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SAND2017-2584PE

Fuel Cycle Technologies

Evaluating Stress Corrosion Cracking of Spent Nuclear Fuel Interim Storage Canisters

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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: Creating a timeline for possible canister failure by SCC**
  - Corrosion incubation
  - Pitting and pit-to-crack transition
  - Crack growth and canister penetration

Background

- United States currently has over 80,000 metric tons of Spent Nuclear Fuel (SNF), about 30% in dry storage systems.
- The dry storage systems are intended as interim storage until a permanent disposal site is developed.
- Until recently, systems were licensed for 20 years with an additional 20 years upon license renewal; in 2011, CFR was modified to allow for initial 40 year license period, with an additional 40 year renewal. 15-20 systems/sites will have to be recertified in the next 10 years.
- However, the United States currently does not have a disposal pathway for SNF. In any conceivable scenario, some SNF will be in storage for decades beyond the original storage system specifications.

Background, cont.

- In most systems, SNF is stored in stainless steel (304 SS) canisters. Canisters are stored in passively-ventilated overpacks, and accumulate surface dust over time. Stress Corrosion Cracking (SCC) of stainless steel due to deliquescence of chloride-rich salts is a potential failure mechanism, especially given longer-than-intended storage times.
- Understanding SCC of interim storage containers has been determined to be a high priority data gap (EPRI 2011; DOE 2012; NRC 2012).
- Efforts to better understand the risk of canister failure by SCC, and to predict timing and conditions of occurrence are being pursued by the DOE and others.

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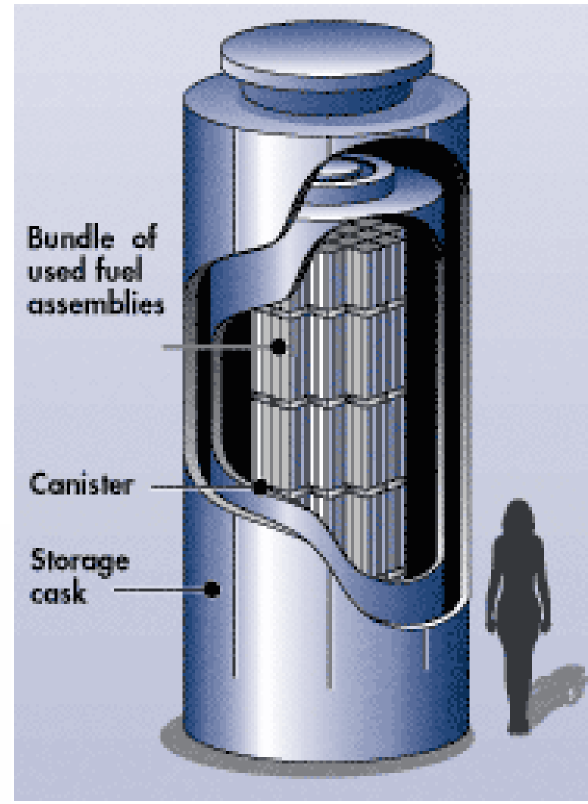
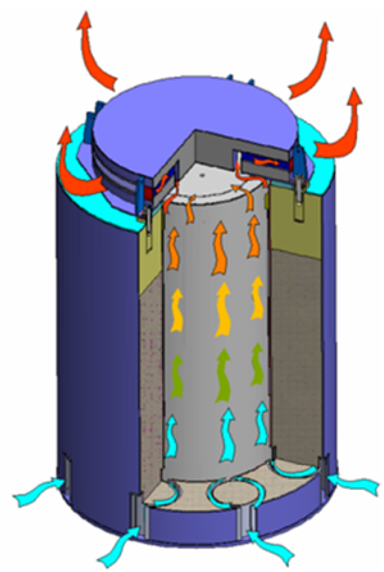
Canistered SNF Dry Storage Systems—Two Standard Designs

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.

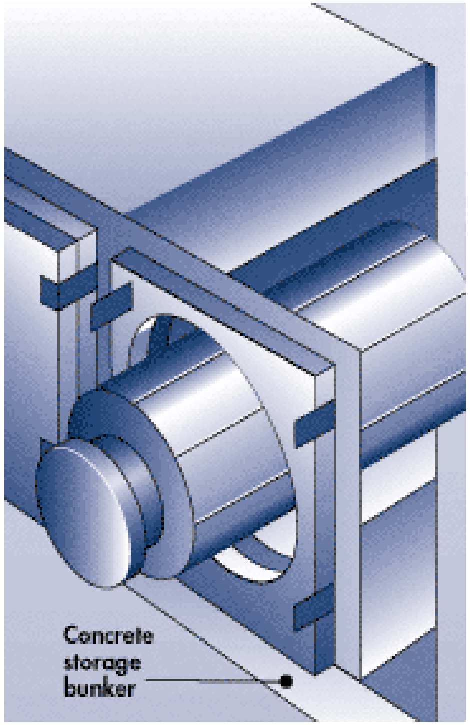


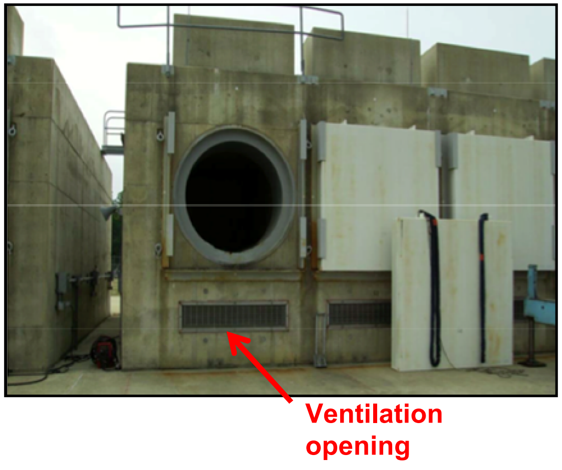


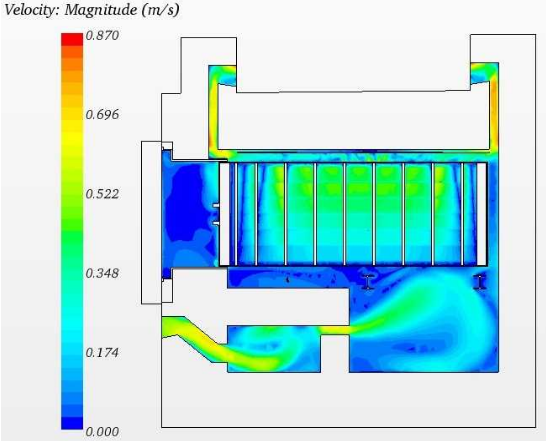
Pathway for air flow through the overpack.

Canistered SNF Dry Storage Systems—Two Standard Designs

<u>Horizontal</u>—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

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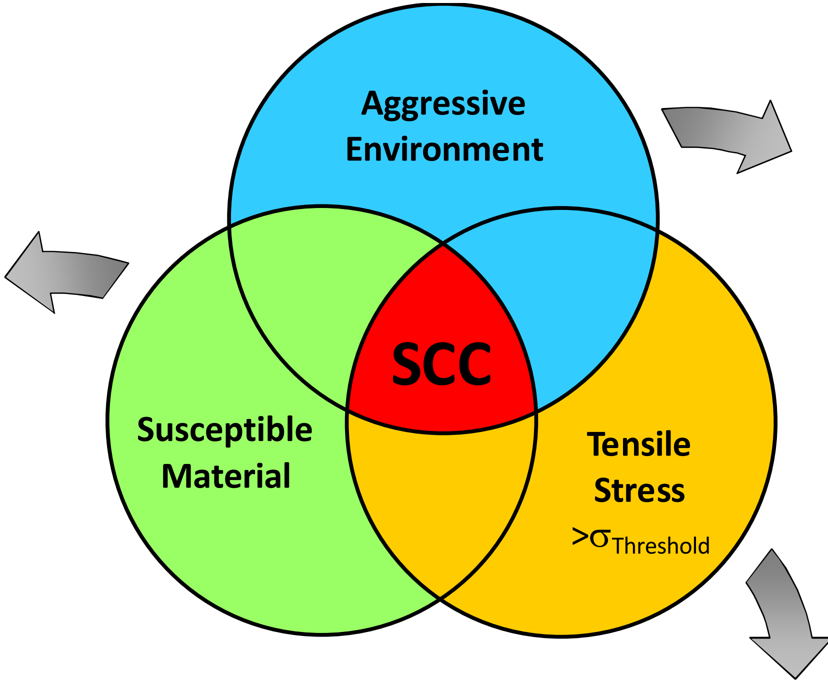
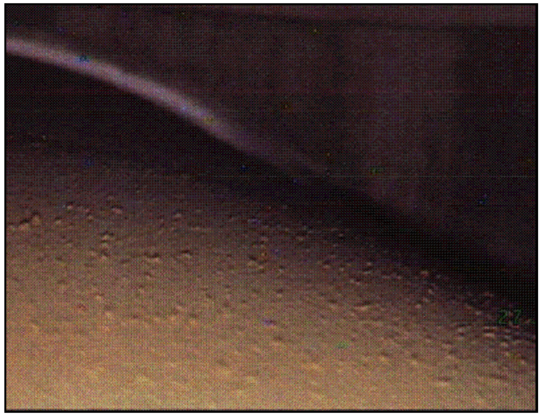
Criteria for Stress Corrosion Cracking

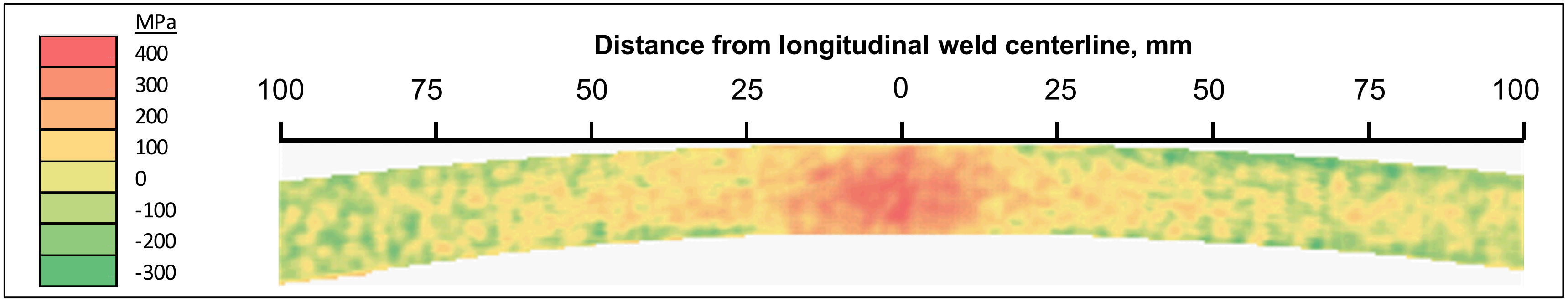
To evaluate the potential for occurrence of SCC, each must be considered

Weld zone, Ranor 304 SS plate





Dust on canister surface at Calvert Cliffs (EPRI 2014)



Measured weld residual stresses (SNL 2016)

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Susceptible Material

- **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 to date has shown SCC can initiate rapidly in this material if the other criteria are met.

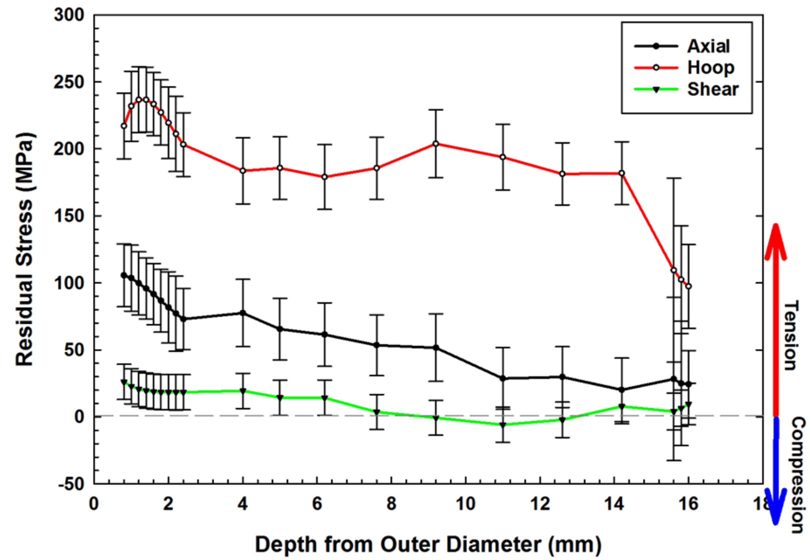
Weld zone, Ranor 304 SS plate

Rust spots on storage canister at Calvert Cliffs

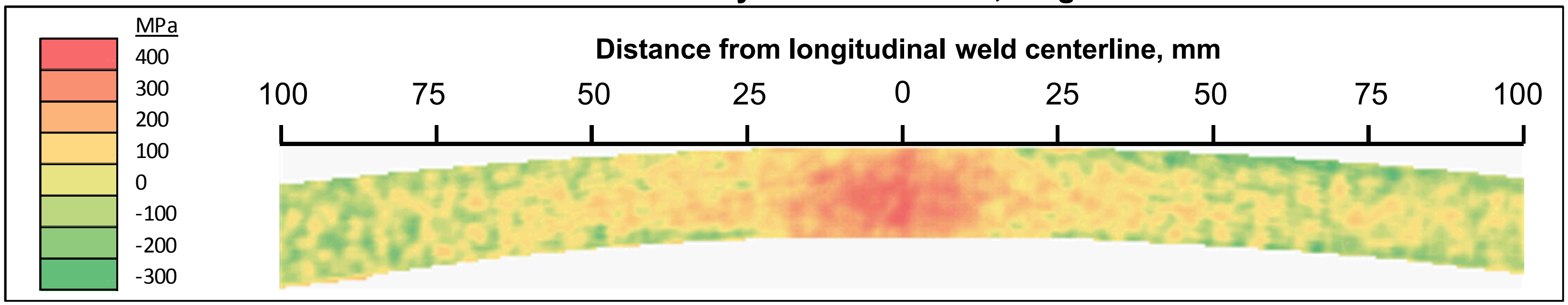
Tensile Stress Sufficient to Support SCC

