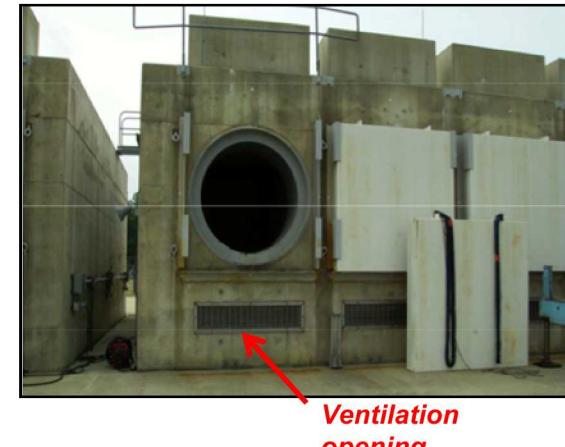
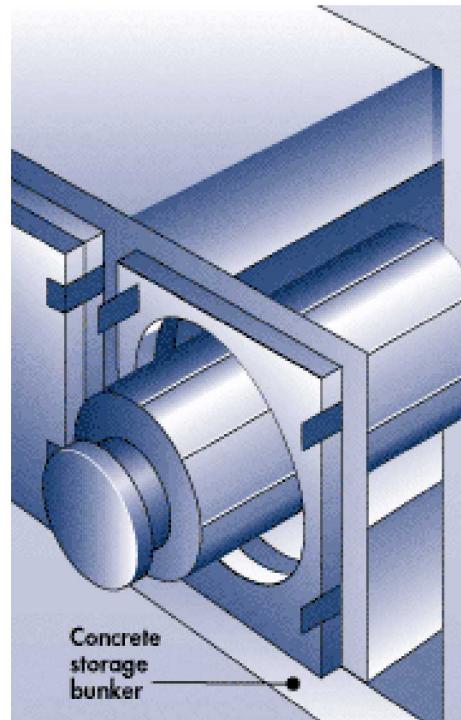
Vertical—In vertical systems, the welded stainless steel canister sits upright within a steel-lined concrete overpack. The canister is passively cooled by air that enters through inlets at the bottom of the overpack and exits through vents near the top.



Pathway for air flow  
through the  
overpack.

# Canistered SNF Dry Storage Systems—Two Standard Designs

Horizontal—In horizontal systems, the welded canister rests on its side upon rails within a concrete vault. Air enters the overpack through a vent in the base (below the door), flows up and around the canister, and exits through vents on the roof.



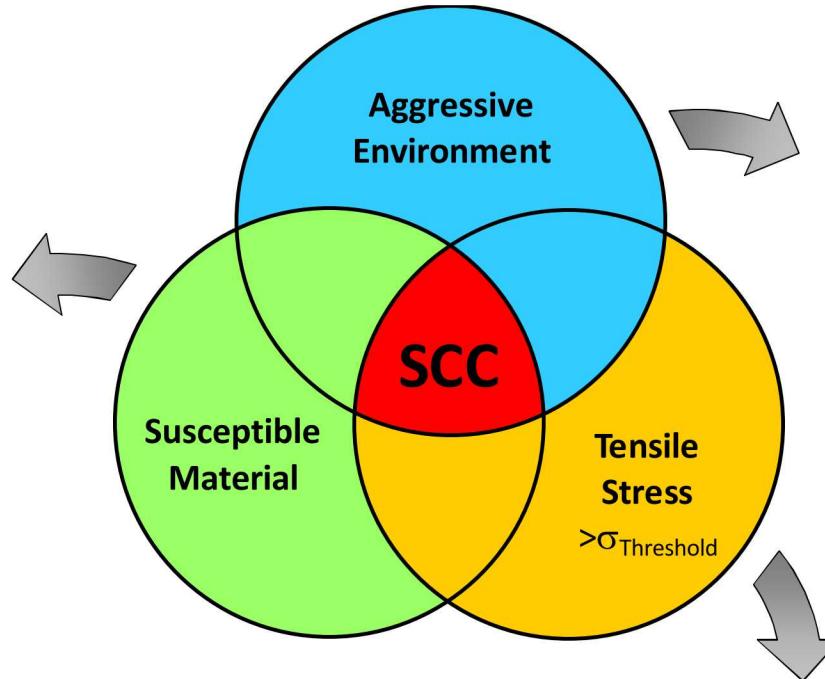
FCRD-UFD-2012-000114  
Figure 7.3

# Criteria for Stress Corrosion Cracking

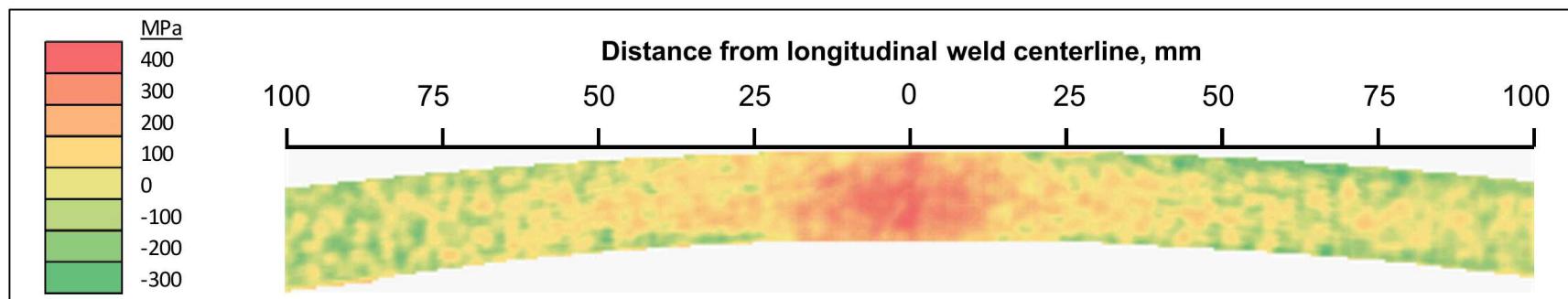
For SCC to Occur, All Three Criteria Must be Met



