



U.S. DEPARTMENT OF  
**ENERGY**

**Nuclear Energy**

SAND2016-11441C

## Fuel Cycle Technologies

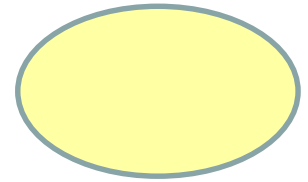
# Evaluating Stress Corrosion Cracking of Spent Nuclear Fuel Storage Canisters

**Charles Bryan**  
**Sandia National Laboratories**  
**Used Fuel Disposition Program**

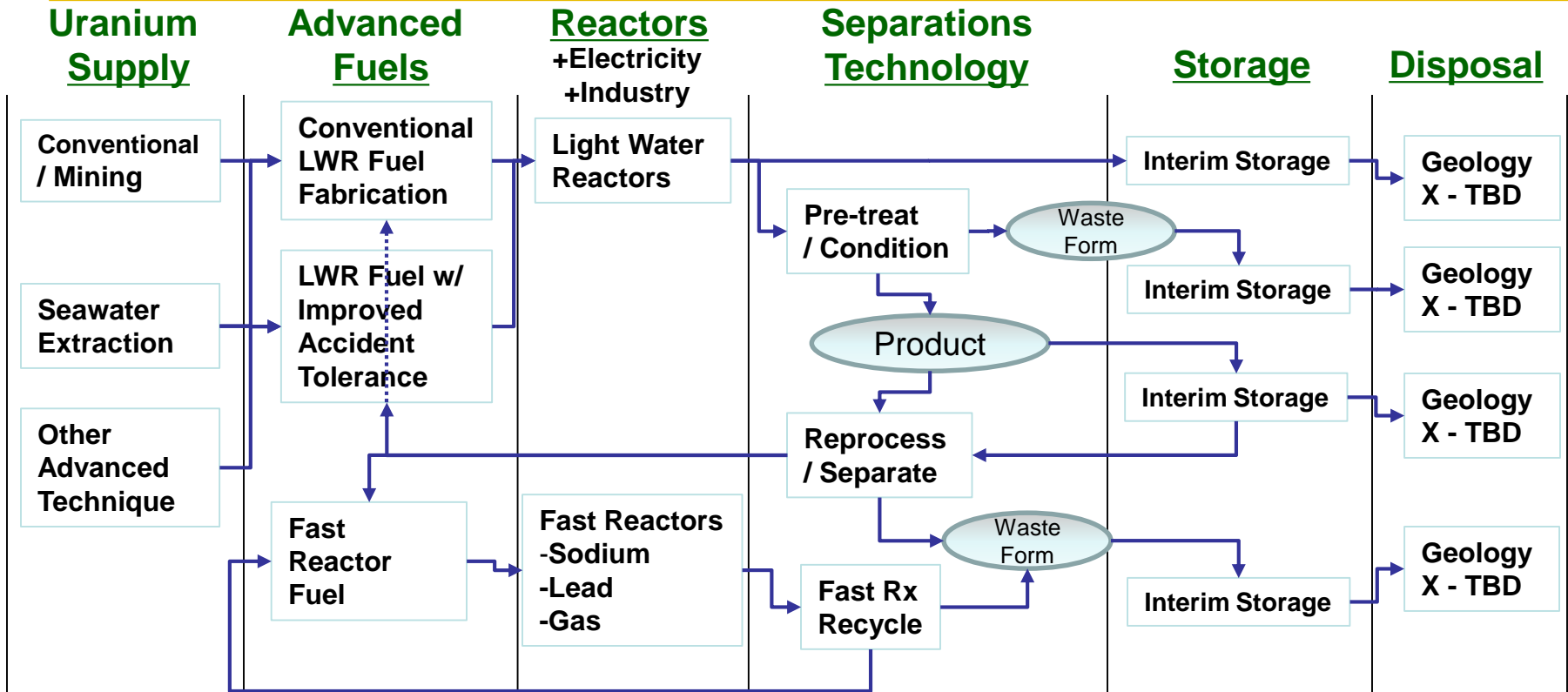
**ANS Winter Meeting, Las Vegas**  
**November 8, 2016**



# Fuel Cycle as a System



## Nuclear Energy



- **Optimized System:** We want the best performance for each step in harmony with other parts of the system
- **Near-Term/Long-Term Balance:** Seek near-term applications while maintaining the long-term objective of a sustainable fuel cycle

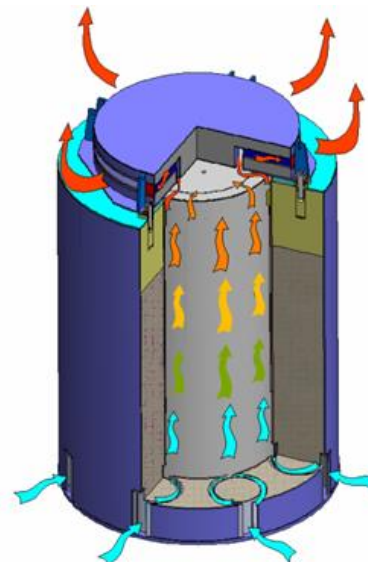
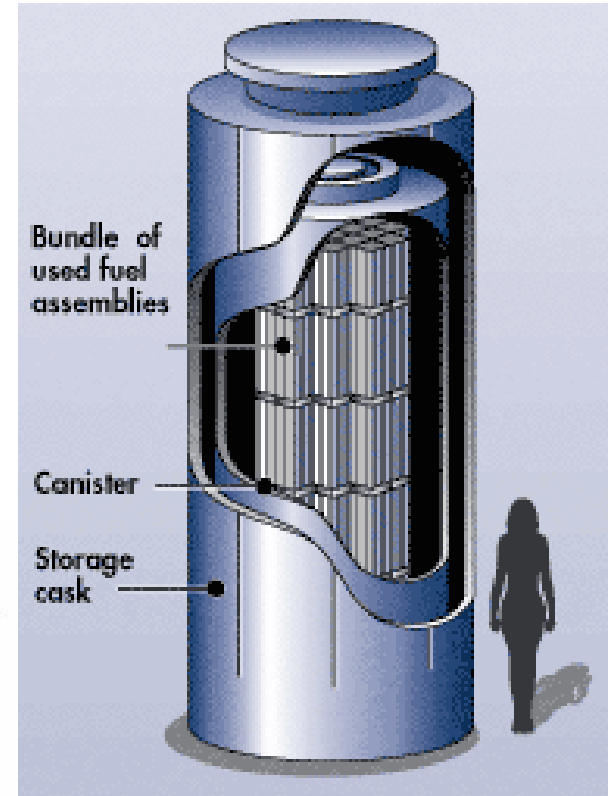