- NRC modeling (NRC 2012) suggested high tensile stresses in welds and 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
  - Weld residual stresses measured using several methods, including DHD and contour method.
  - High through-wall tensile stresses measured at both longitudinal (seam) welds and circumferential welds. Highest tensile stresses parallel to welds.
  - Highest tensile stresses (up to 600 Mpa) measured at simulated weld repairs.
  - 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



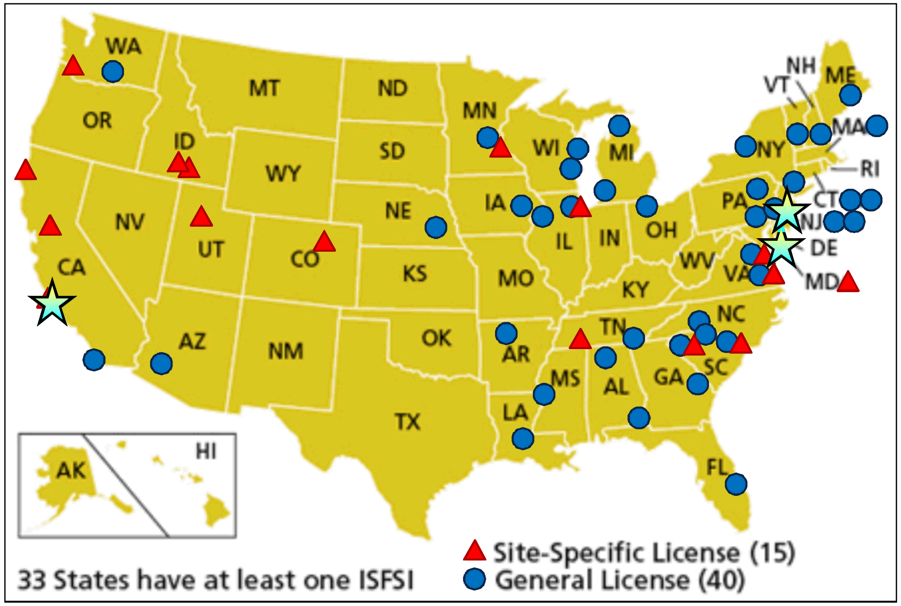
Residual stresses measured by contour method, longitudinal weld



Canister Surface Environment

- **Many Independent Spent Fuel Storage Installations are at coastal sites. Anticipated deposition of chloride-rich sea-salts, and formation of corrosive brines.**
- **EPRI sampling program: Assess the composition of dust on the surface of in-service stainless steel SNF storage canisters, with emphasis on the deliquescent salts.**
- **ISFSI locations sampled:**
  - **Calvert Cliffs, MD: Transnuclear NUHOMS system, horizontal storage canister (June, 2012)**
  - **Hope Creek, NJ: Holtec HI-STORM system, vertical canister (Dec, 2013)**
  - **Diablo Canyon, CA: Holtec HI-STORM system (Jan, 2014)**

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

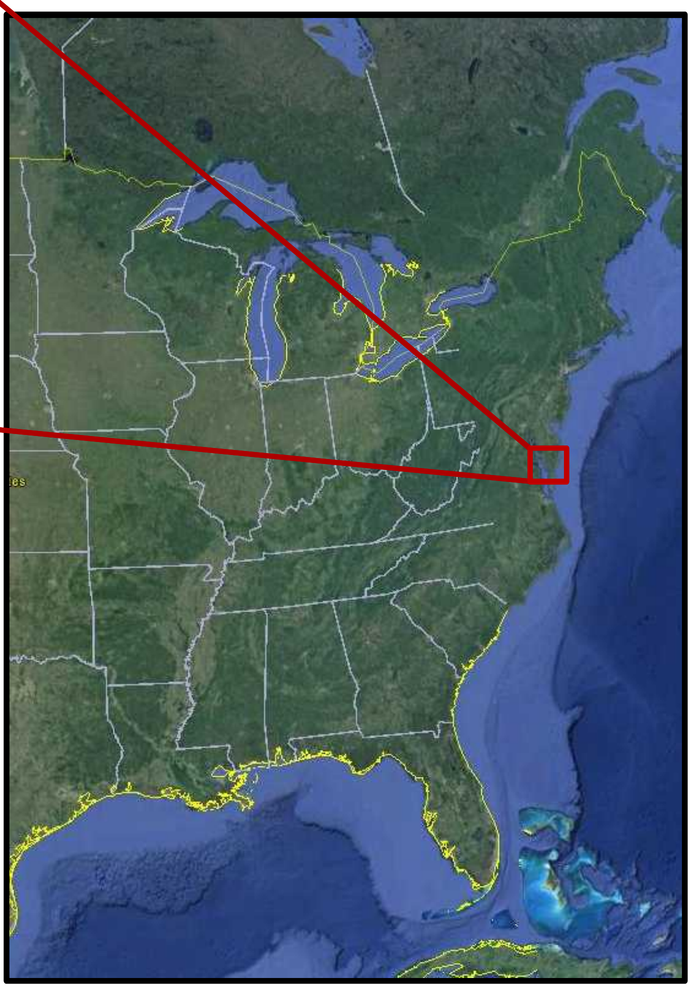


ISFSI locations sampled.

Samples delivered to Sandia National Labs for analysis

Calvert Cliffs ISFSI

Eastern U.S.



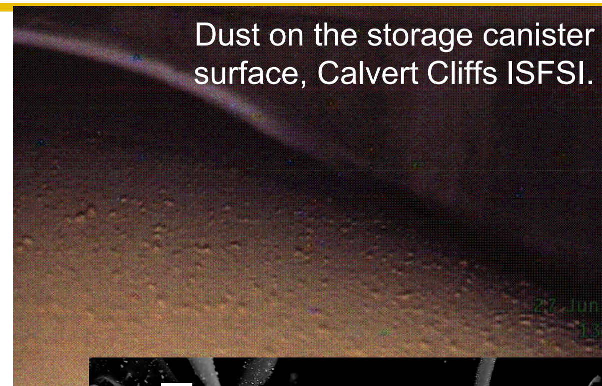
ISFSI is ~0.5 miles from Chesapeake Bay

- Sheltered bay
- Brackish water

Results: Calvert Cliffs

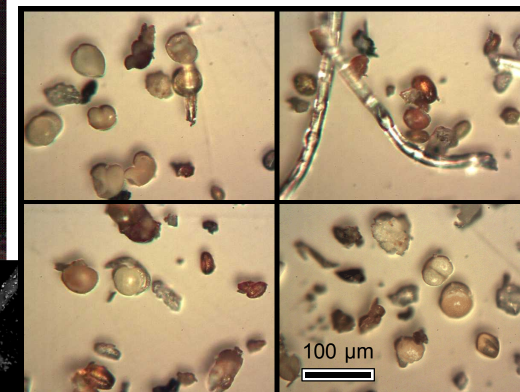
- Horizontal canisters 16-19 years in service, fairly cold.
- The canister upper surface was heavily coated with dust and salts due to gravitational settling. Samples from upper surface contained abundant pollen.
- The soluble salts are Ca- and SO_4-rich. Gypsum is the dominant salt phase present.
- Chlorides comprise a small fraction of the total salt load, and are dominantly NaCl.

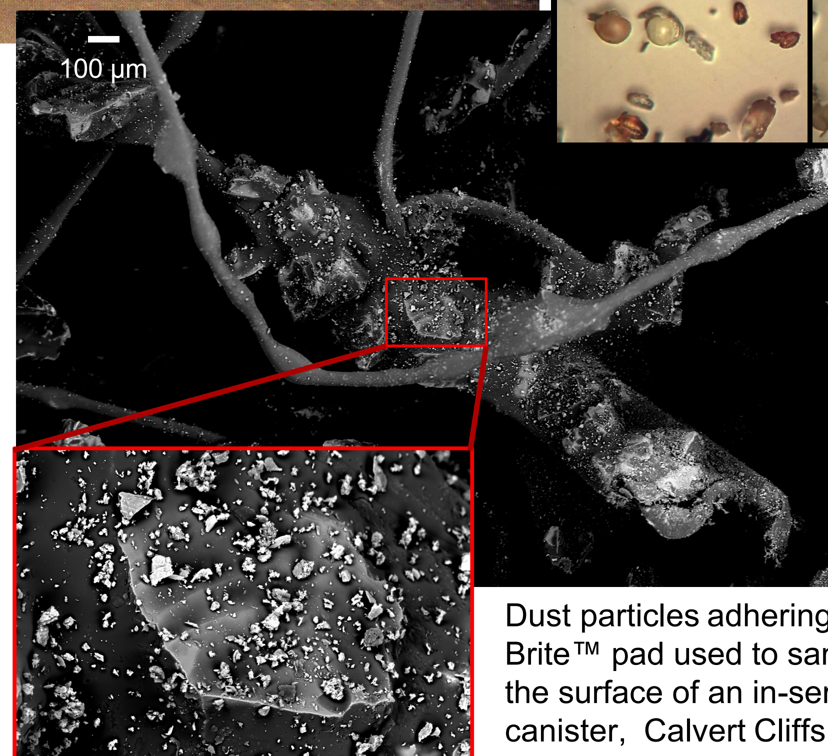
Despite the proximity to the coast, the dusts sampled from in-service containers at Calvert Cliffs do not appear to have a large sea salt component. Chesapeake Bay is brackish, and may be sheltered sufficiently to limit wave-generated sea-salt aerosols.



Dust on the storage canister surface, Calvert Cliffs ISFSI.

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

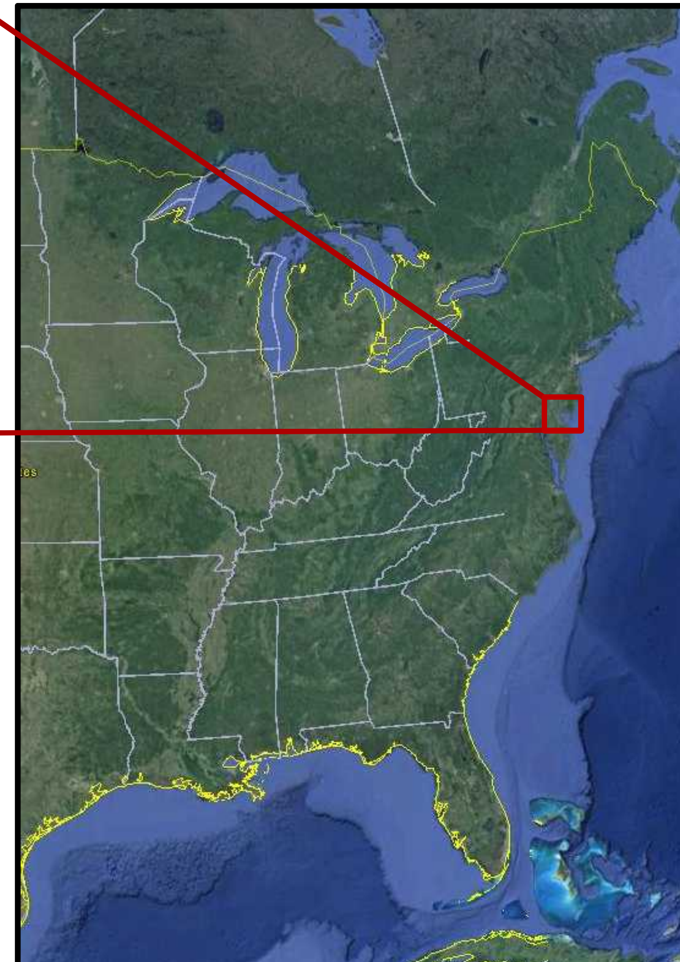




Dust particles adhering to a Scotch-Brite™ pad used to sample dust on the surface of an in-service storage canister, Calvert Cliffs ISFSI.

Hope Creek ISFSI

Eastern U.S.

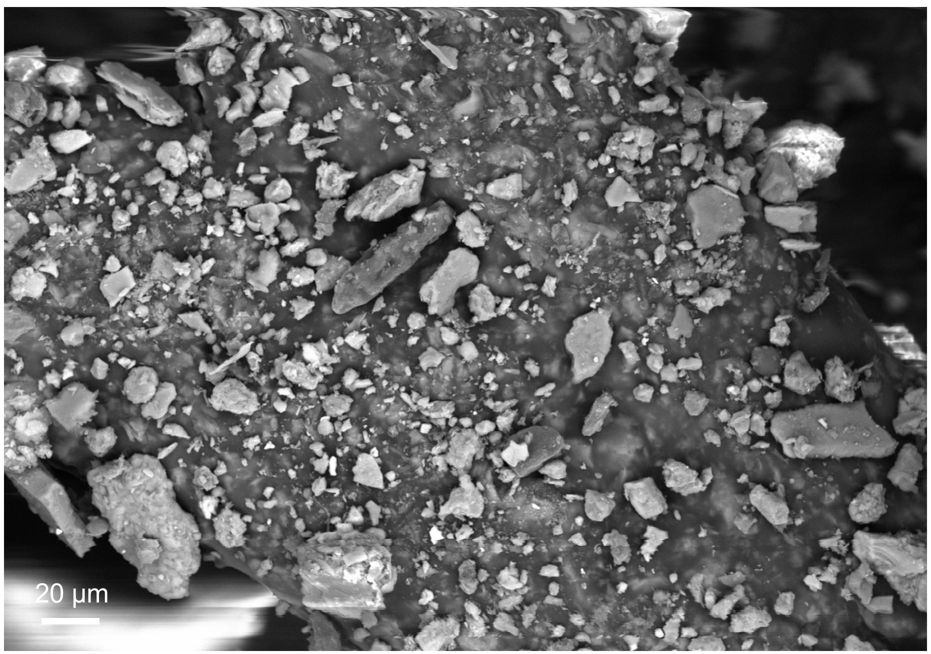


ISFSI is ~0.25 miles from the Delaware River, 15 miles upstream from Delaware Bay

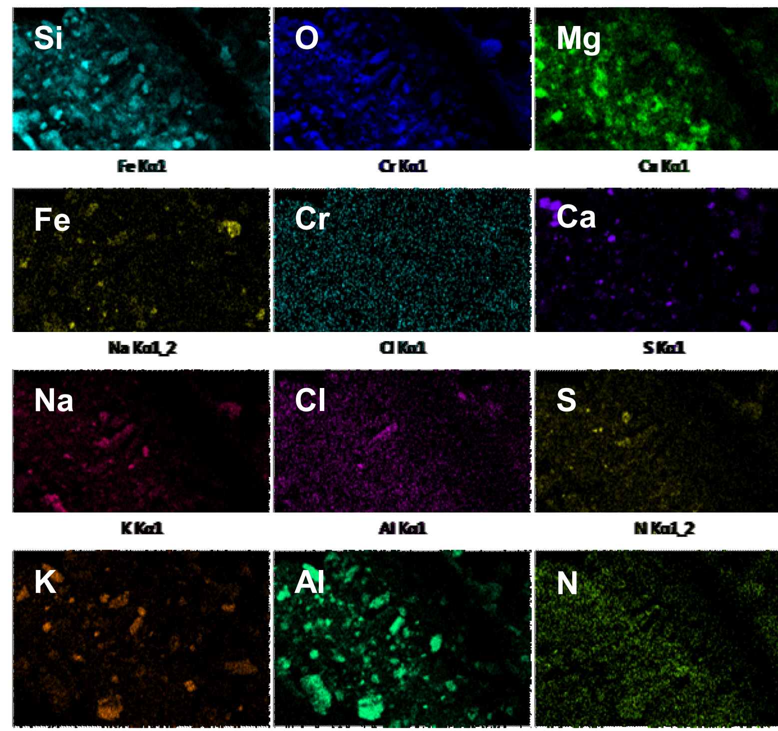
- Brackish water
- Sheltered from open ocean

Results: Hope Creek

- Vertical HOLTEC canisters.
- Flat canister top much more heavily coated than vertical sides.
- Dust dominated by insoluble minerals (quartz, clays, aluminosilicates). Soluble salts minor; dominantly gypsum, carbonates. Sparse chlorides, mostly isolated grains of NaCl.