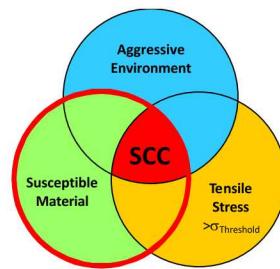
Weld zone, Ranor  
304 SS plate



Dust on canister surface at  
Calvert Cliffs (EPRI 2014)



Measured weld residual stresses (SNL 2016)

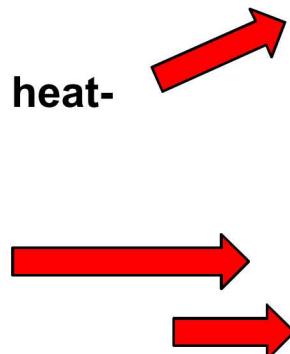


- Most canisters are 304 SS, known to be susceptible to atmospheric SCC. Susceptibility is a function of many factors:
- Degree of sensitization, in weld heat-affected zones (HAZ)
- Degree of cold work
- Surface finish
- Presence of iron contamination

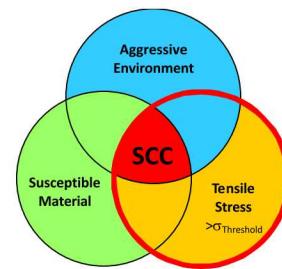
Experimental testing and operational experience has shown that SCC can initiate rapidly in 304 SS if the other criteria are met.



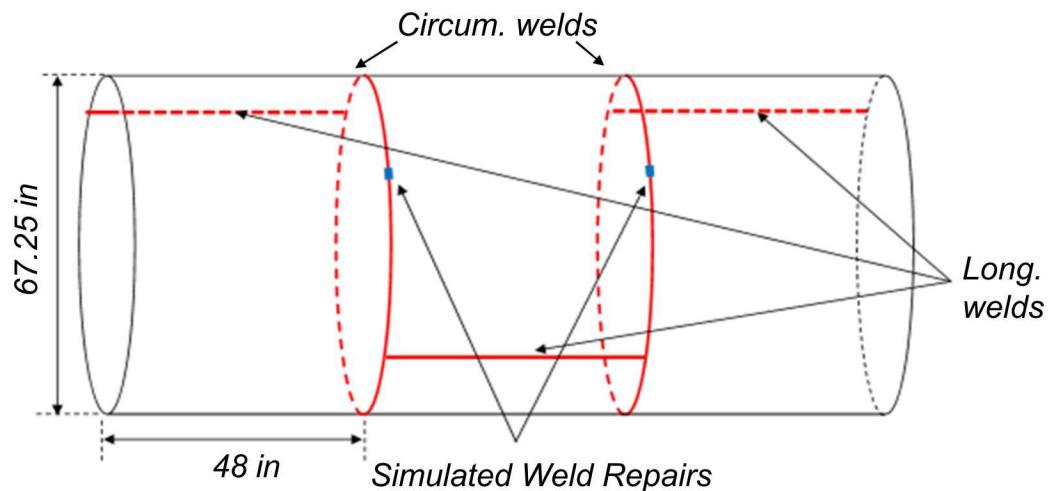
Weld zone, Ranor  
304 SS plate



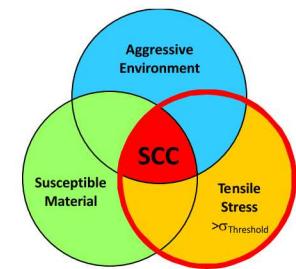
Rust spots on storage canister at Calvert Cliffs



- NRC modeling (NRC 2012) suggested high tensile stresses in welds and heat-affected zones (HAZ)
- Sandia full-diameter cylindrical mockup: experimental evaluation of WRS
  - Made by Ranor (old supplier to Transnuclear) using identical procedures to in-service NUHOMS canisters
  - 5/8" thick, 304/304L SS, 67.25" diameter
  - Mockup contained both circumferential and longitudinal welds, and two simulated weld repairs.
  - Stresses measured in base metal, all weld types, and associated HAZ