- **United States currently has over 80,000 metric tons of Spent Nuclear Fuel (SNF), about 30% in dry storage systems.**
- **The dry storage systems are intended as interim storage until a permanent disposal site is developed. Until recently, systems were licensed for 20 years with an additional 20 years upon license renewal; in 2011, CFR was modified to allow for initial 40 year license period, with an additional 40 year renewal.**
- **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.**
- **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.**



# 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.

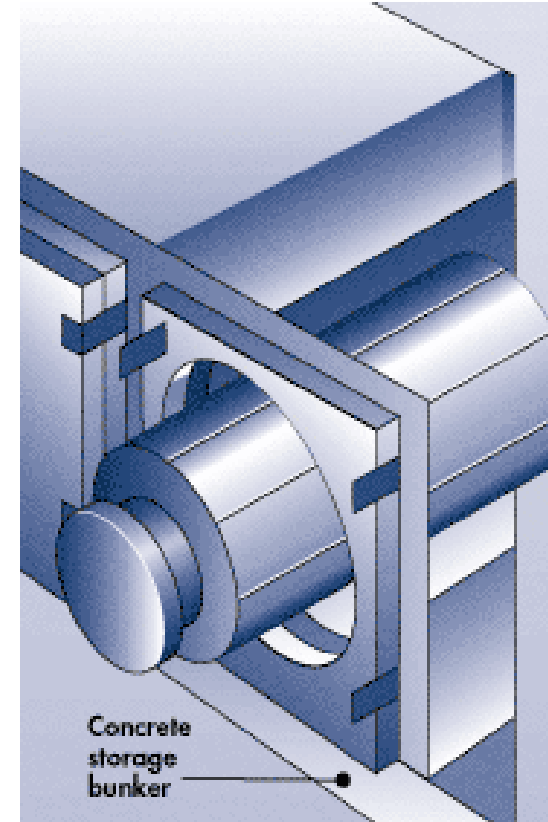


Pathway for air flow through the overpack.



# 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, flows up and around the canister, and exits through vents on the roof.



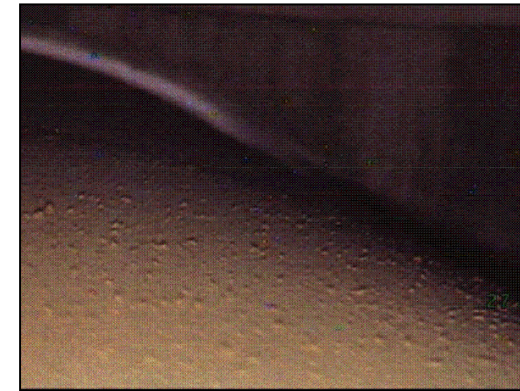
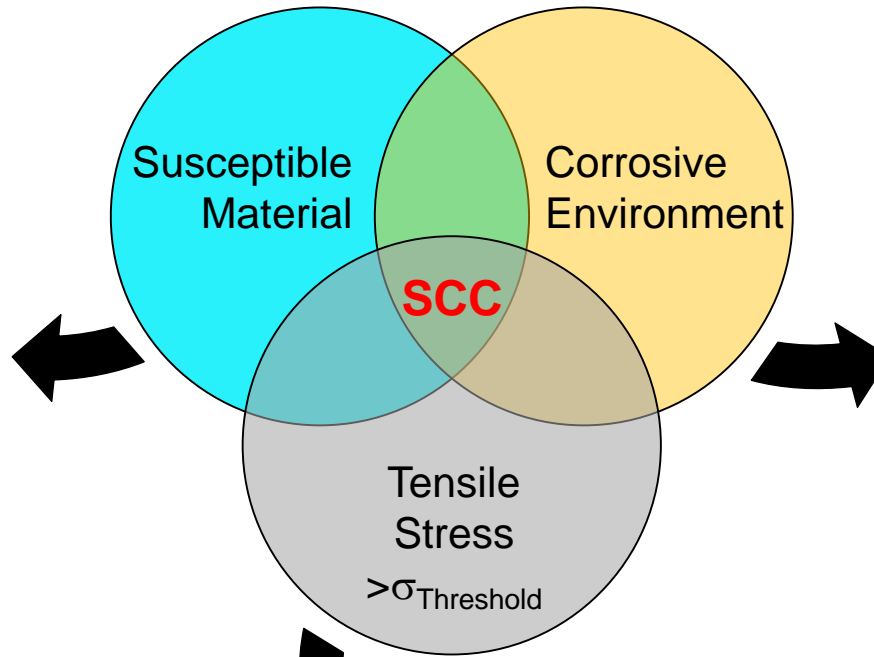