Dust particles adhering to a Scotch-Brite™ pad used to sample dust at Hope Creek (Bryan and Enos, 2014: SAND2014-16383)



Results: Hope Creek, continued

Chemistry of soluble salts

Sample	Loc.	Depth, ft	Temp, °F	Amount present, µg/sample										SUM
				K	Ca	Mg	Na	NH_4^+	F^-	Cl^-	NO_3^-	PO_4^{3-}	SO_4^{2-}	
144-008	Side	13.0	93.2	*0.8*	3.4	0.6	*0.1*	2.7		0.9	2.7		4.1	15.4
144-009	Side	7.5	116.5	1.7	4.5	0.5	*0.1*	2.7		0.9	6.4	*1.1*	6.5	24.3
144-010	Side	1.0	133.9	1.4	4.2	0.4	*0.4*	2.4		1.2	5.0		4.4	19.4
144-013	Top	0.0	138	18	102	33	42	2.8	0.4	4.2	19	4.8	91	317
144-014	Top	0.0	141.2	6.4	29	8.0	13.4	2.7	0.4	18	7.3	*1.3*	55	142
144-003				*0.6*	2.2	0.4		1.4		*0.5*	3.3	1.2	2.1	11.6
144-004				*0.3*	3.2	0.6		2.9		0.8	1.8	*0.5*	1.7	11.8
145-006*	Side	13.0	70.6	2.2	4.4	0.6	*0.5*	2.3		2.2	8.1		4.7	25.1
145-007	Side	7.5	100.8	1.0	2.4	0.6	*0.7*	2.9		2.1	2.2	*0.7*	5.3	17.9
145-014	Side	1.0	130.3	*0.9*	3.2	0.8	*0.6*	3.2		1.2	2.5		9.1	21.5
145-013**	Top	0.0	174.1	15	91	30	32	2.8		2.2	15	3.5	82	273
145-011**				*0.2*	2.3	0.3		3.0		*0.7*	1.3		1.7	9.6
145-002				1.2	4.8	0.5		2.7		*0.7*	5.9	*0.8*	2.0	18.5
SS-Bl-8 min-1					1.3	0.2		1.1		*0.4*	1.6		*0.6*	5.1
SS-Bl-8 min-2					1.2	0.2		1.5		*0.7*	*0.9*	*0.5*	*0.2*	5.2
SS-Bl-15 min					1.5	0.5		5.7	0.2	*0.7*	1.1	1.6	1.7	12.9

Salts are calcium, sulfate, nitrate-rich. Minor chloride salts in some samples.
(Bryan and Enos, 2014: SAND2014-16383)

Despite the proximity to the coast, the dusts sampled from in-service containers at Hope Creek do not have a large sea salt component. Delaware River is brackish, and may be sheltered sufficiently to limit wave-generated sea-salt aerosols.

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Diablo Canyon ISFSI

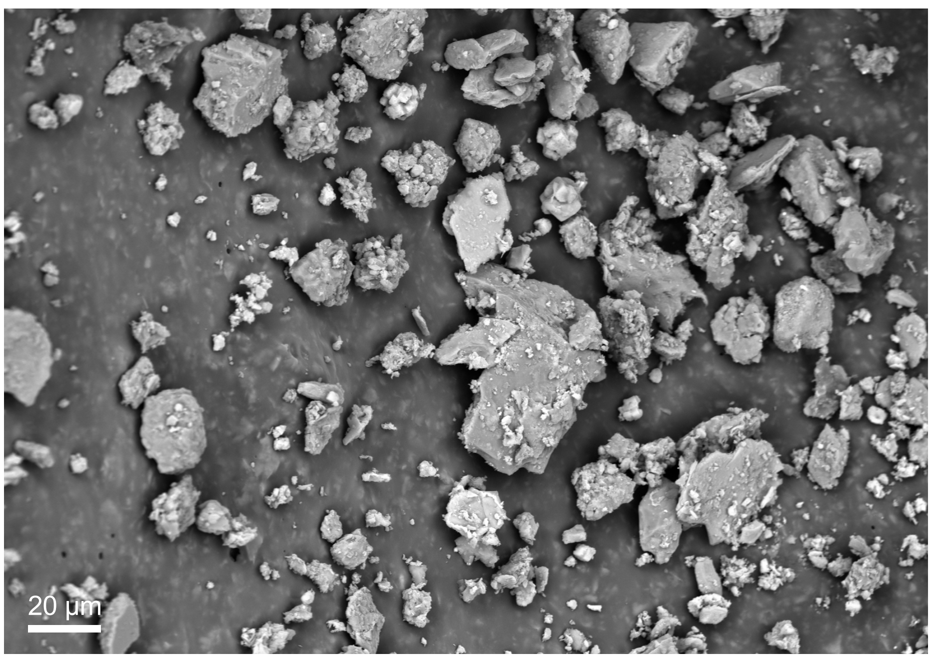
Western U.S.

ISFSI is ~1/3 mile from the shoreline, on a hill above the plant.

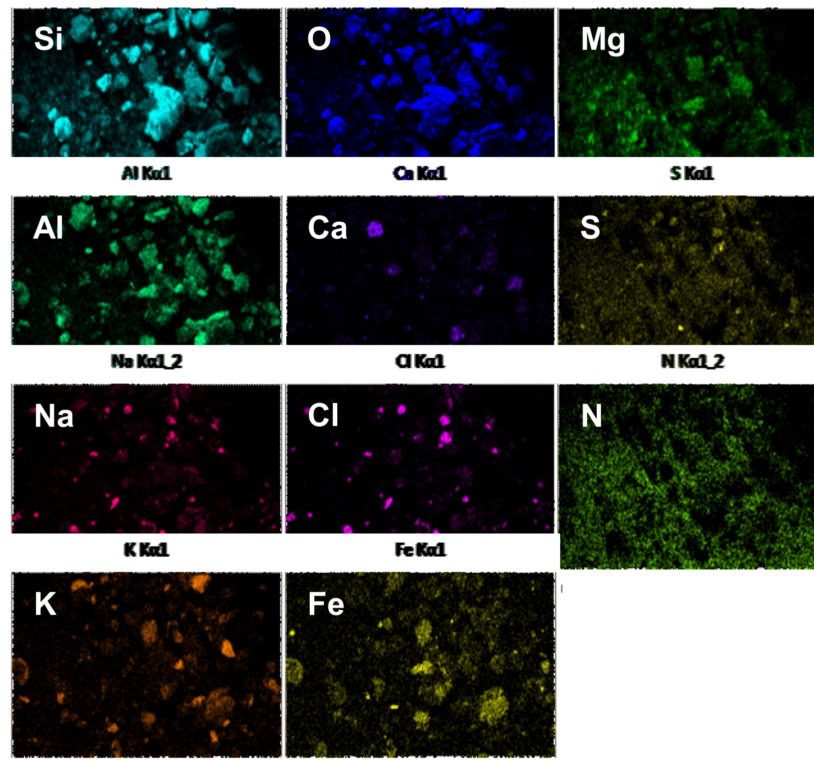
- Elevated (~400 feet) above sea level
- Rocky shore, breaking waves
- Open ocean

Results: Diablo Canyon

- Vertical HOLTEC canisters, 2-4 years in storage (hot!).
- Canister sides lightly coated, tops more heavily coated.
- Dust dominated by insoluble minerals (quartz, clays, aluminosilicates), but chloride-rich soluble salts are abundant, (10-20%) present as sea-salt aggregates.

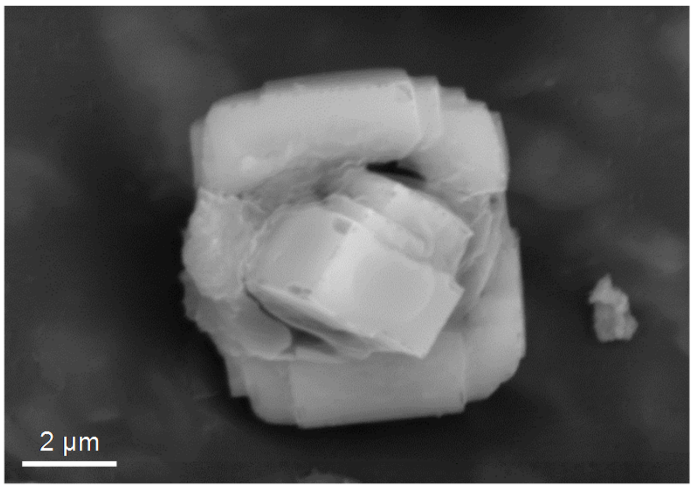


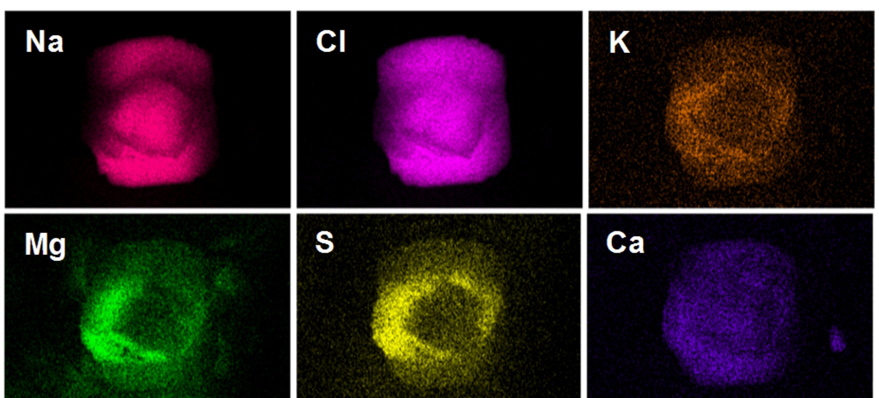
Dust particles adhering to a Scotch-Brite™ pad used to sample dust at Diablo Canyon



Diablo Canyon Sea-Salt Aggregates

Salt aggregate: dominantly NaCl with interstitial $MgSO_4$ and trace K, Ca phases. Consistent with seawater ion compositions.



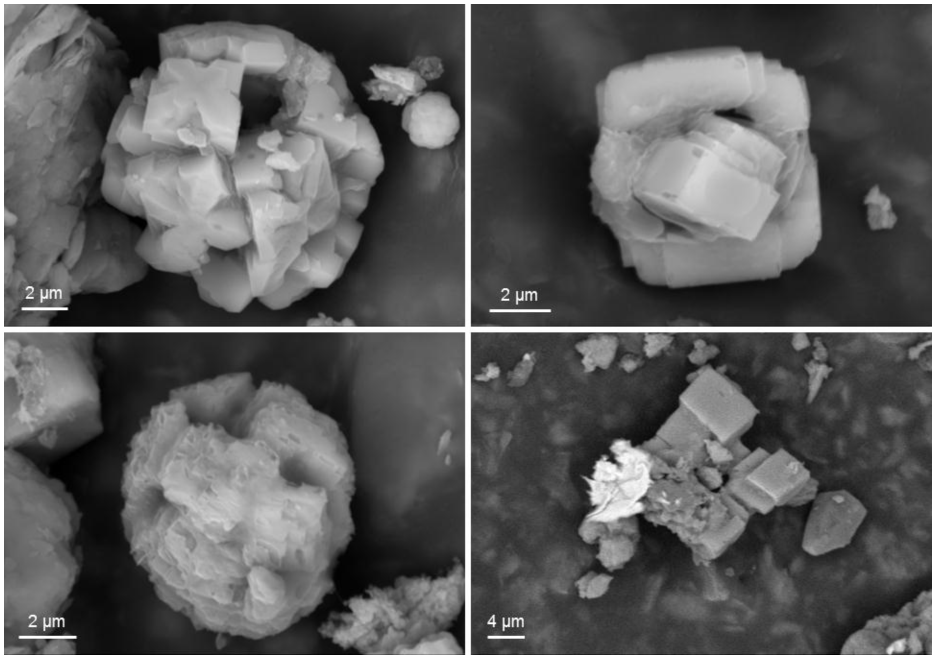


At near-marine sites, salt aggregates that form by evaporation of seawater can be deposited on canister surfaces, and will deliquesce to form chloride-rich brines.