# Tensile Stresses Sufficient to Support SCC?



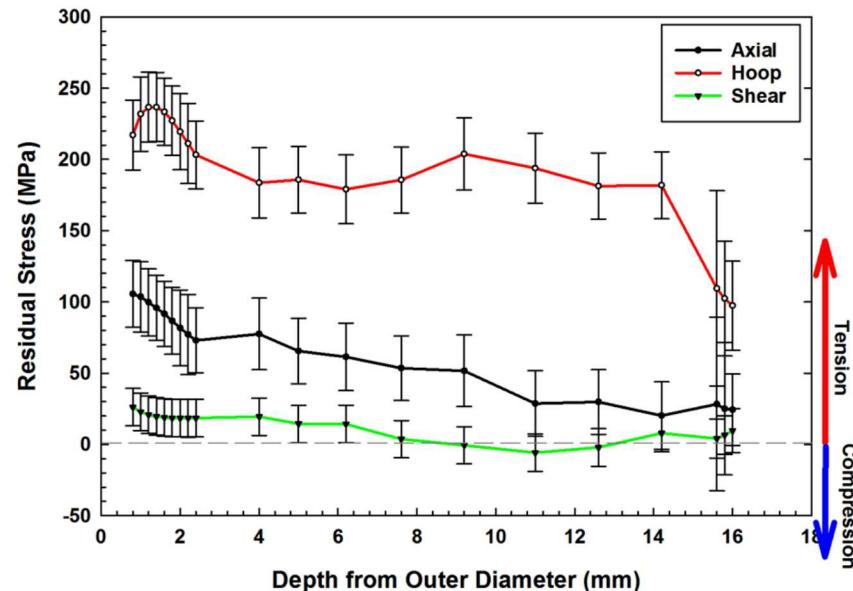
## WRS measurement results

- DHD and contour mapping results are consistent.
- High *through-wall* tensile stresses measured in all weld types and in all HAZ. Highest tensile stresses are parallel to welds, but tensile stresses also occur perpendicular to welds.
- Highest tensile stresses measured at simulated weld repairs.

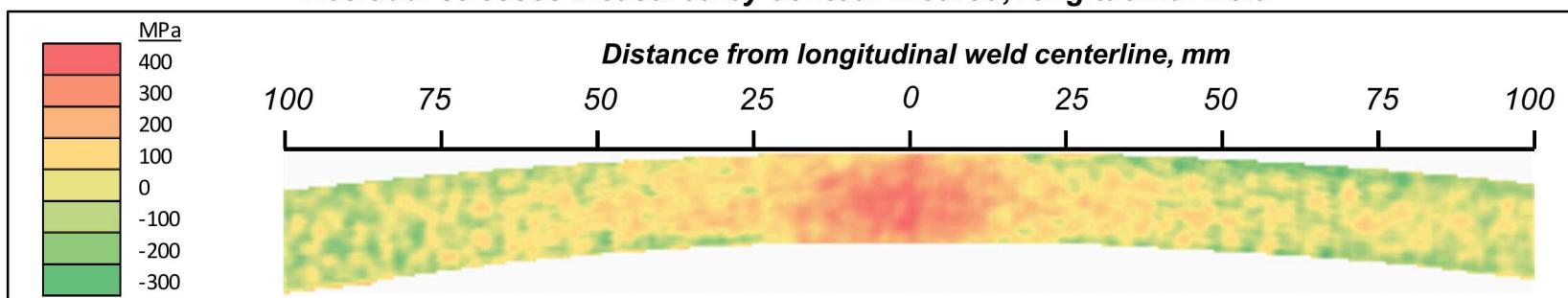
Conclusion: Sufficient tensile stresses occur in weld regions to support through-wall SCC

Enos D. and Bryan C., 2016. *Final Report: Characterization of Canister Mockup Weld Residual Stresses*, FCRD-UFD-2016-000064, U.S. DOE.

*Residual stresses measured by deep-hole drilling (DHD) method, circumferential weld HAZ*

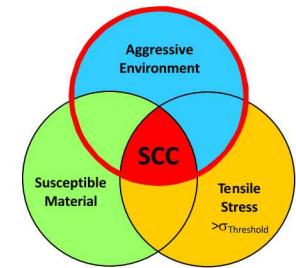


*Residual stresses measured by contour method, longitudinal weld*



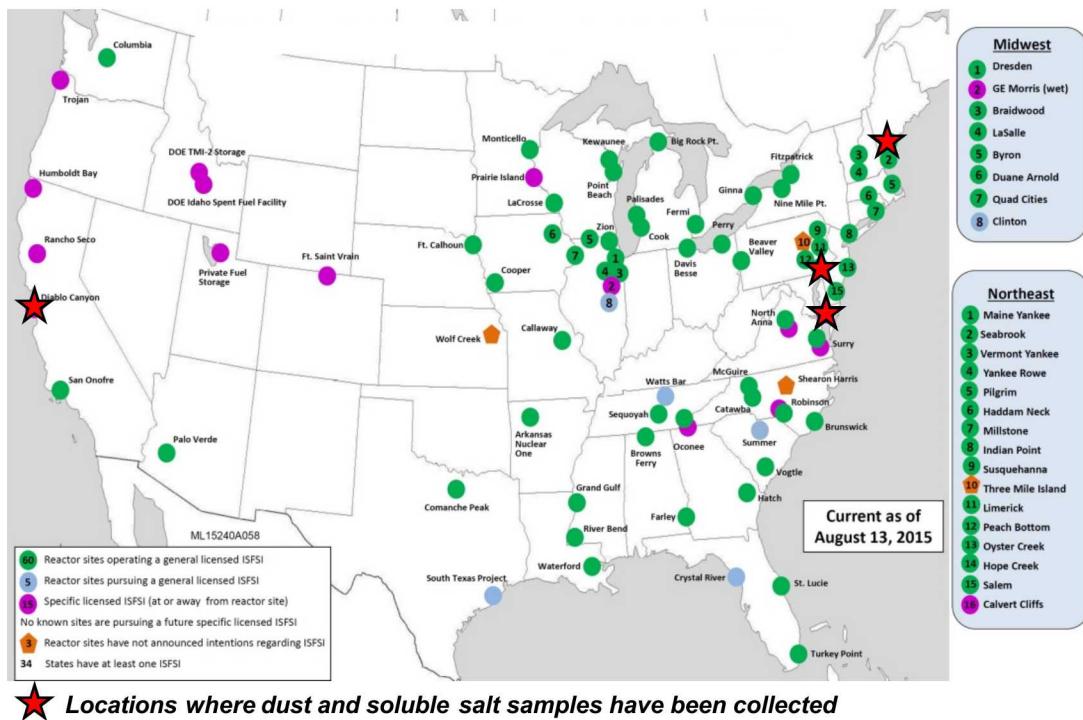
# Spent Fuel and Waste Science and Technology