# Criteria for Stress Corrosion Cracking

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

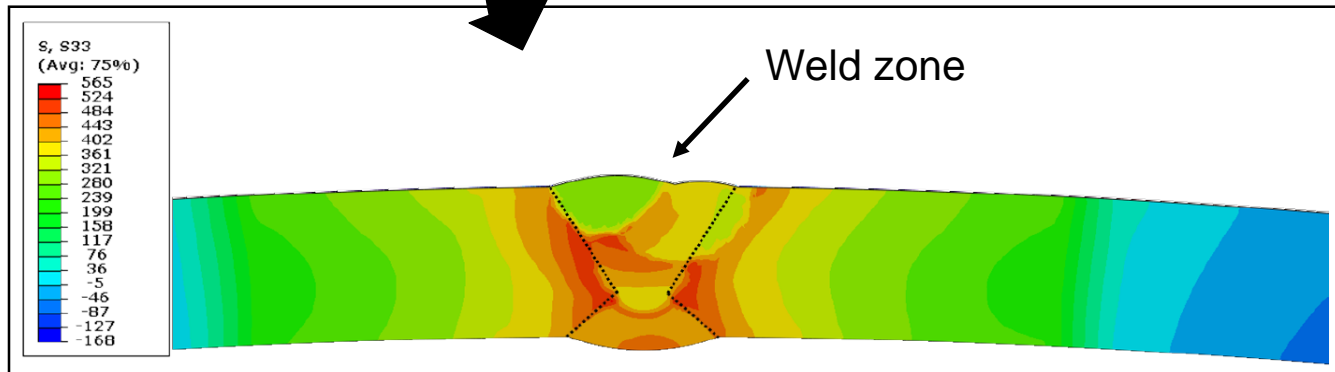


Weld zone, Ranor 304 SS plate



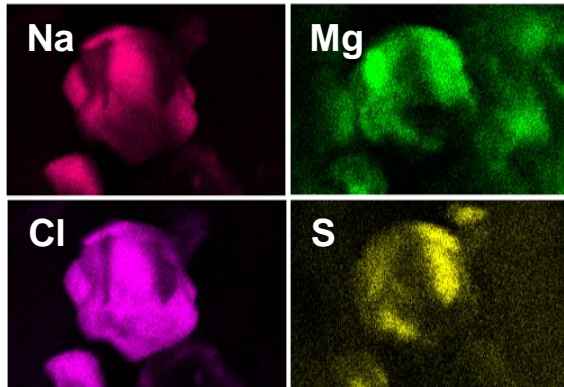
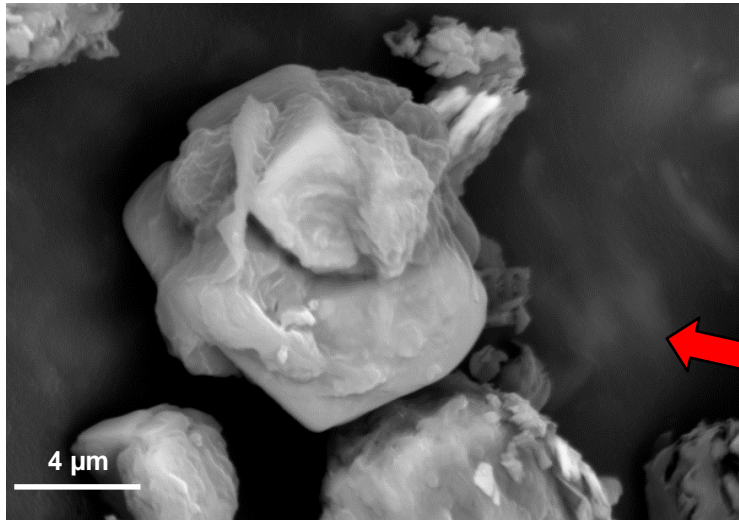
Dust on storage canister surface at Calvert Cliffs (EPRI 2014)

NRC (2013) modeled weld residual stresses

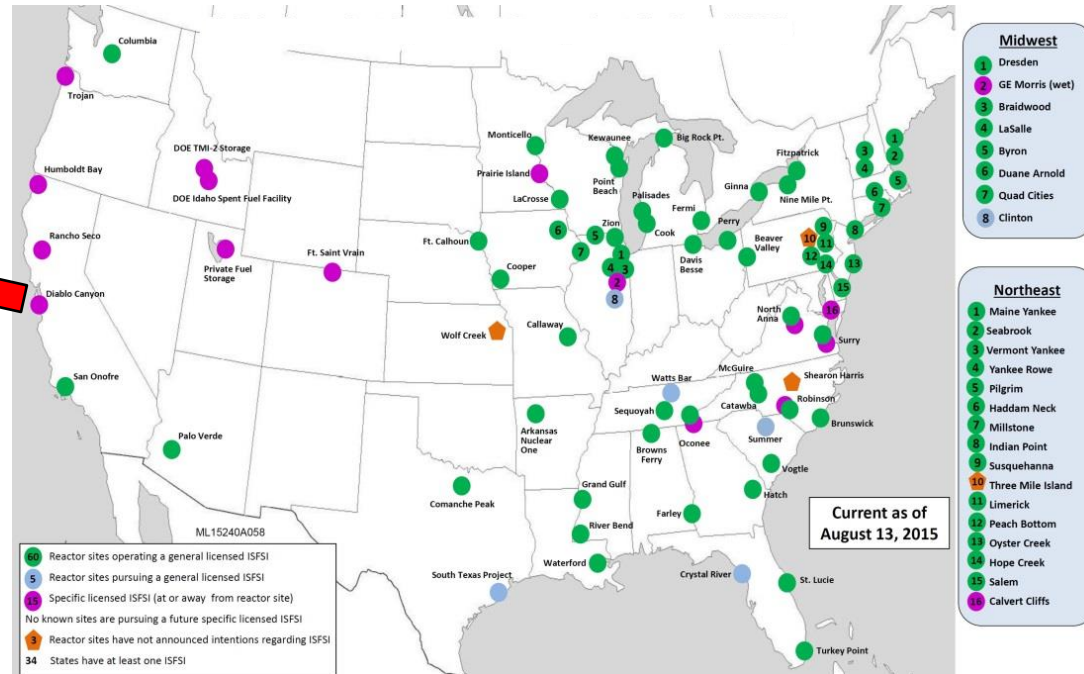




Sea-salt aggregate on Diablo Canyon ISFSI storage canister






Locations of U.S. Independent Spent Fuel Storage Installations



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



# Susceptible Material

- **304 SS is known to be susceptible to atmospheric SCC. Susceptibility is a function of many factors:**
- **Degree of sensitization** 
- **Degree of cold work**
- **Surface finish** 
- **Presence of iron contamination** 
  