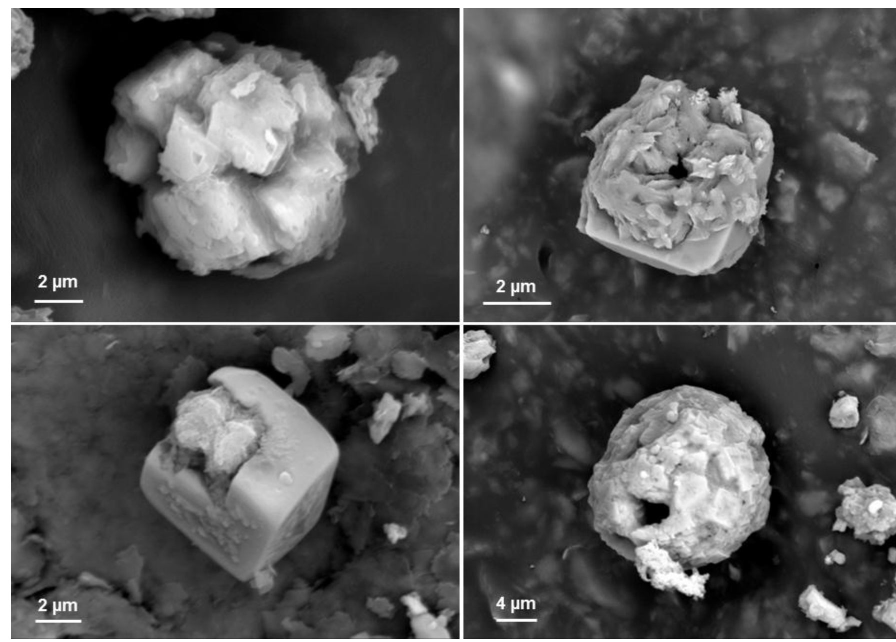
Synthetic ocean water (ASTM D1141-98)

Species	Conc., mg/L ASTM D1141-98
Na^+	11031
K^+	398
Mg^{2+}	1328
Ca^{2+}	419
Cl^-	19835
Br^-	68
F^-	1
SO_4^{2-}	2766
BO_3^{3-}	26
HCO_3^-	146
pH	8.2

Diablo Canyon Sea-salt Aerosols

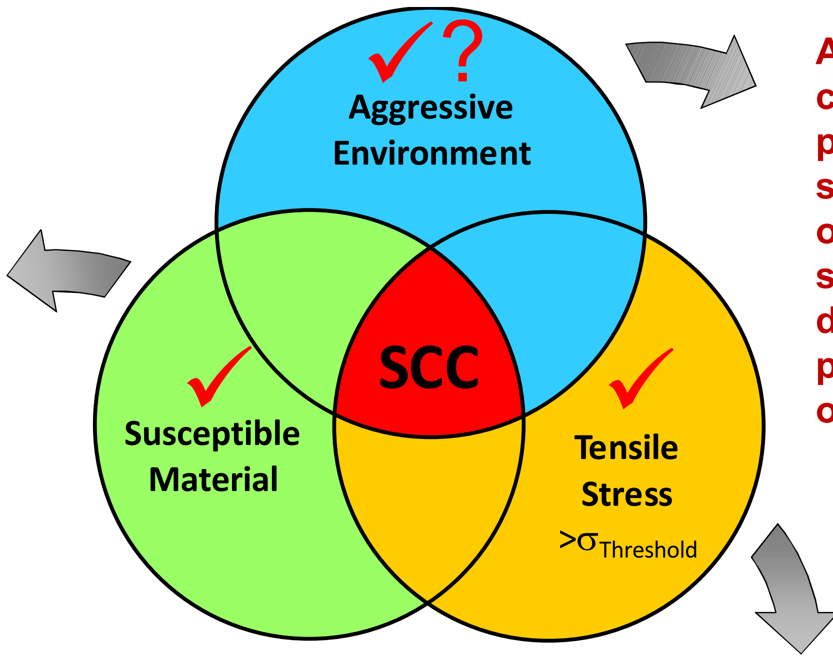


Sea-salt aerosols: Sub-spherical or euhedral aggregates of sodium chloride with interstitial magnesium sulfate, trace calcium and potassium phases. Morphology suggests formation by crystallization of an aerosol droplet from the outside inwards.



Heavy wave action at the Diablo Canyon site generates abundant sea-salt aerosols. Although 400 feet above sea level, Diablo Canyon canisters have a significant amount of sea-salts on the canister surfaces, which may have entered overpacks as “sea-fog”.

Criteria for Stress Corrosion Cracking



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

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.

FE modeling and experimental measurements on prototypical weld regions indicate that high through-wall tensile stresses are present.

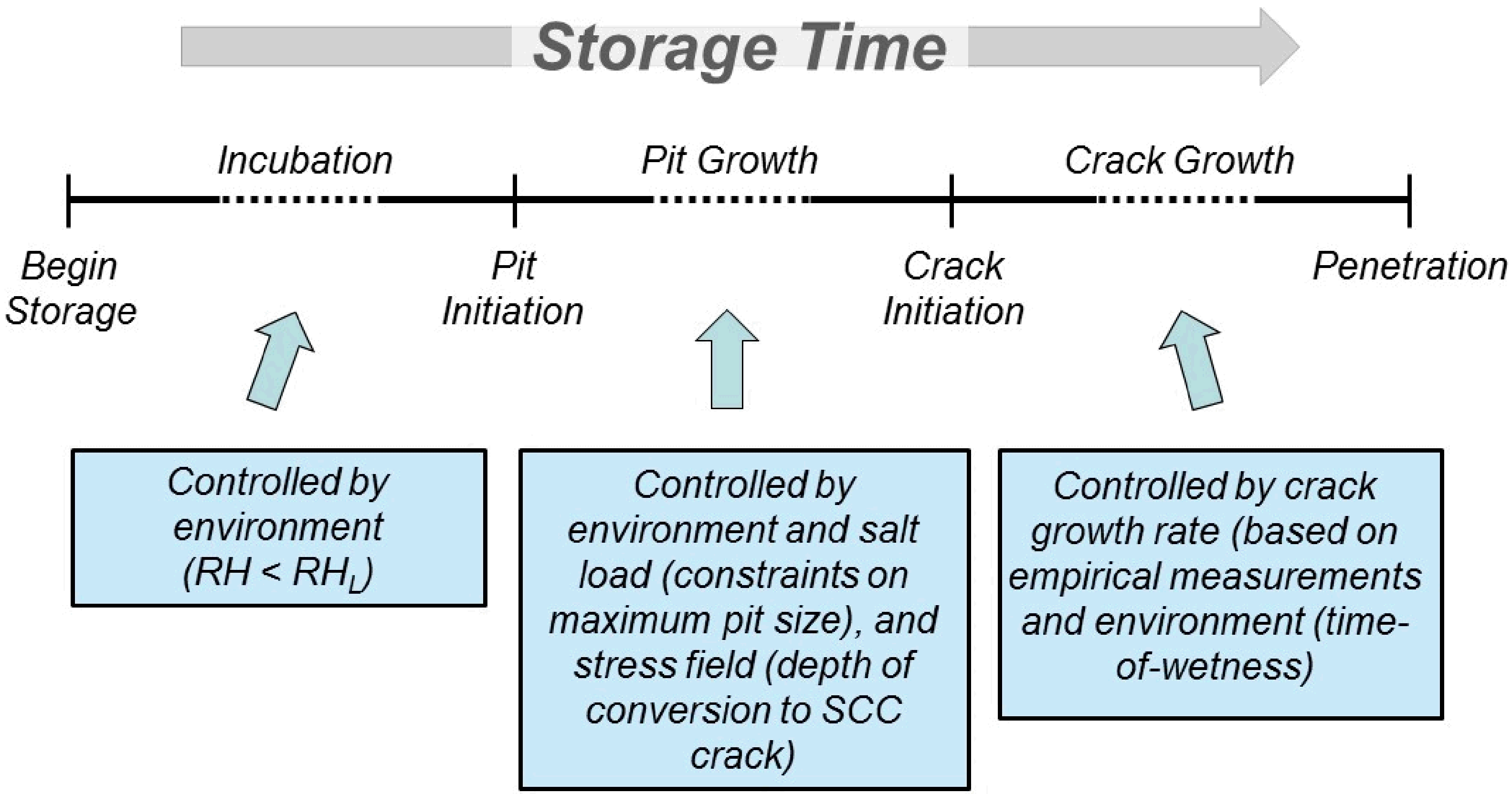
Conclusion: At some near-marine ISFSI sites, all three criteria for stress corrosion cracking are likely to eventually be met, and SCC may occur.

Evaluating Long-Term Canister Performance

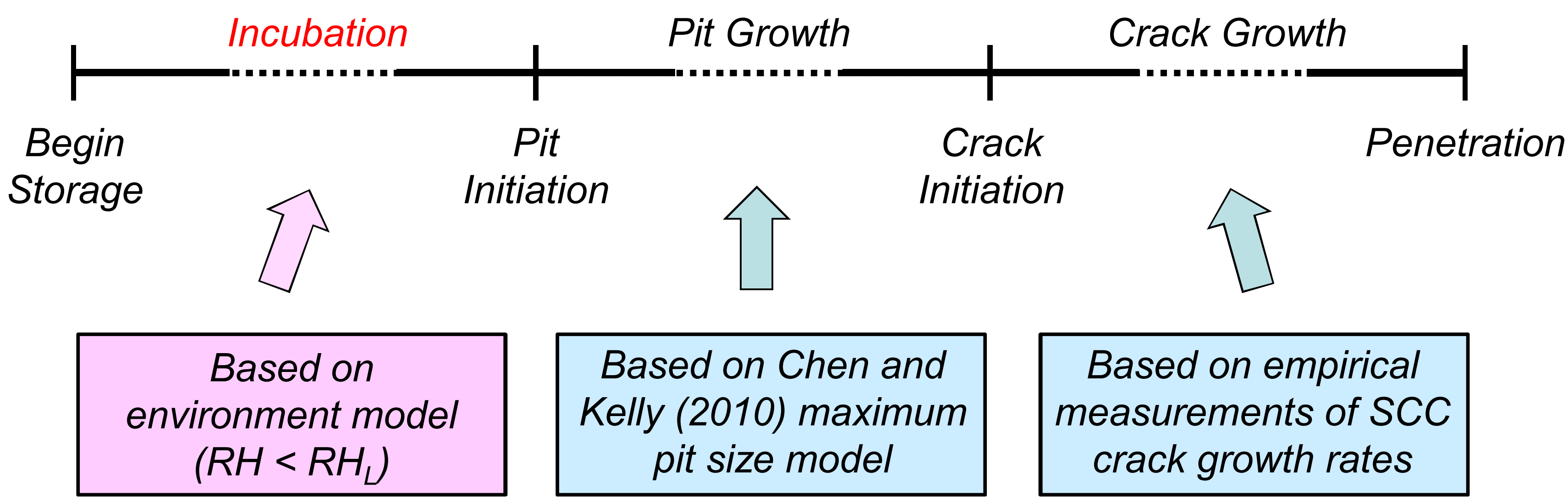
Sandia probabilistic SCC model

- **Places important events and processes into a time-line for canister stress corrosion cracking. Includes models for:**
  - Evolution of the chemical and physical environment on the canister surface
  - Pitting and SCC crack initiation
  - SCC crack growth
- **Major goals**
  - Identify most important parameters controlling canister corrosion performance (SCC penetration times).
  - Model used to prioritize research needs for predicting SCC occurrence/growth
- **Current model is simplified**
  - Develops the functional form and identifies data needs for the model
  - Allows testing of parameter sensitivities.
  - Model is constantly evolving to include latest data and to improve process modeling

General Timeline for SCC Initiation and Penetration



Canister Surface Environment



Canister Surface Environment Model

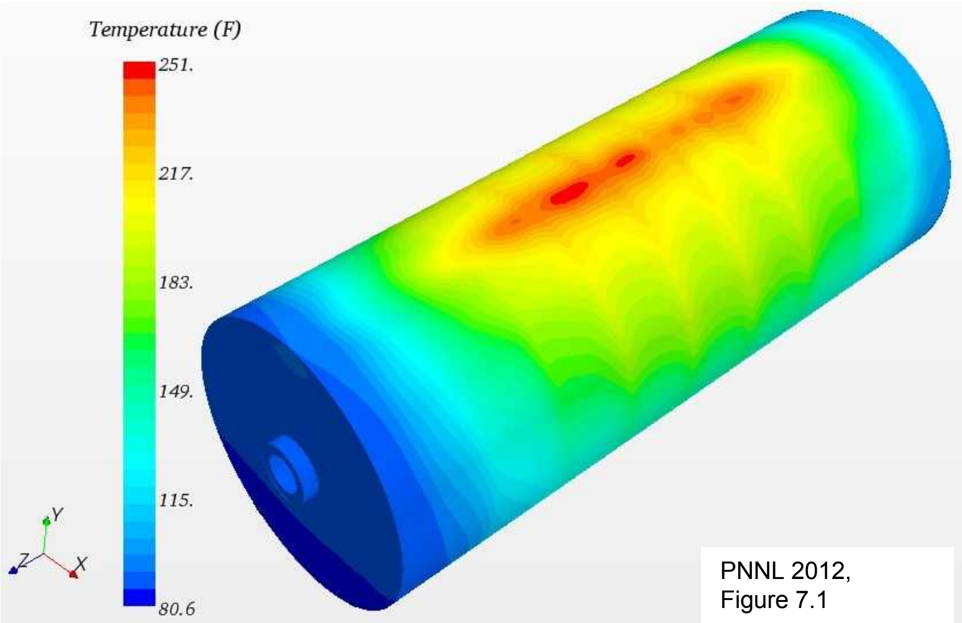
Corrosion can occur when a threshold RH is reached

The following conditions must be met:

- **Aqueous conditions—a deliquescent brine must be present.**
  - **Deliquescence is *f* (T, AH, salt composition):**
    - Temperature on the waste package surface at any given location, as a function of time and heat load. PNNL CFD thermal models of horizontal and vertical canisters in overpack
    - Ambient environment: absolute humidity (AH) and temperature as a function of time. NOAA site data from 64 sites near each existing ISFSI
    - Composition of salts (controls deliquescence RH) *Assume sea-salt assemblage*
- **Brine must be corrosive (chloride-rich).** *Assume sea-salts*
- **Threshold chloride concentration for corrosion?** *SCC model implements a chloride concentration threshold implicitly, within a model for the maximum possible pit size at any given time step.*