# Canister Surface Environment

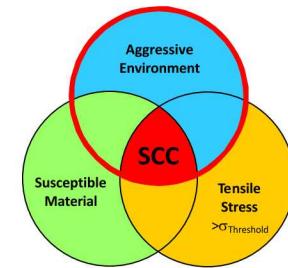


- Many ISFSIs are at coastal sites; anticipated deposition of chloride-rich sea-salts, which could eventually form corrosive brines.
- Samples have been collected from in-service storage canisters at coastal 4 sites to assess dust compositions, with emphasis on deliquescent salts ISFSI locations sampled:
  - Calvert Cliffs, MD
  - Hope Creek, NJ
  - Diablo Canyon, CA
  - Maine Yankee, ME
- Samples delivered to Sandia National Labs for analysis. Soluble salts leached and analyzed, dust characterized by SEM.

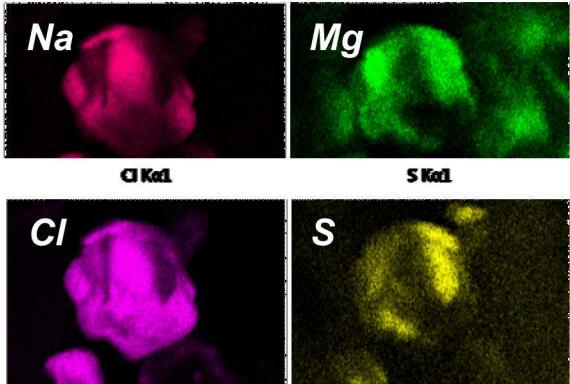
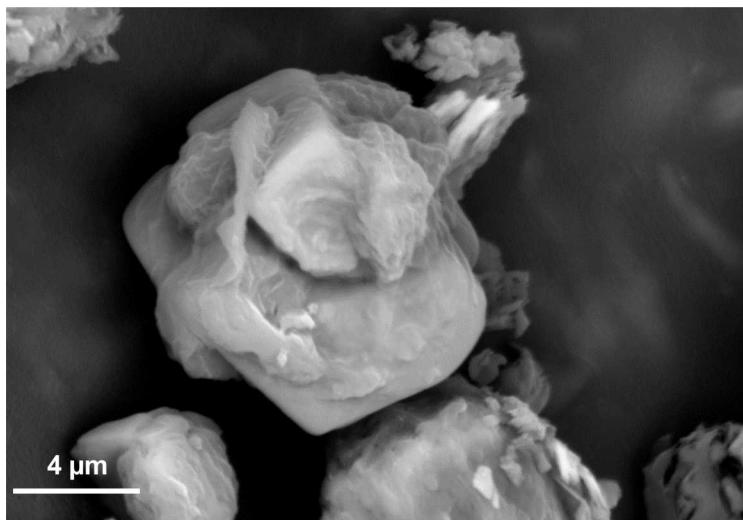
## Locations of U.S. Spent Nuclear Fuel Independent Storage Installations (ISFSIs)



Near-marine sites are considered to especially at risk because of potentially high concentrations of chloride-rich sea-salt aerosols



Sea-Salt Aggregate Collected from  
Diablo Canyon ISFSI Storage Canister



### Diablo Canyon ISFSI

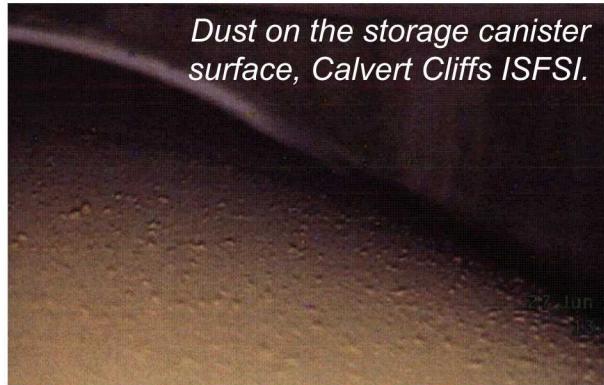
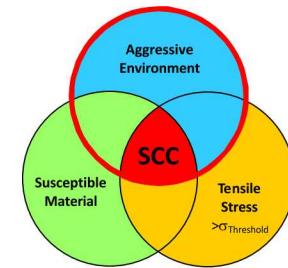
Dust is mostly mineral grains, but soluble salts particles are common, and are mostly chlorides.



Western U.S.