- **Experimental testing to date has shown rapid initiation of SCC under some conditions.**



Weld zone, Ranor  
304 SS plate

Rust spots on storage  
canister at Calvert Cliffs

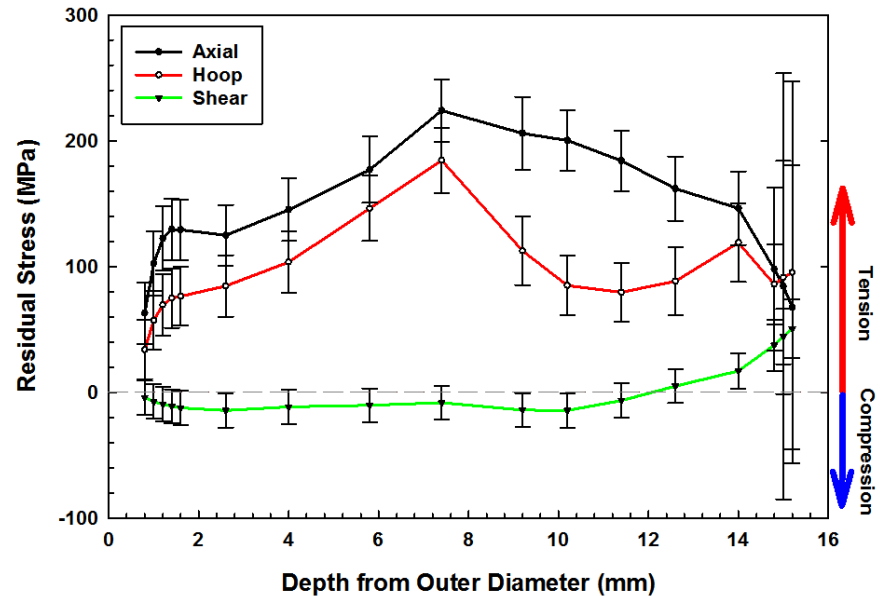




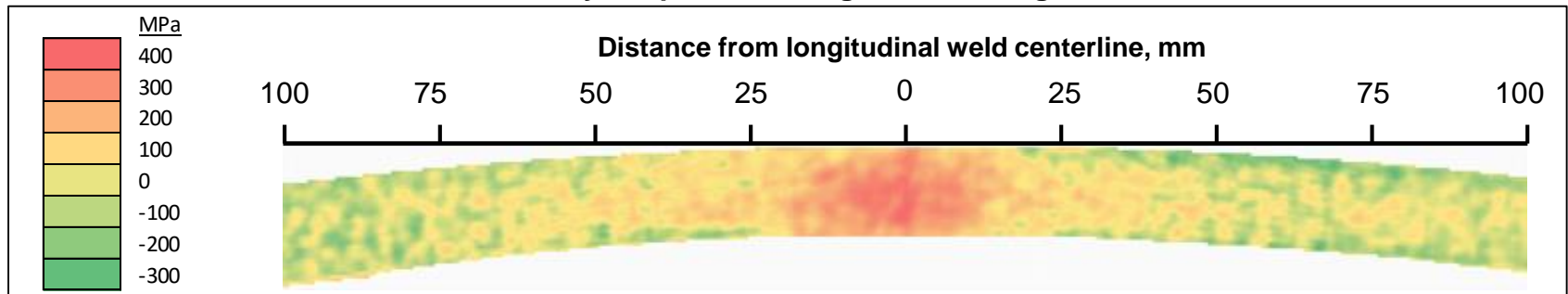
# Tensile Stress

- Sandia full-diameter cylindrical mockup
- Made By Ranor (old supplier to Transnuclear) using identical procedures to real NUHOMS canisters
- Weld residual stresses measured using several methods.
- High through-wall tensile stresses measured at both longitudinal (seam) welds and circumferential welds.
- Highest tensile stresses measured at simulated weld repairs.

Residual stresses measured by deep-hole drilling method, longitudinal weld heat-affected zone