Environment Model: Waste Package Surface Temperatures

Horizontal canister

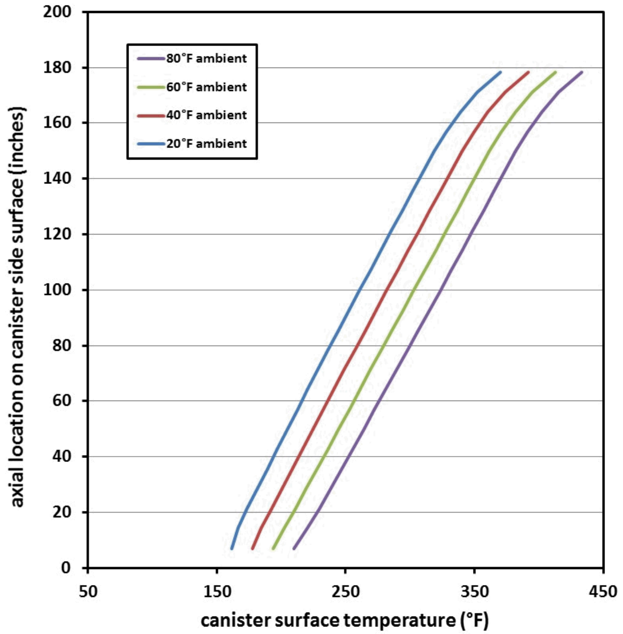


PNNL 2012, Figure 7.1

Location-specific evolution of surface temperature is important to determine:

- Timing of deliquescence and potential corrosion initiation
- Temperature-dependent corrosion parameters (e.g. crack growth rate)

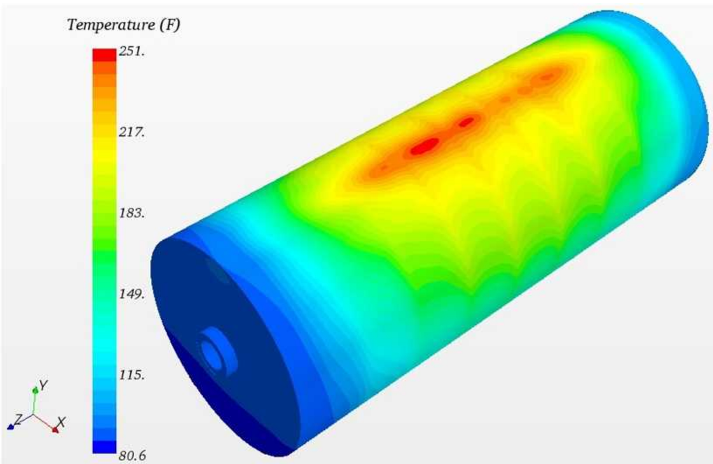
Vertical canister



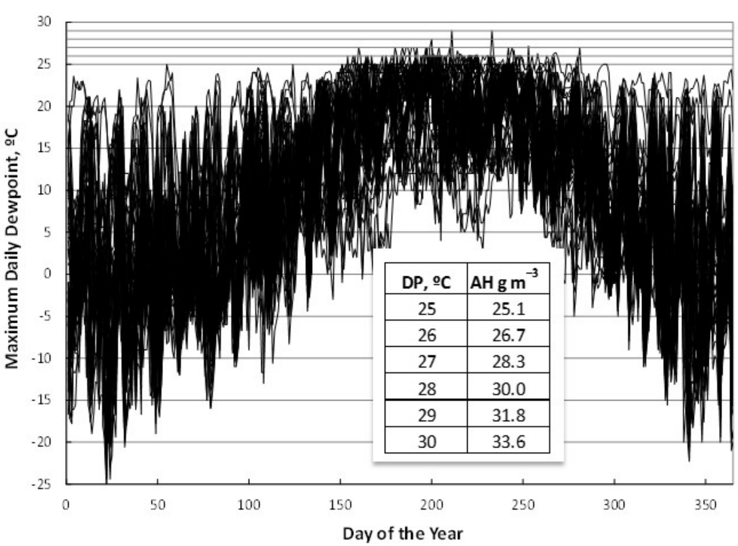
PNNL thermal models

- provide surface temperature maps through time
- horizontal canister (NUHOMS 24P)
- vertical canister (HOLTEC HI-STORM 100)

Incubation Period



Surface temperature estimates

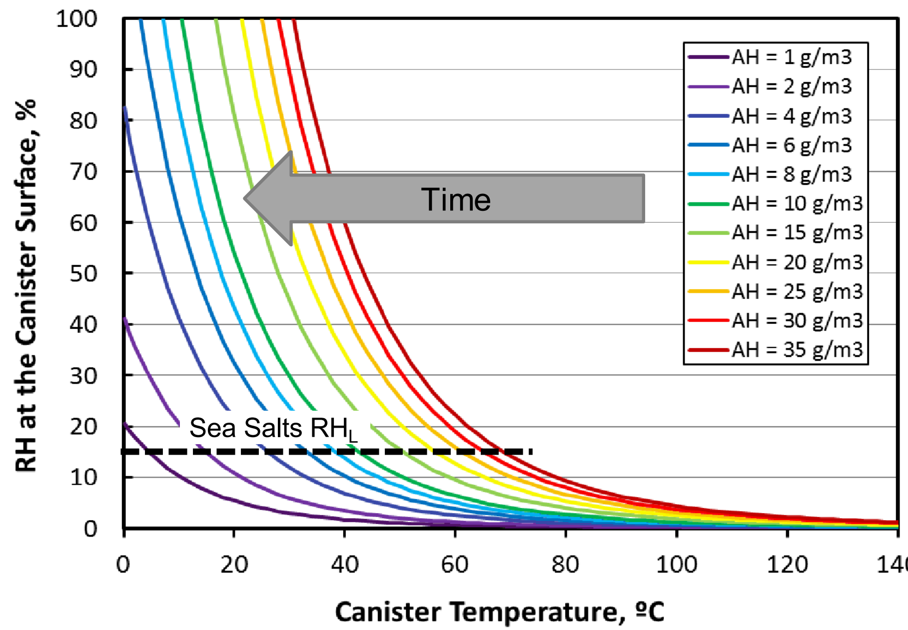


DP, ºC	AH g m^{-3}
25	25.1
26	26.7
27	28.3
28	30.0
29	31.8
30	33.6

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

Canister surface RH =
***f*(Ambient AH, canister surface T)**

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



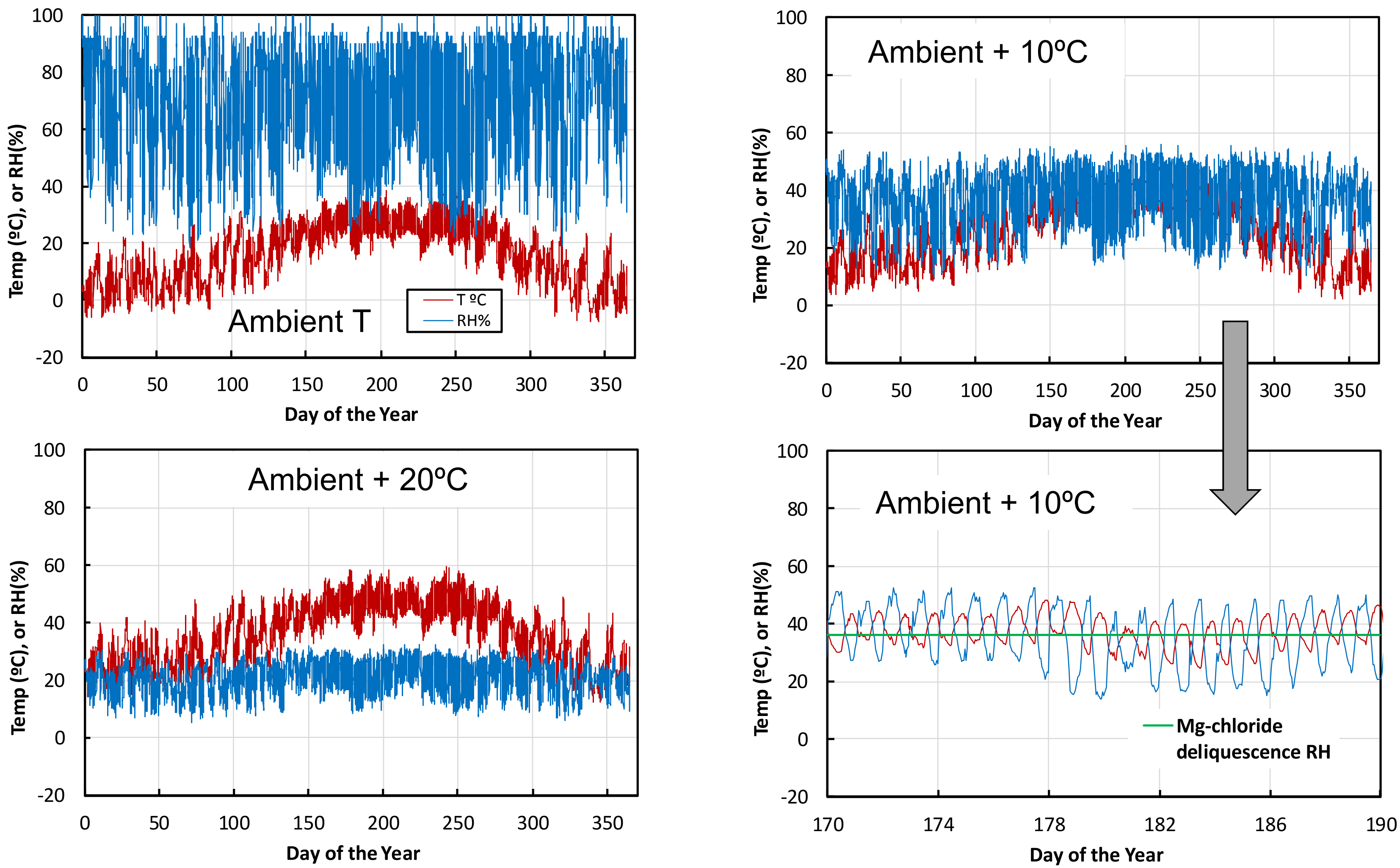
Timing of corrosion initiation—point in time at which RH$_L$ is first reached.

Summing time when RH>RH$_L$ provides "time of wetness"; time when corrosion can actually occur.

Both vary with canister surface location.

Effect of Elevated Canister Surface Temperatures

Weather Data for Arkansas Nuclear 1 Power Station (2013)



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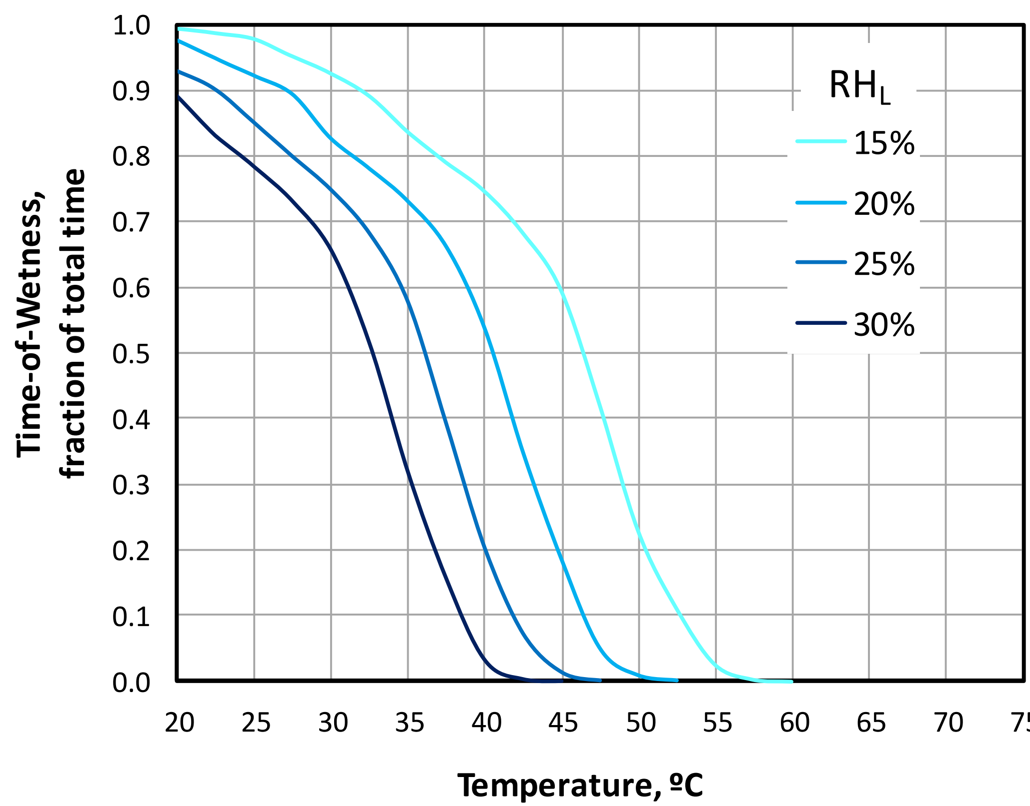
Concept of Time-of-Wetness

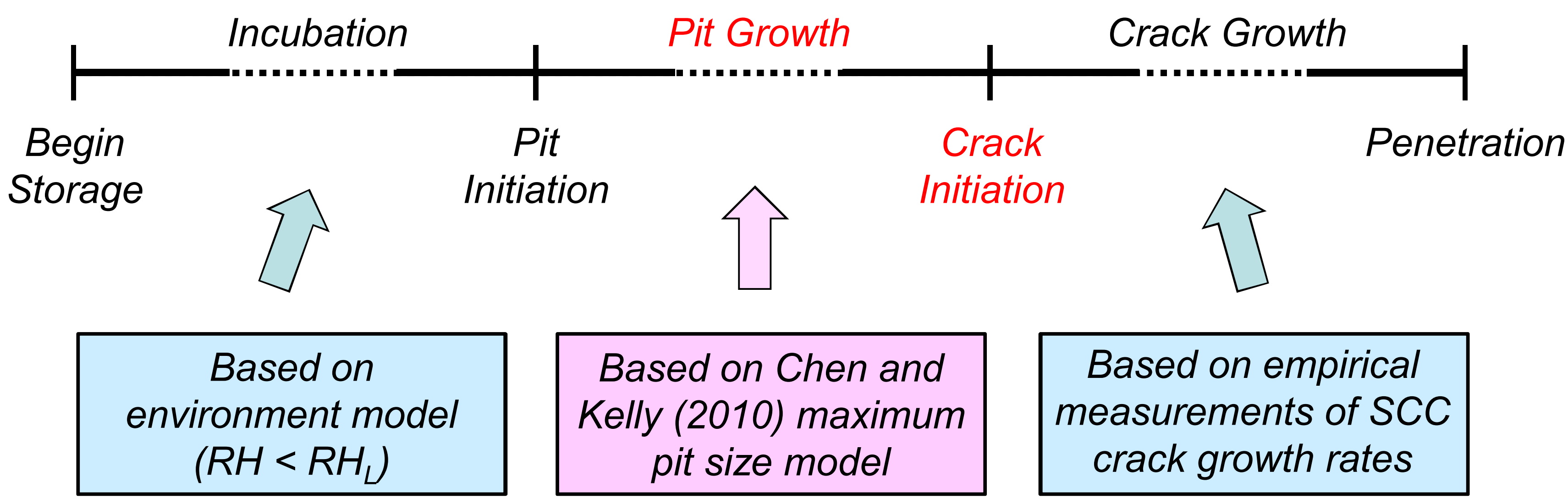
Time-of-wetness:

- Corrosion only occurs when a threshold RH (RH_L) is exceeded.
- Corrosion can start-stop on a daily or seasonal basis, as a function of changing T, AH
- Total integrated time when aqueous conditions exist is time-of-wetness
- Rationale: External cathode, required to support corrosion at the anode, is only present when aqueous conditions exist on the metal surface.
- Widely accepted approach (ASTM procedure G84-89). *However, many reasons to challenge time of wetness on a conceptual basis. (e.g., Schindelholz et al., 2013)*

Calculated time-of-wetness as a function of canister surface temperature and RH_L. Based on one year of weather data from the Oceanside Municipal Airport near the San Onofre ISFSI.