ISFSI  
Located  
~0.35 miles  
from the  
shoreline

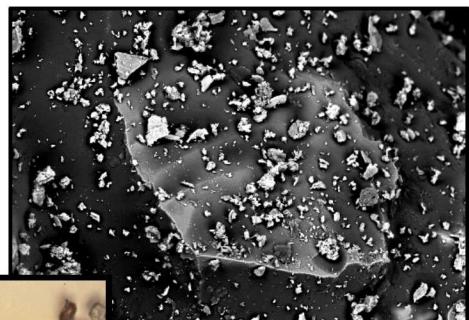


*Dust on the storage canister surface, Calvert Cliffs ISFSI.*

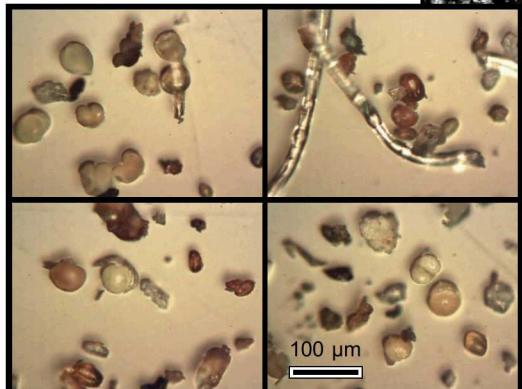
## Calvert Cliffs ISFSI

Dust is dominantly mineral grains and pollen. Most soluble salts are sulfates, nitrates; only minor chloride.

Eastern U.S.



Dust particles collected from the surface of the storage canister, Calvert Cliffs ISFSI.



*Pollen grains in dust from the upper surface of the canister.*

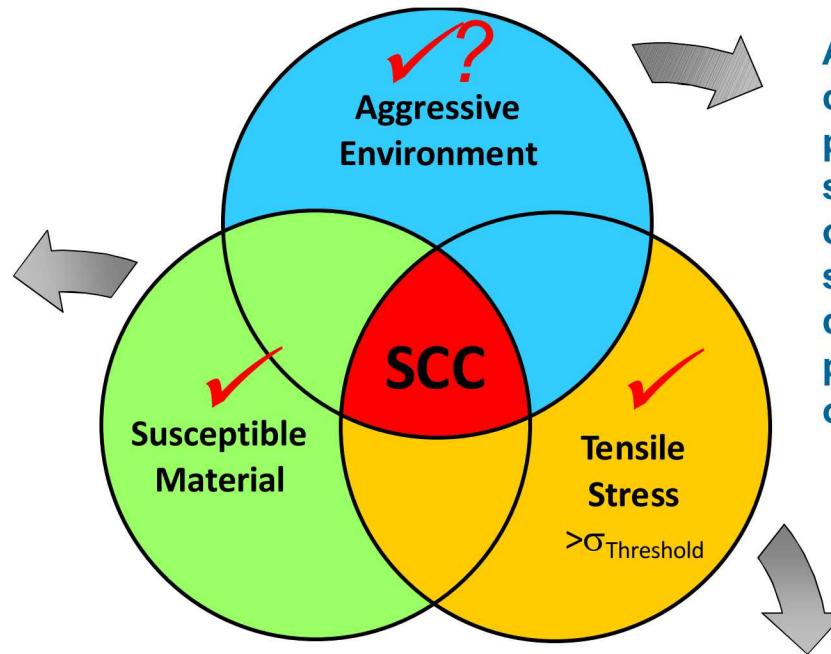


*ISFSI is ~0.5 miles from Chesapeake Bay*

- *Sheltered bay*
- *Brackish water*

# Criteria for Stress Corrosion Cracking

304 SS is known to be susceptible to SCC, and will be more susceptible in weld HAZ.



**Conclusion:** At some near-marine ISFSI sites, all three criteria for stress corrosion cracking are likely to eventually be met, and SCC may occur.