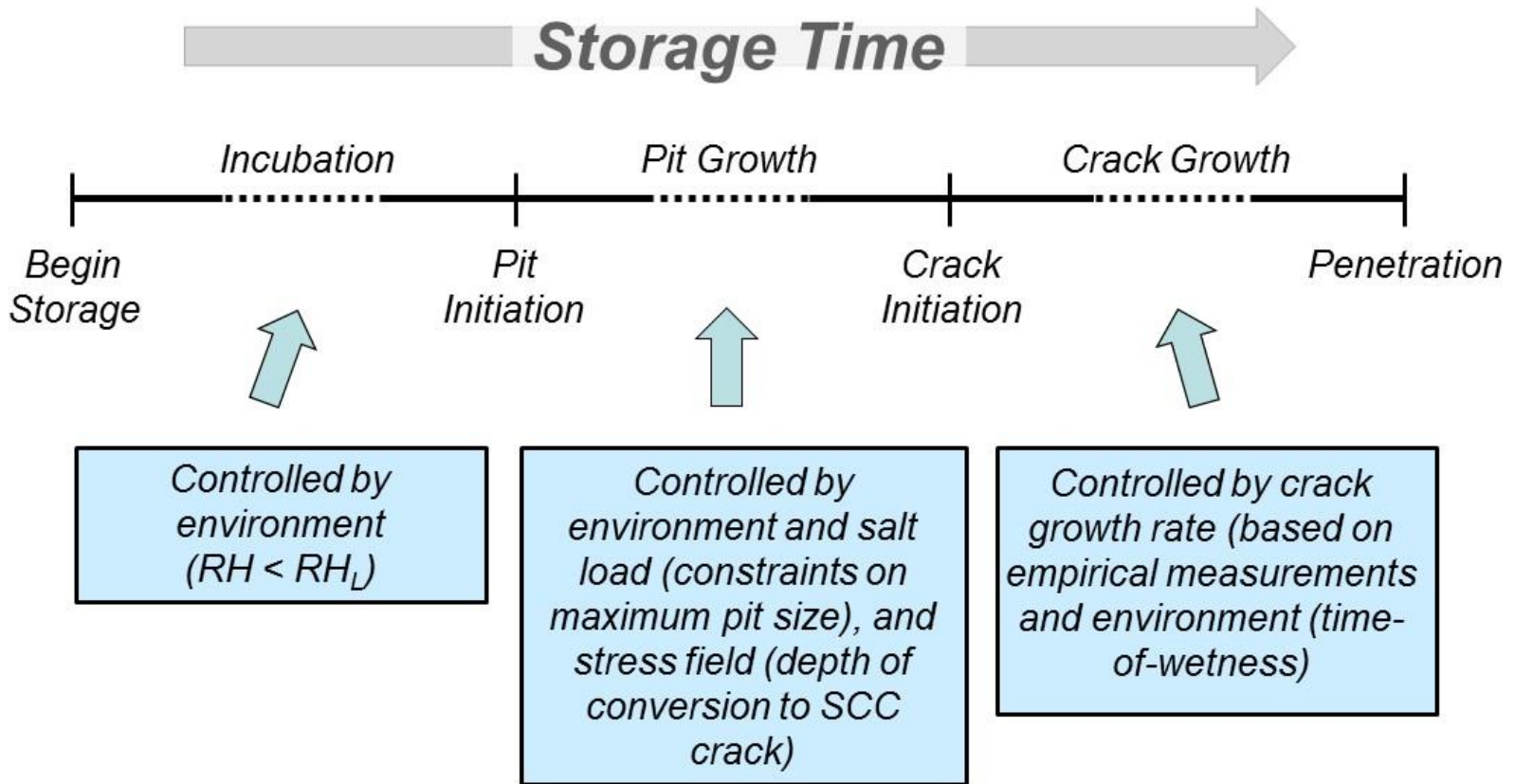


Residual stresses measured by deep-hole drilling method, longitudinal weld heat-affected zone





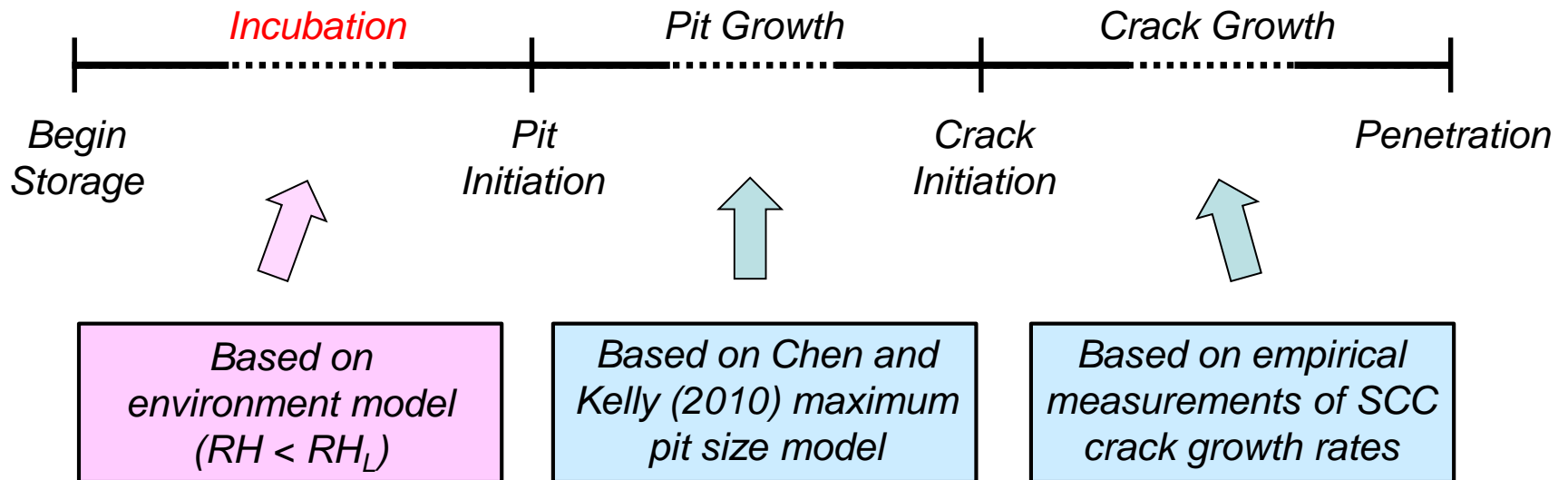
# General Timeline for SCC Initiation and Penetration



Used  
Fuel  
Disposition

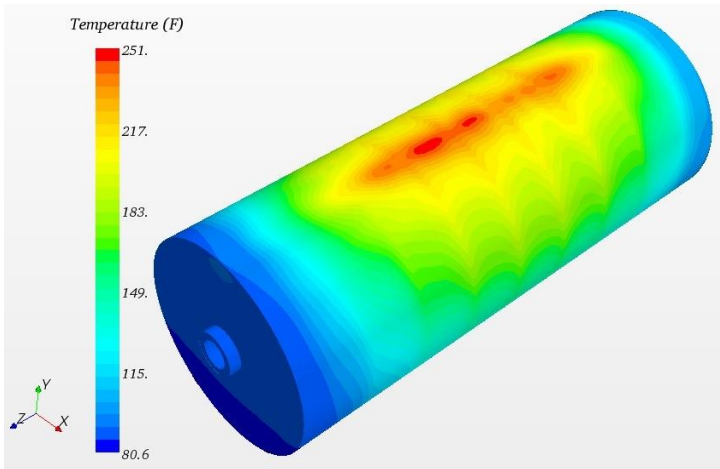
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## Canister Surface Environment