Pit Growth and SCC Crack Initiation



Pitting and SCC Crack Initiation

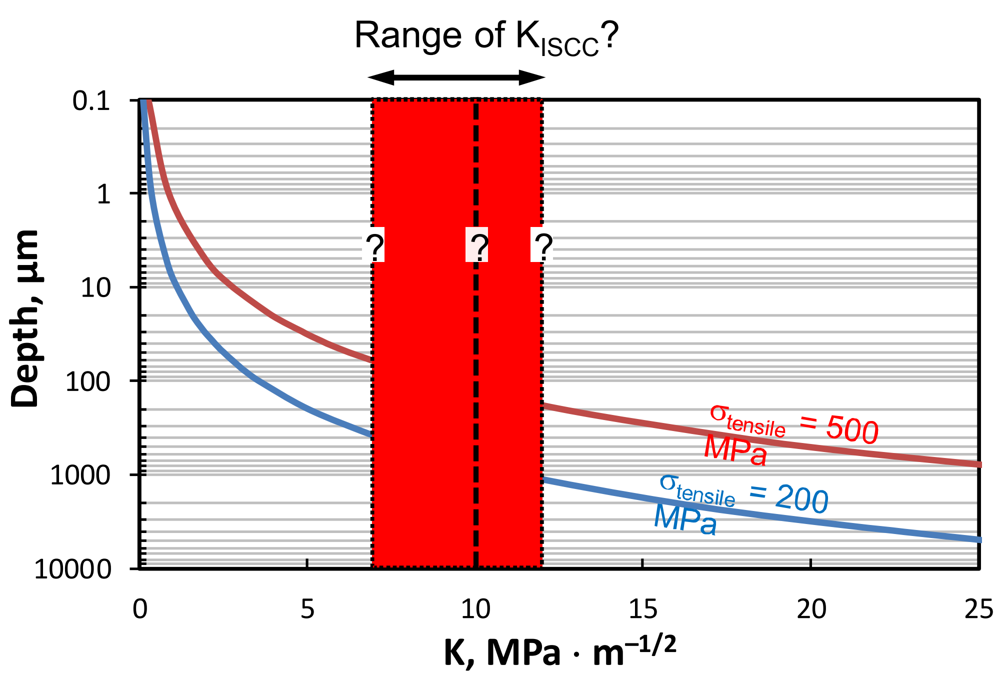
Pitting and SCC crack initiation

- Corrosion starts (pits form) once the RH_L is exceeded.
- *Pits grow over time, but for any given set of environmental conditions (T, RH, salt load), pit depths reach a limiting value.*
- Maximum pit depth model (Chen and Kelly 2010) can be used to calculate the maximum possible pit depth over time as a function of environmental parameters (temperature, RH, and deposited salt density).
- 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 can be estimated using the “Kondo criterion” (Kondo, 1985).

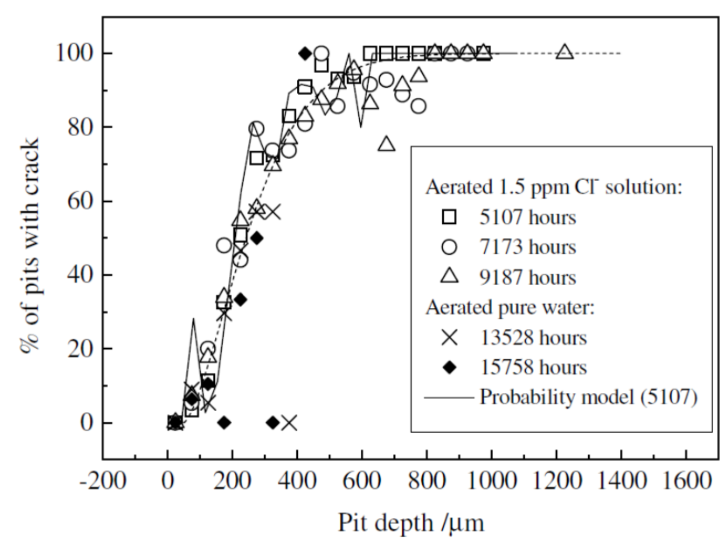
Crack Initiation: the "Kondo Criterion"

"Kondo criterion". **Pit-to-crack transition** occurs when the pit depth is equal to the depth at which an equivalent-depth SCC crack would have a crack-tip stress intensity factor (*K*) that exceeds K_{ISCC} for the metal.

- *K* is calculated as a function of crack depth using a sampled through-wall stress profile.
- K_{ISCC} is sampled from a compilation of literature values.



Experimental data from Turnbull et al. (2006)

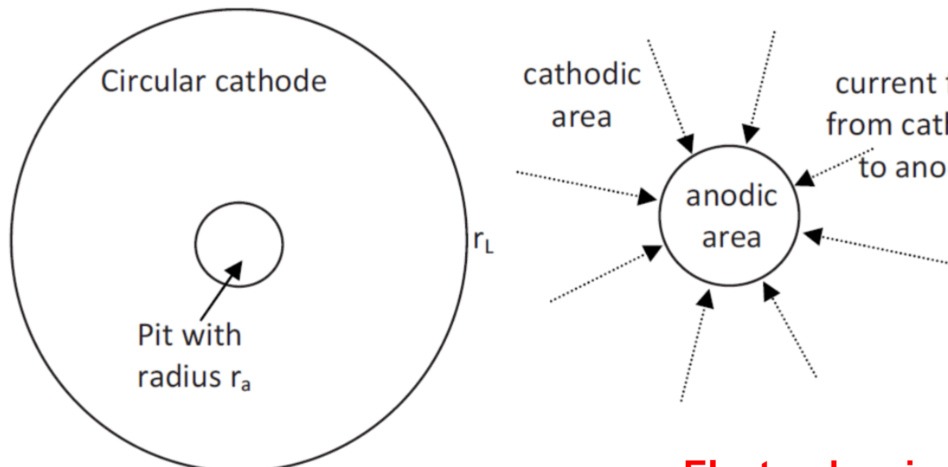


Stochastic process: As a pit deepens, the likelihood of a crack initiating increases.

Maximum Pit Size Model

Chen and Kelly (2010): Max pit size is a function of the maximum cathode current.

Pits are modeled as being hemispherical, and stifle once the pit becomes so large that the anodic current requirement exceeds the available cathode current.



Max. cathode current

Brine conductivity

Brine layer thickness

Electrochemical term (from cathodic polarization curve)

$$\ln I_{c,max} = \frac{4\pi k W_L \Delta E_{max}}{I_{c,max}} + ln\left[\frac{\pi e r_a^2 \int_{E_{corr}}^{E_{rp}} (I_c - I_p) dE}{\Delta E_{max}}\right]$$

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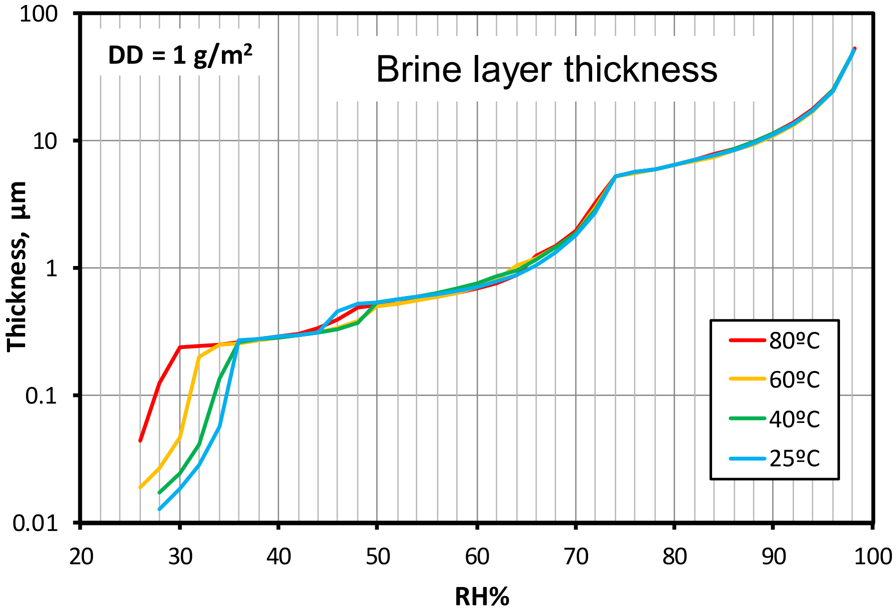
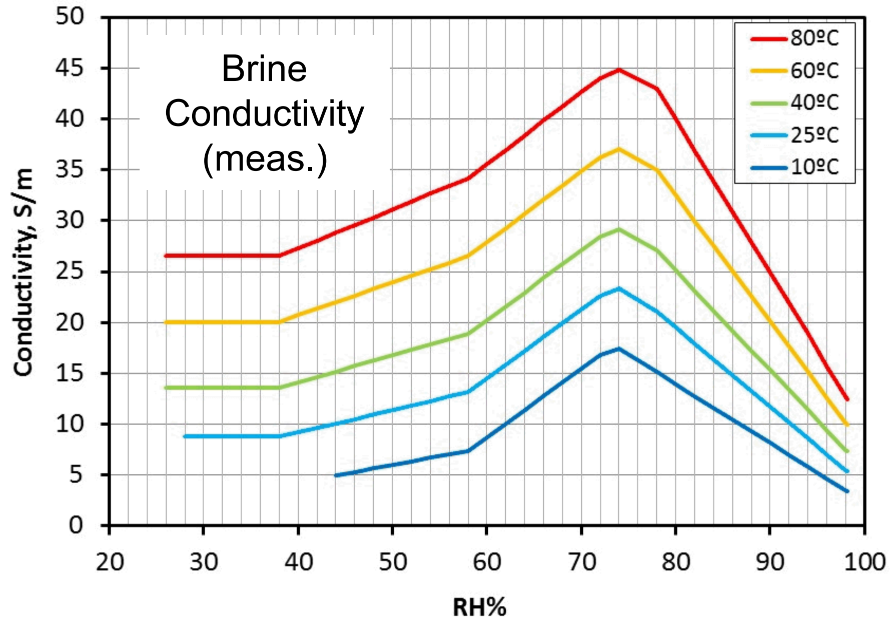
Parameterizing the Model for Sea-Salt Brines

Values are based on geochemical modeling, literature data, and measured data for brine densities and conductivities (4 brines, from 98-38% RH).





Original *Chen and Kelly* (2010) model: Based on NaCl brines (dry out at ~75% RH).

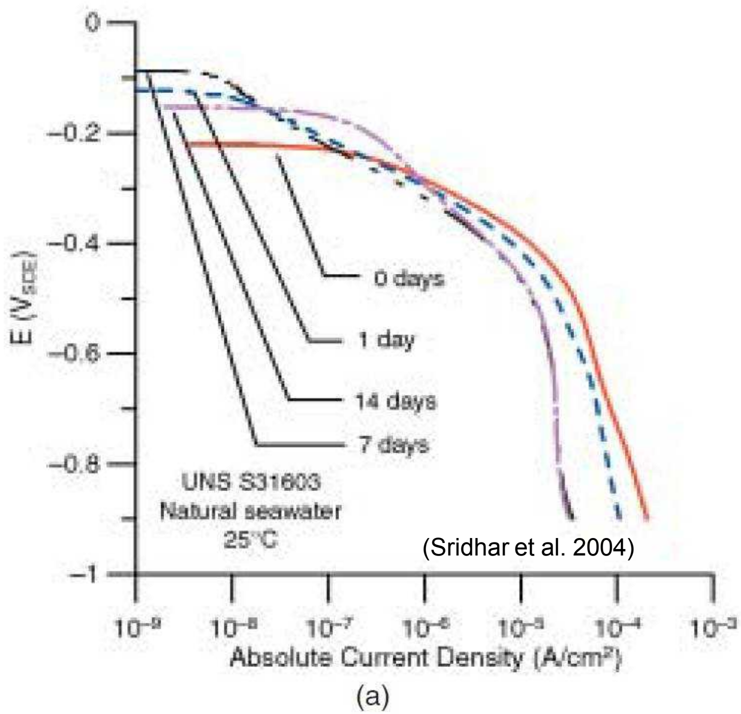
Assumption: *Brine forms a continuous brine layer.* This seems unlikely at low salt loads or low RH values. However:

- Experimental data suggest that the cathode can extend well beyond the perimeter of salt grains (Schindelholz et al., 2013).
- Insoluble dust particles will increase brine film continuity via capillary effects.