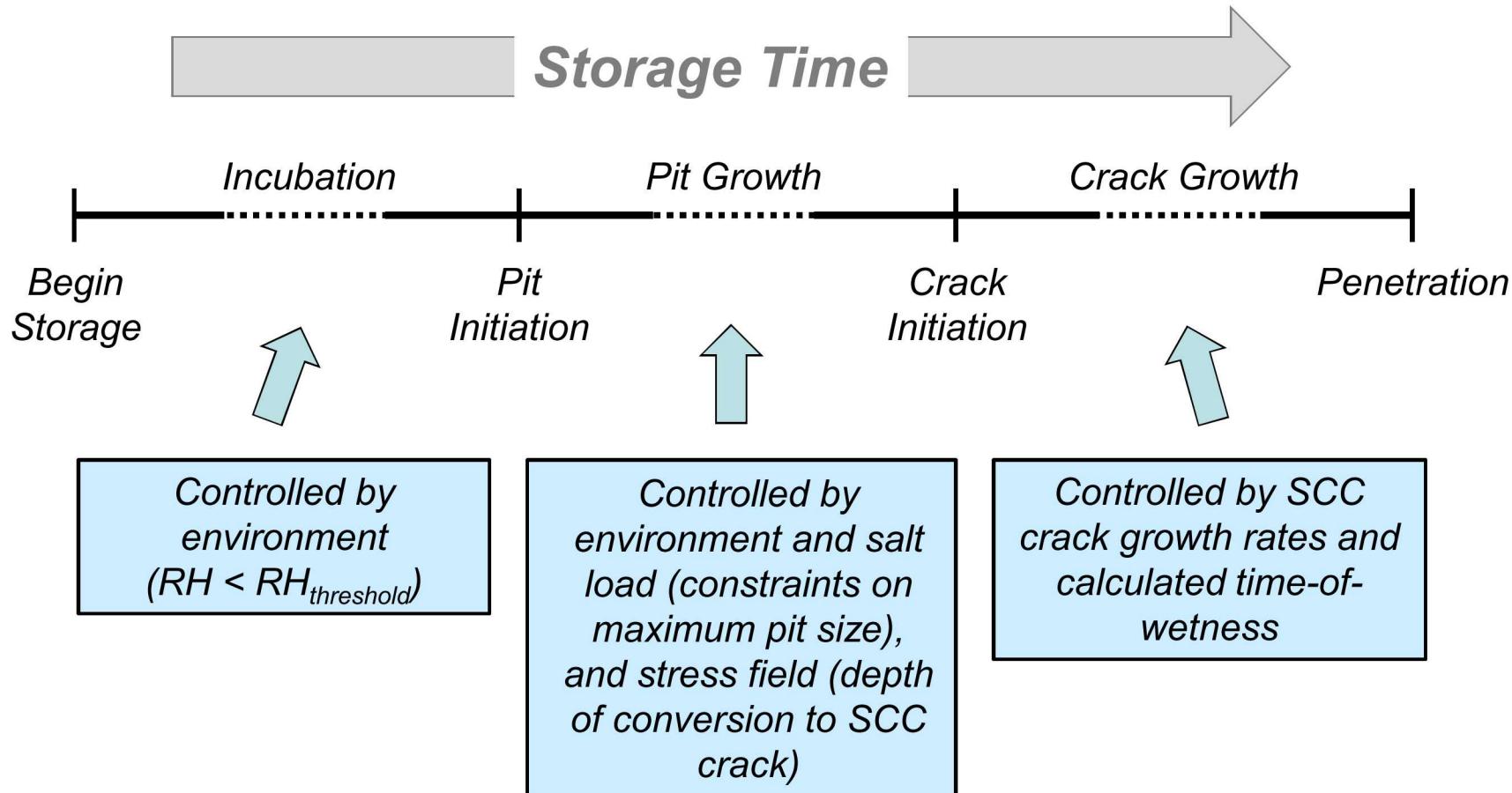
At some ISFSI sites, chloride-rich salts are present on canister surfaces, and eventually, once canisters cool sufficiently to allow salt deliquescence, will form potentially corrosive brines on the canister surface.

Stress modeling and experimental measurements on prototypical weld regions indicate that high through-wall tensile stresses are present in canister weld zones.

## Sandia probabilistic SCC model

- **Places important events and processes into a time-line for canister stress corrosion cracking. Includes sub-models for:**
  - Evolution of the chemical and physical environment on the canister surface
  - Pitting and SCC crack initiation
  - SCC crack growth
- **Existing data do not allow accurate prediction of occurrence and growth of SCC cracks. Current model is used primarily to:**
  - Identify the most important parameters controlling canister corrosion performance (SCC penetration times).
  - Prioritize research needs for predicting SCC occurrence and growth
- **Model is being constantly updated as additional information becomes available**

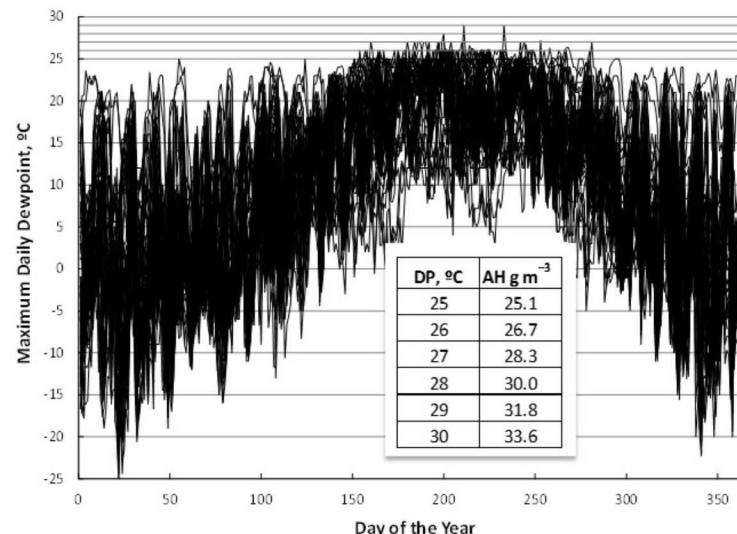
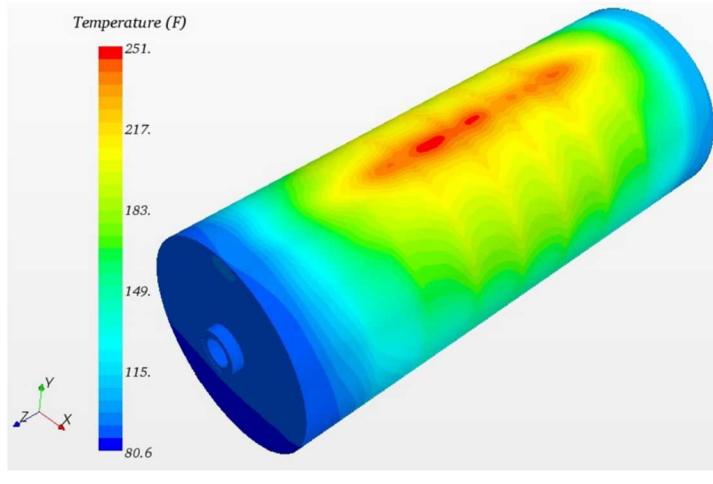
## General Timeline for SCC Initiation and Growth