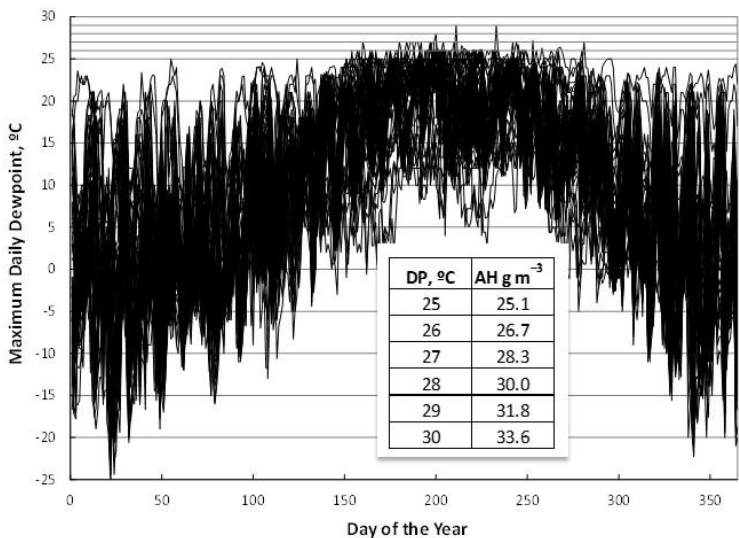




# Incubation Period



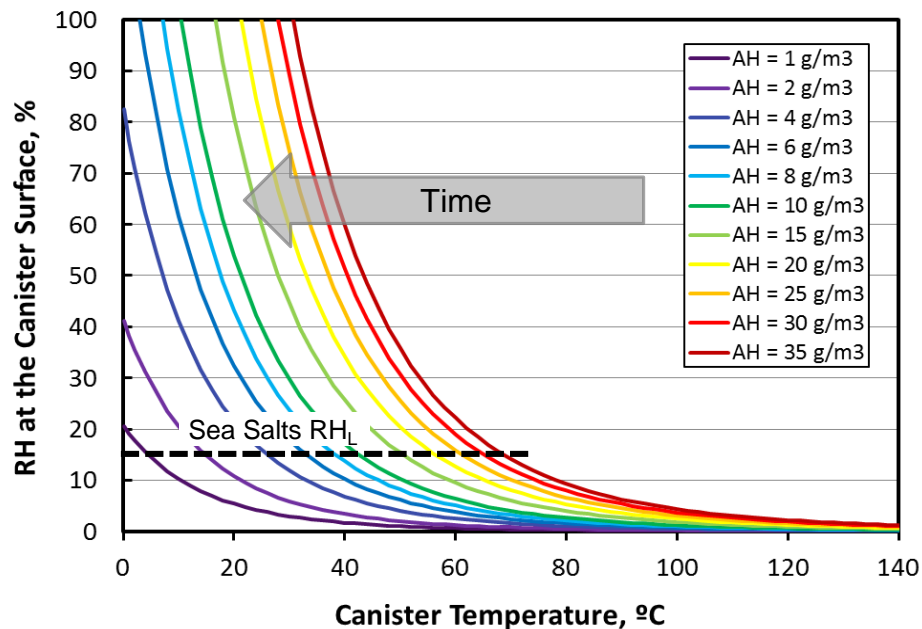
Surface temperature estimates



Site-specific National Weather Service data: daily and seasonal variations in 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  $RH_L$  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.**