Other Needed Data:

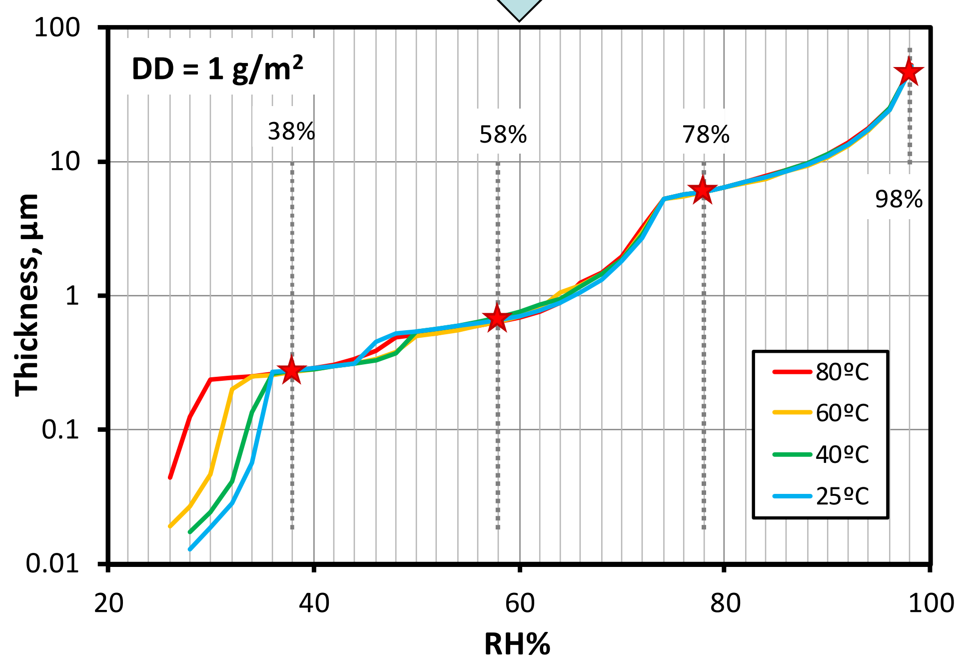
Cathodic Polarization Curves and electrochemical parameters for concentrated brines

Currently, limited available data:
Cathodic polarization curve for seawater at 25ºC, 316 SS (Sridhar et al. 2004)



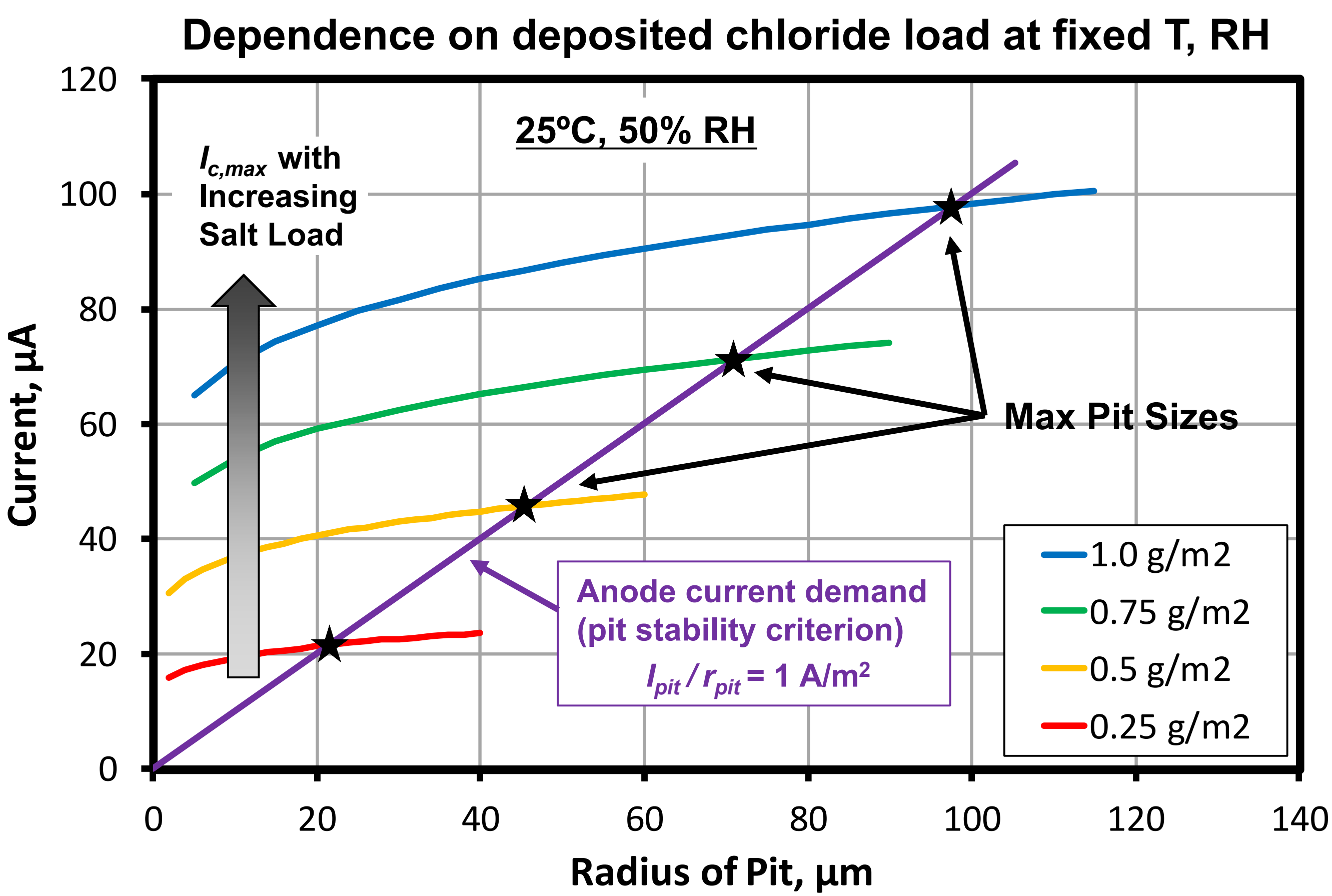
To characterize variability in cathode kinetics with brine composition, SNL is measuring polarization curves and other electrochemical parameters in four brines corresponding to:

- Unevaporated Seawater (98% RH)
- Evap. to 78% RH
- Evap. to 58% RH
- Evap. to 38% RH



Determining Maximum Pit Size

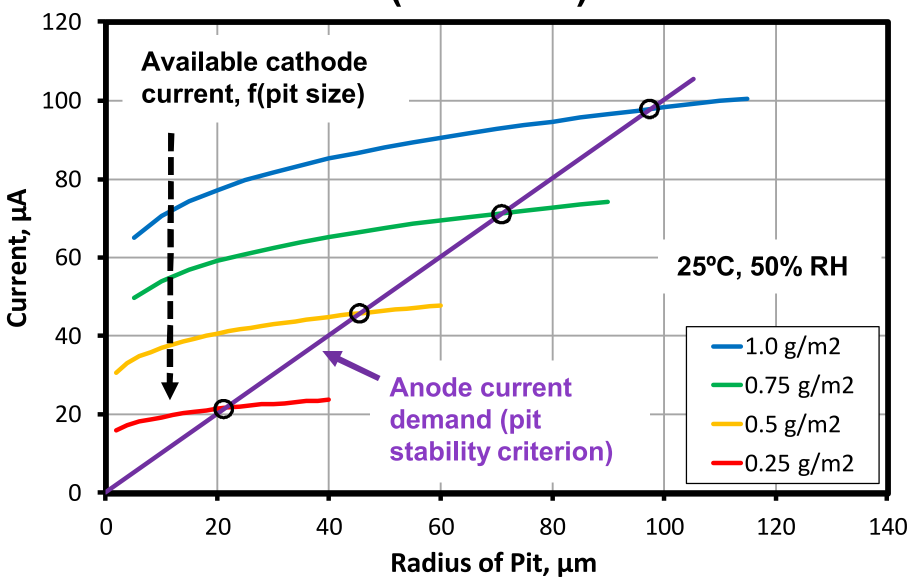
Maximum pit size is determined the intersection of the anode current demand and the available cathode current.



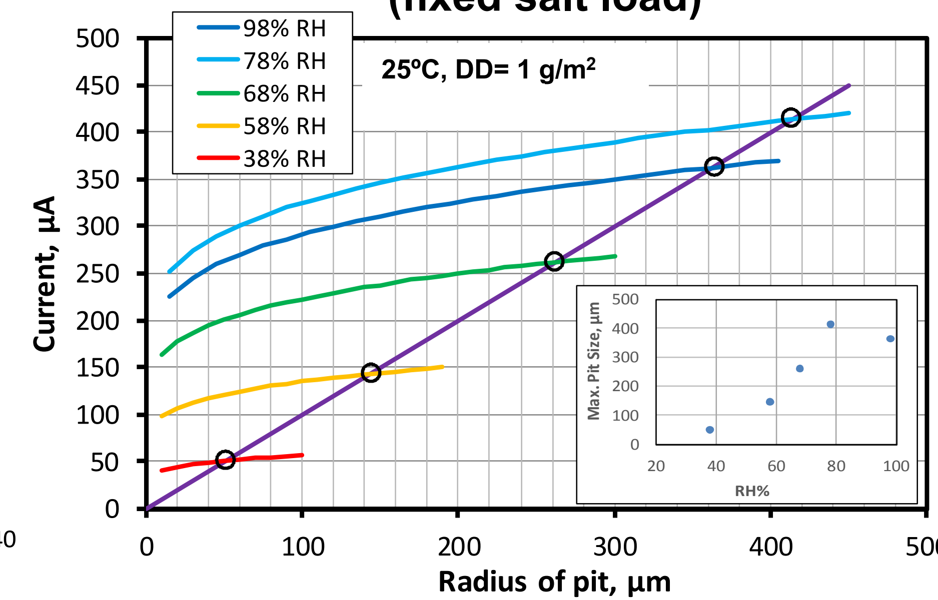
Example: Calculated Maximum Pit Sizes

Examples of Chen and Kelly maximum pit size model results

Dependence on deposited chloride load (fixed RH)



Dependence on relative humidity (fixed salt load)



Conclusion: Strong pit size dependence on salt surface load and RH (below 75-80% RH).

Caveat: Results are based on seawater (98% RH) cathodic polarization data---only available data at this time

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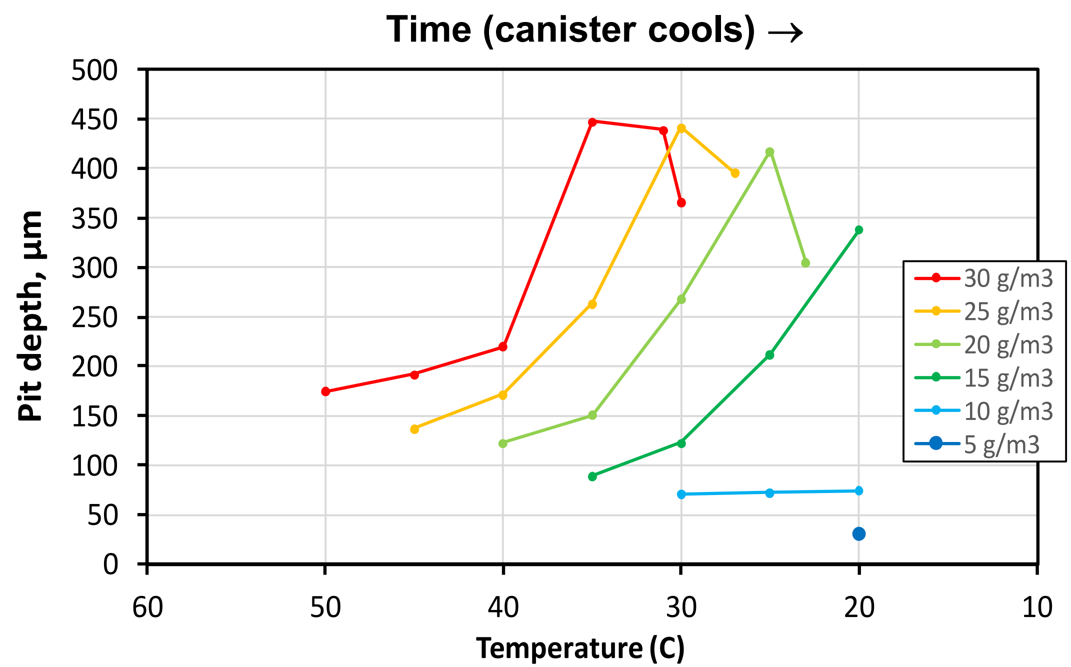
Model Results

As the canister cools, T drops, and RH increases

Assume constant AH: As T is varied, RH modified to hold AH constant. Salt load held constant throughout.

- **Largest pits possible at temperatures corresponding 75-85% RH**
  - *Higher T = lower RH, thinner brine films*
  - *Lower T = higher RH, more dilute solutions.*
- **Overall trends:**
  - *As the canister cools, maximum possible pit size increases*
  - *Increasing salt load with time will also result in larger pits*

Risk of meeting Kondo Criterion and initiating SCC will increase with time.



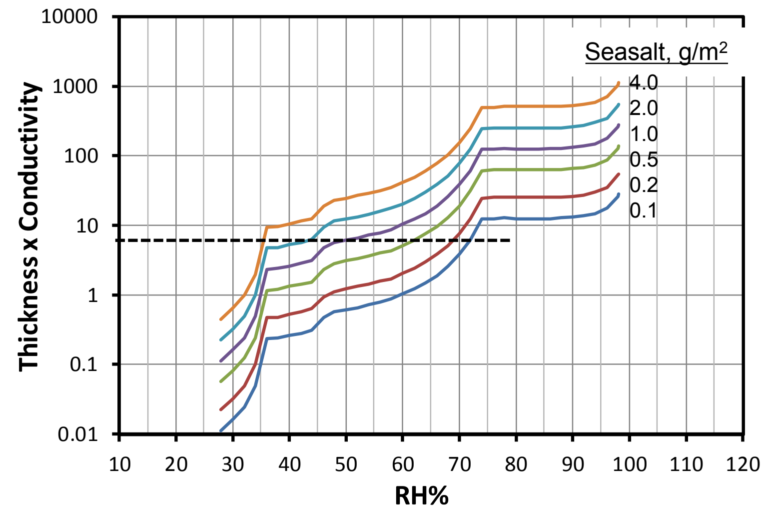
Caveat: Electrochemical terms are held constant. Realistically, they vary with brine composition and temperature.