**Predicting SNF canister performance requires developing models to evaluate each process, and the duration of each period.**

## Incubation Period

Ends when temperatures drop sufficiently to allow salt deliquescence and corrosion to occur



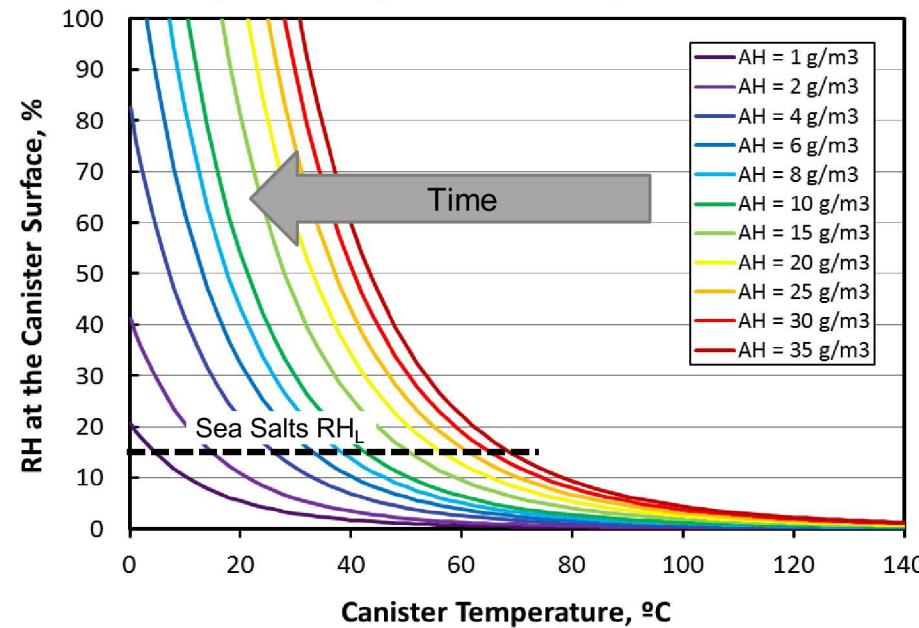
Surface temperature estimates



Site-specific National Weather Service data: daily and seasonal variations in T, AH

$$\text{Canister surface RH} = f(\text{Ambient AH, canister surface T})$$

Use weather data and predicted canister surface temperature to predict RH at any location and time.



Timing of corrosion initiation—point in time at which RH<sub>L</sub> is first reached.

Summing time when RH>RH<sub>L</sub> provides “time of wetness”; time when corrosion can actually occur.

Both vary with canister surface location.

### Pitting and SCC crack initiation

- Corrosion starts (pits form) once the  $RH_{threshold}$  is exceeded.
- Pits grow over time, but for any given set of environmental conditions ( $T$ ,  $RH$ , salt load), *pit depths reach a limiting value*.
- The maximum possible pit depth increases over time as the canister cools and more salt accumulates.
- Pits generate aggressive chemistry and act as local stress focusers. SCC cracks initiate from corrosion pits on the metal surface.
- Depth of pit-to-crack transition is a function of the tensile stress field at the pit location. As pits grow larger, they are more likely to initiate cracks.

Once a crack forms, crack growth rates are a function of many parameters:

- In general:

$$\frac{dx_{crack}}{dt} = \dot{x}_{crack} = \alpha_{crack} f(T) f(K) f(R_a) f([Cl^-]) f(m_{Cl}) f(pH) f(\sigma_{ys}) \dots$$

- A version incorporating only the effect of T and crack tip stress intensity factor K is commonly used:

$$\frac{dx_{crack}}{dt} = \alpha_{crack} \cdot \exp \left[ -\frac{Q}{R} \left( \frac{1}{T} - \frac{1}{T_{ref}} \right) \right] \cdot (K - K_{th})^{\beta_{crack}}$$

$dx_{crack}/dt$  = crack growth rate

$\alpha_{crack}$  = crack growth amplitude

$Q$  = activation energy for g=crack growth

$R$  = universal gas constant ( $8.314 \text{ J mol}^{-1} \text{ K}^{-1}$ )

$T$  = temperature (K) of interest

$T_{ref}$  = reference temperature (K) at which  $\alpha$  was derived.

$K$  = crack tip stress intensity factor

$K_{th}$  = threshold stress for SCC

$\beta_{crack}$  = stress intensity factor exponent.