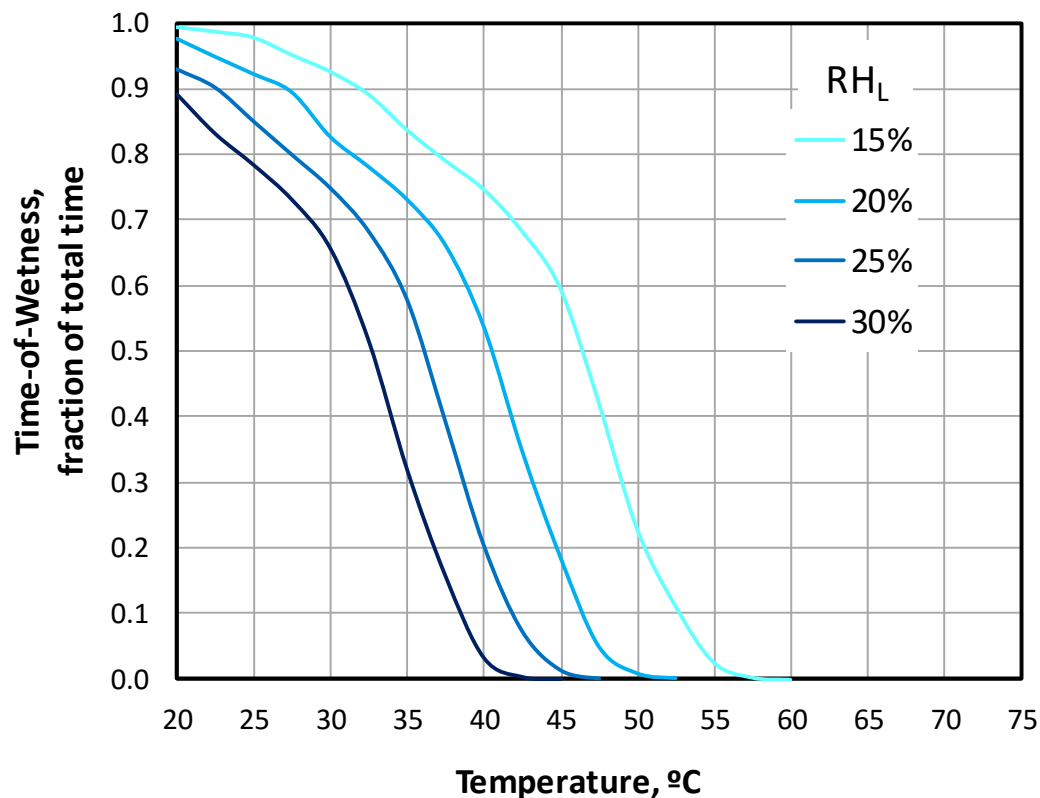


# Concept of Time-of-Wetness

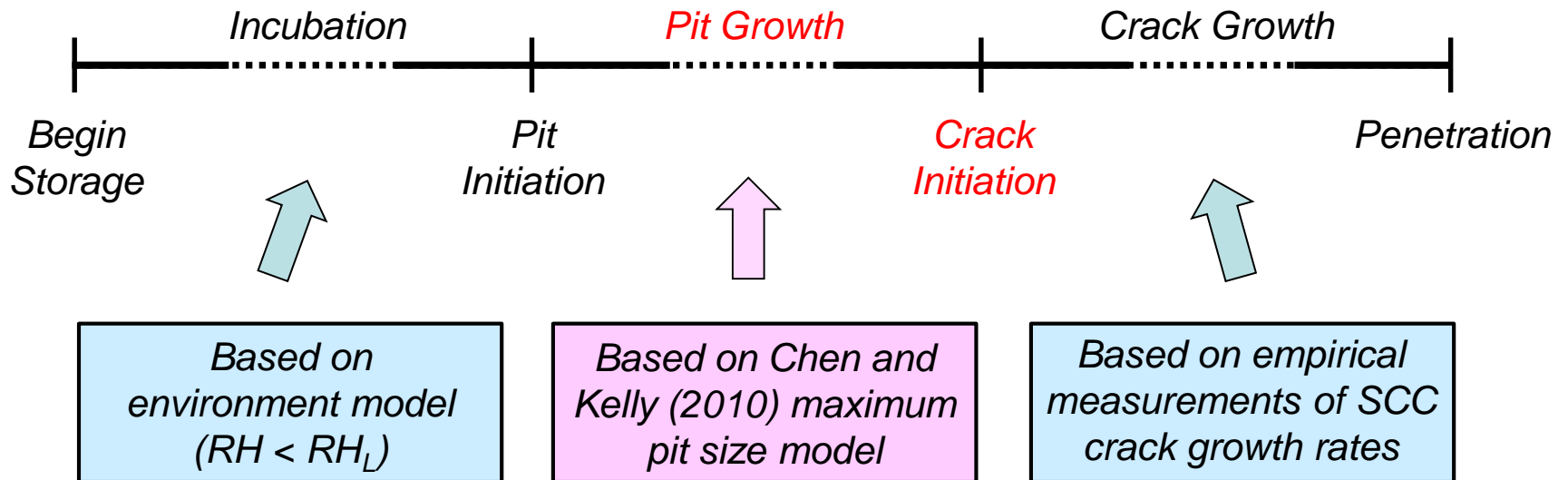
## Time-of-wetness:

- 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

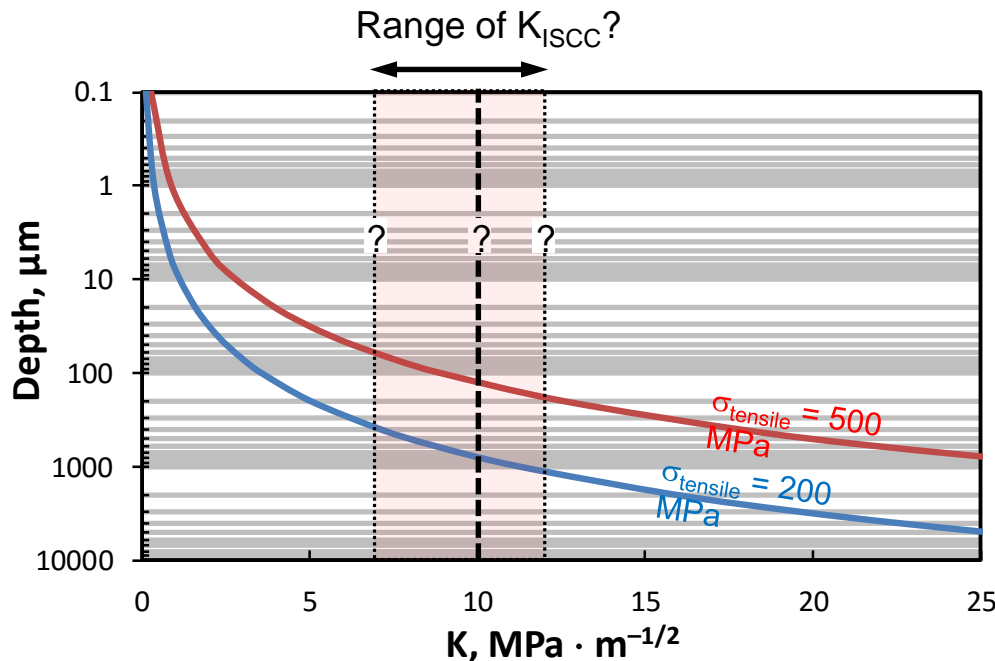
- Corrosion starts (pits form) once the  $RH_L$  is exceeded.
- *Pits grow over time, but for any given set of environmental conditions ( $T$ ,  $RH$ , salt load), pit depths reach a limiting value.*
- Maximum pit depth model (Chen and Kelly 2010) can be used to calculate the maximum possible pit depth 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).



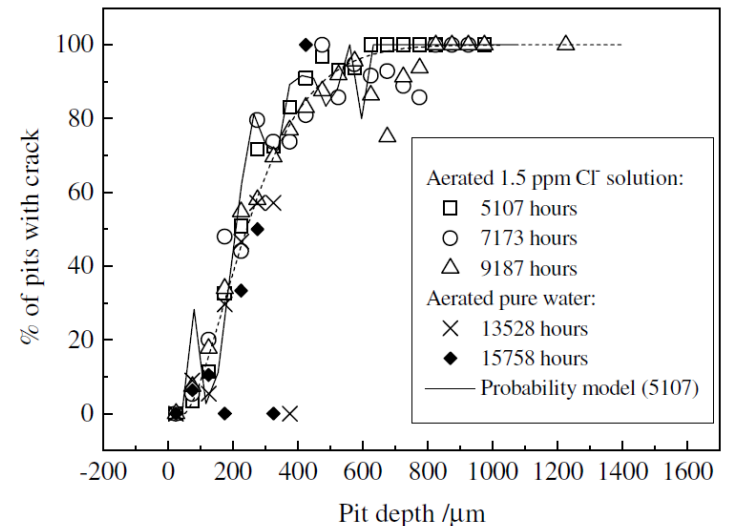
# Crack Initiation: the “Kondo Criterion”