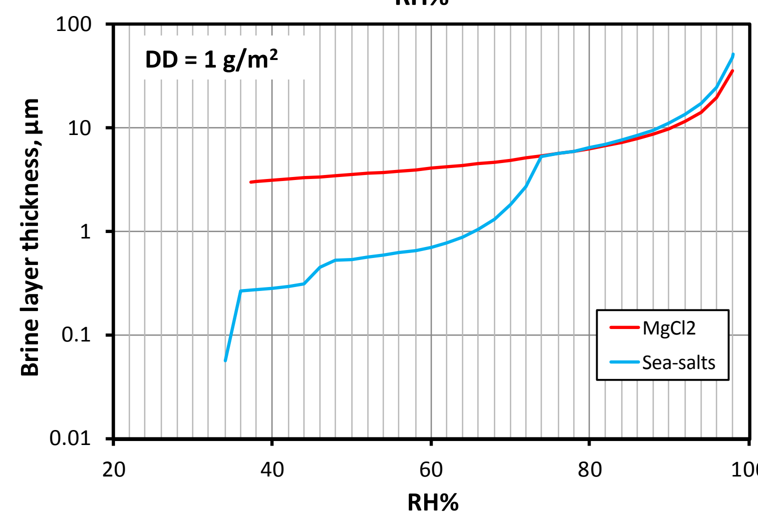
(Woldemedhin et al. 2014, 2015; Srinivasan et al. 2015)

Implications of Combining Maximum Pit Size Model and Kondo Criterion

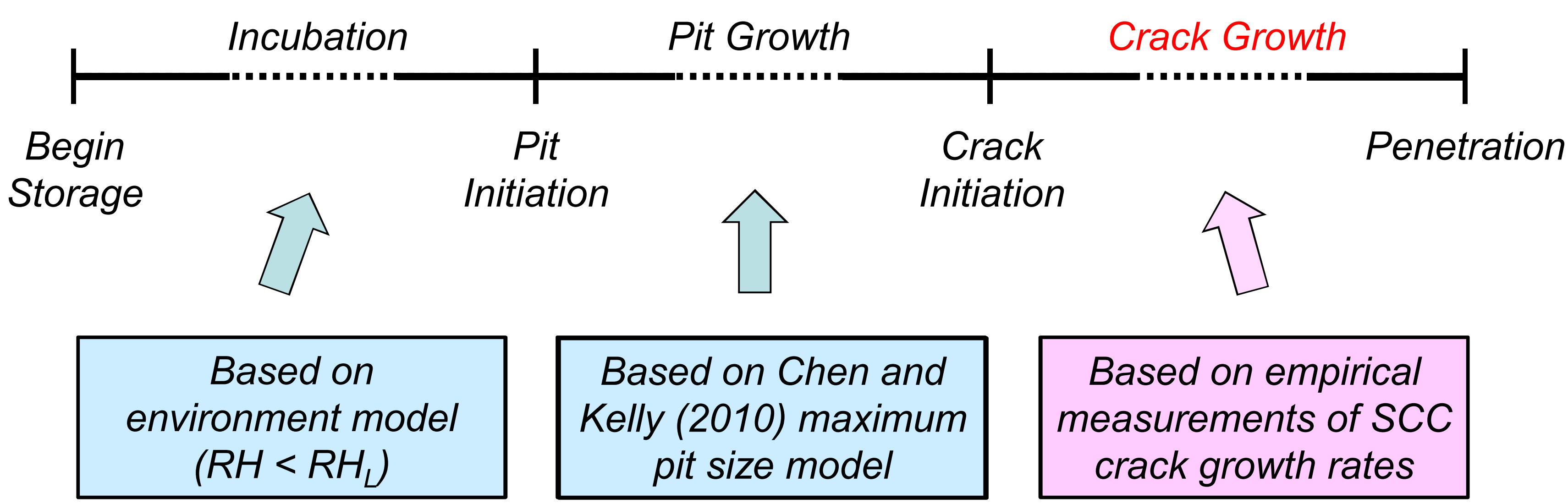
- **Brine volume/concentration will control pit size and hence, initiation of SCC.**
  - RH threshold for SCC will decrease as salt load increases.
  - Higher RH values will increase brine volume and cathode size, resulting in SCC at lower salt loads. This matches experimental data, at least, at low RH values (NRC 2014, Table 3-3)
- **Magnesium chloride is a poor analog for sea-salts in corrosion experiments—for a given chloride load, the brine volume is much higher)**

Large variations in literature values for RH thresholds for SCC may be due to failure to control these parameters independently.





Model for SCC Crack Growth



Stress Corrosion Cracking Model:
Crack growth

Model for crack growth:

- In general:

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

- Implemented a version incorporating only effect of T, K:

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

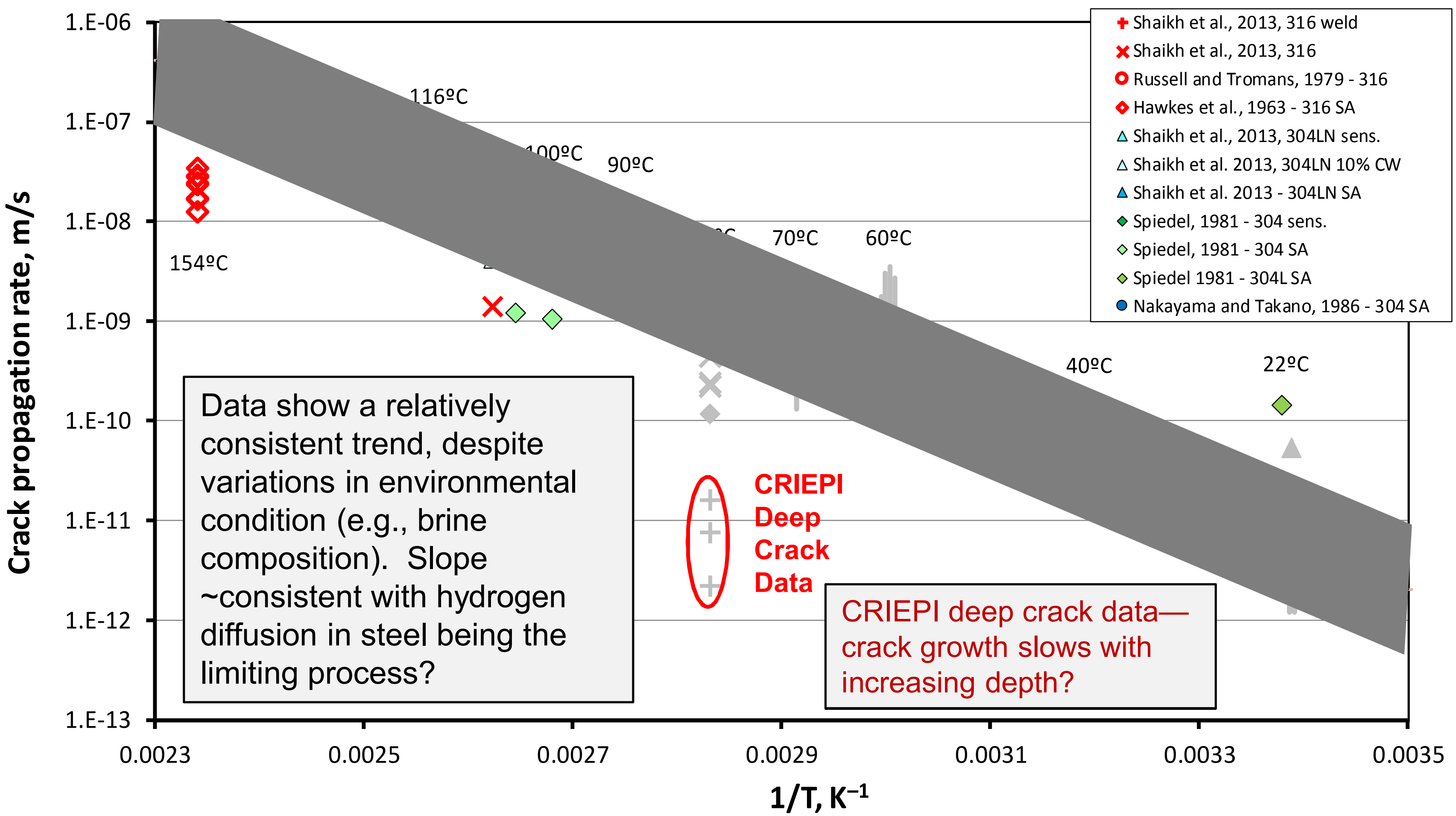
dx_{crack}/dt = crack growth rate
α_{crack} = crack growth amplitude
Q = activation energy for g=crack growth
R = universal gas constant (8.314 J mol^{-1} K^{-1})
T = temperature (K) of interest
T_{ref} = reference temperature (K) at which α was derived.
K = crack tip stress intensity factor
K_{th} = threshold stress for SCC
β_{crack} = stress intensity factor exponent.

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

$\sigma_{applied}$ = tensile stress
Y = shape factor
x_{crack} = depth

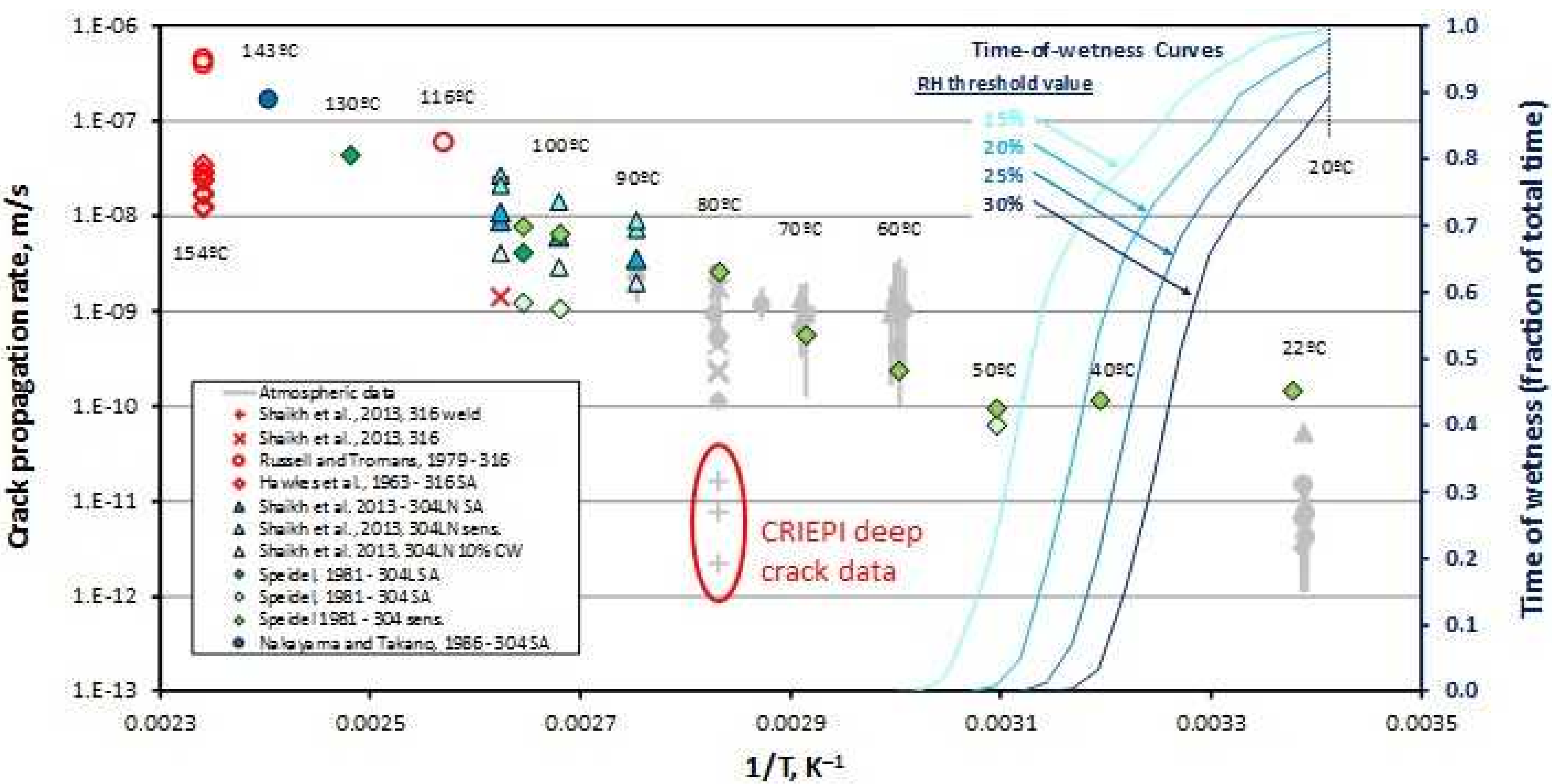
Literature Crack Growth Rates

Data for 304 SS and 316 SS



Role of Time-of-Wetness

Overall Crack Penetration Rate = Crack Growth Rate × Time-of-wetness



Intended Model Uses

- **Assessing risk of SCC initiation and penetration as a function of:**
  - Estimated heat decay curve (canister surface temperatures)
  - ISFSI location (weather parameters, salt deposition rates)
  - Canister surface location (variation in T, salt load)
- **Support development of appropriate ISFSI Aging Management Plans: informing required canister inspections for SCC**
  - Maps of at-risk locations on the canister surfaces
  - Determination of necessary SCC inspection intervals

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Model Needs and Proposed Future Work

Environment

- Lacking information on dust/salt compositions and deposited salt loads
- Lack of SNF- and storage system-specific thermal models

Pitting and SCC initiation

- Maximum pit size model insufficiently parameterized
  - Poor understanding of pit growth processes at low RH (below 75% RH); effect of discontinuous brine films, nature of cathode
  - Lack of electrochemical data for concentrated sea-salt brines
  - Effect of surface roughness or surface dust on brine film continuity (capillary condensation)
- Factors affecting SCC initiation
  - Capturing local stresses, plastic deformation, local topography (how deep a pit is really needed to initiate a crack on a canister?)

Crack growth

- Factors controlling crack growth under thin films: do crack growth rates slow as cracks deepen?

Collaborations

- **Colorado School of Mines Integrated Research Project**
  - Controls on pitting and crack initiation
  - Weld residial stress models
  - Characterization (microstructure, stresses) of weld zones
  - Identification of most susceptible HAZ regions
  - Crack growth rates and controls (NCSU)
- **Ohio State University (EFRC)**
  - Pit-to-crack transition, and effects of limited salt load on crack growth
- **Southwest Research Station, Savannah River National Lab**
  - Attempts to measure crack growth rates under atmospheric SCC conditions

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