Where  $K = \sigma_{applied} Y \sqrt{\pi x_{crack}}$

$\sigma_{applied}$  = tensile stress

$Y$  = shape factor

$x_{crack}$  = depth

Once a crack forms, crack growth rates are a function of many parameters:

- Controls on crack growth rate:
  - Temperature
  - Tensile stress
  - Material properties (composition, yield strength, sensitization)
  - Brine properties (pH, chloride concentration)
  - SCC crack growth
- A version incorporating only the effect of T and crack tip stress intensity factor K is commonly used:

$$\frac{dx_{crack}}{dt} = \alpha_{crack} \cdot \exp \left[ -\frac{Q}{R} \left( \frac{1}{T} - \frac{1}{T_{ref}} \right) \right] \cdot (K - K_{th})^{\beta_{crack}}$$

$dx_{crack}/dt$  = crack growth rate

$\alpha_{crack}$  = crack growth amplitude

Q = activation energy for g=crack growth

R = universal gas constant (8.314 J mol<sup>-1</sup> K<sup>-1</sup>)

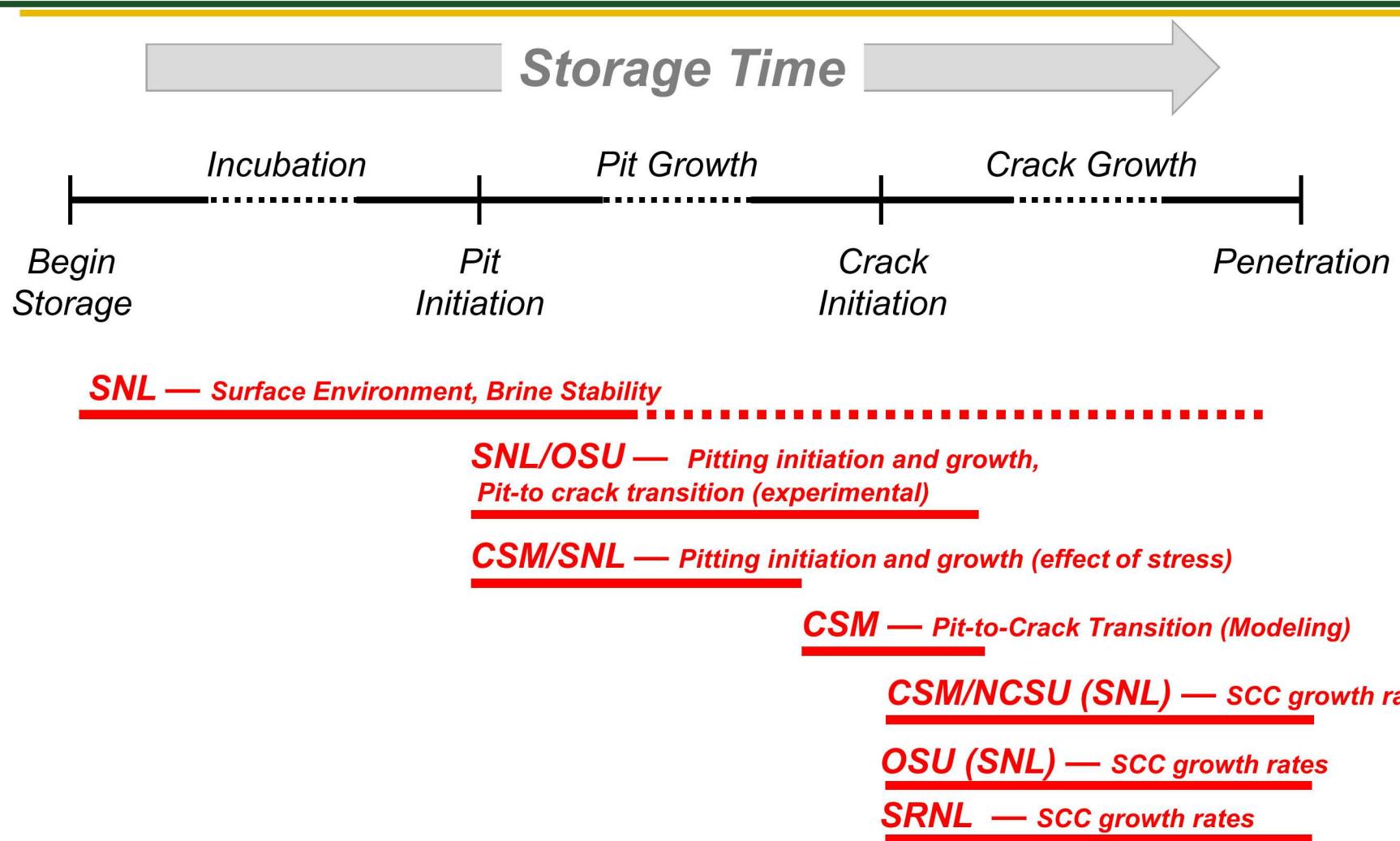
T = temperature (K) of interest

$T_{ref}$  = reference temperature (K) at which  $\alpha$  was derived.

K = crack tip stress intensity factor

$K_{th}$  = threshold stress for SCC

$\beta_{crack}$  = stress intensity factor exponent.



- **Criteria for Stress Corrosion Cracking of SNF interim storage canisters have been met, or will be met in the future**
  - Canister material (304 SS) is susceptible to SCC
  - High through-wall tensile stresses are present in welds and HAZ
  - Chloride-rich salts are present on canister surfaces at some ISFSI locations, and will eventually deliquesce to form corrosive brines.
- **Corrosion timeline can be divided into 3 periods**
  - Incubation period—controlled by environment (T, RH), ends when canister cools to the point that corrosion can occur.
  - Pitting period—localized corrosion; extent of damage (e.g., pit depth) controlled by salt load and RH. Crack initiation is more likely as pits deepen (Kondo criterion).
  - Crack Growth—Temperature-dependent crack growth rates determine when penetration occurs. Other controlling parameters?
- **DOE-funded efforts are underway to develop predictive models for canister performance; existing data do not support accurate prediction of crack initiation or possible penetration times**