“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)



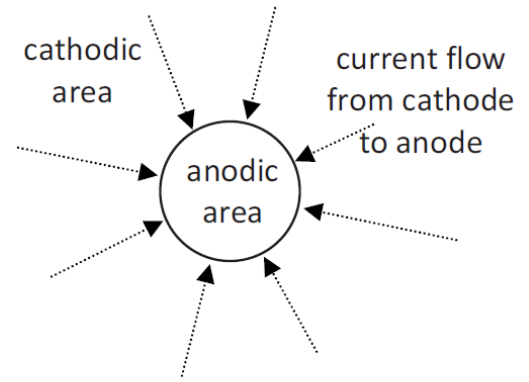
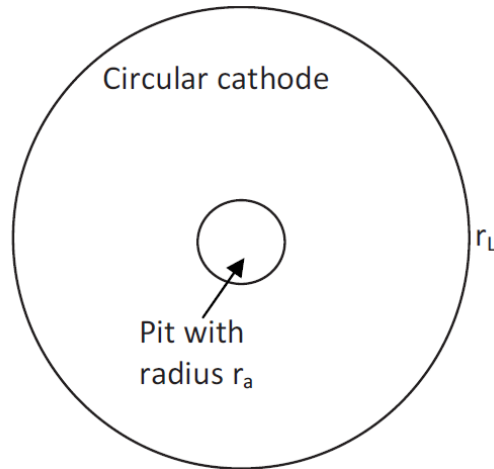
Stochastic process: As a pit deepens, the likelihood of a crack initiating increases.



# 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.



**Electrochemical term (from cathodic polarization curve)**

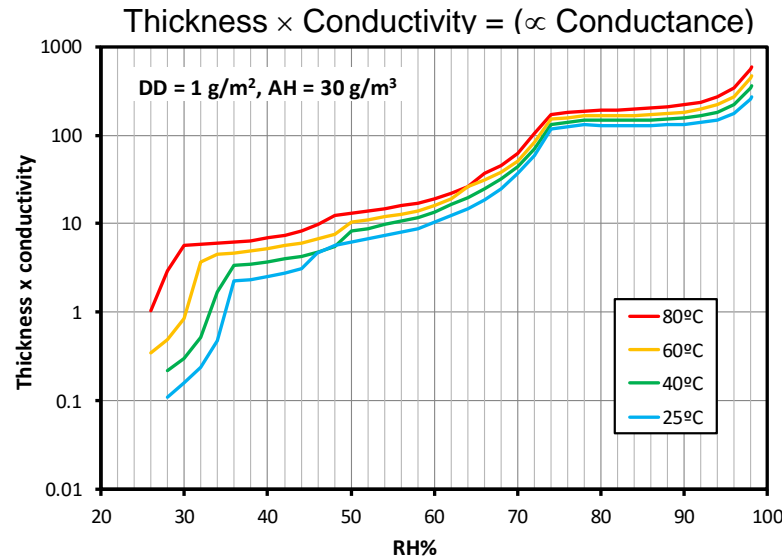
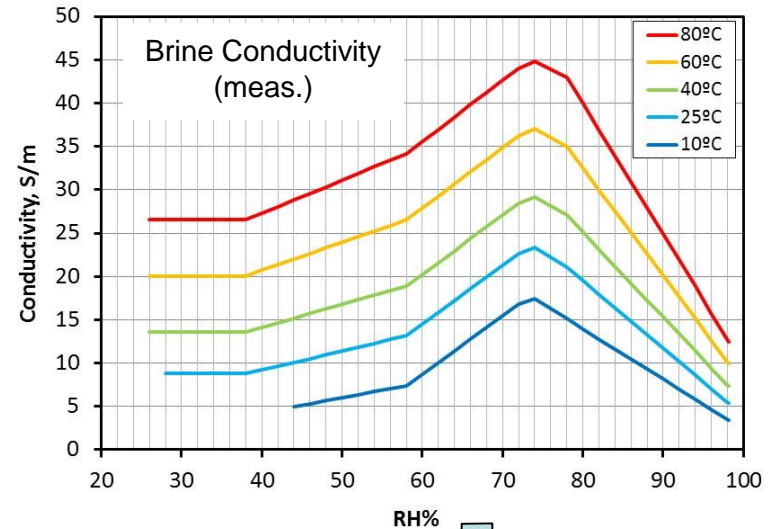
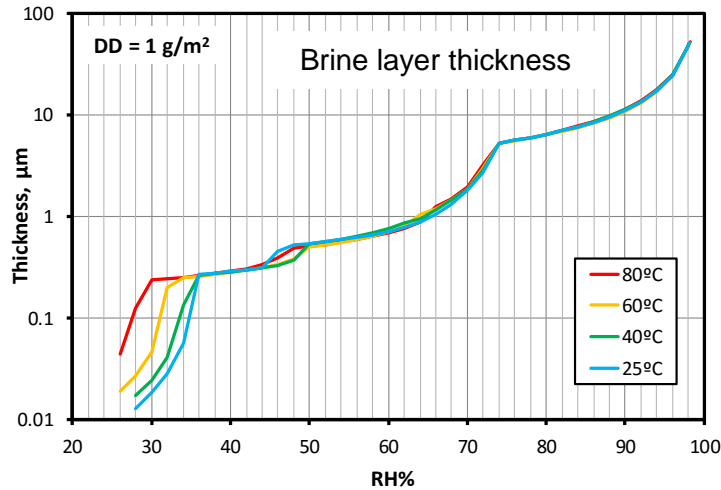
$$\ln I_{c,max} = \frac{4\pi k W_L \Delta E_{max}}{I_{c,max}} + \ln \left[ \frac{\pi e r_a^2 \int_{E_{corr}}^{E_{rp}} (I_c - I_p) dE}{\Delta E_{max}} \right]$$

Labels for the equation:

- Max. cathode current:  $\ln I_{c,max}$
- Brine conductivity:  $k$
- Brine layer thickness:  $W_L$



# Parameterizing the Model for Sea-Salt Brines



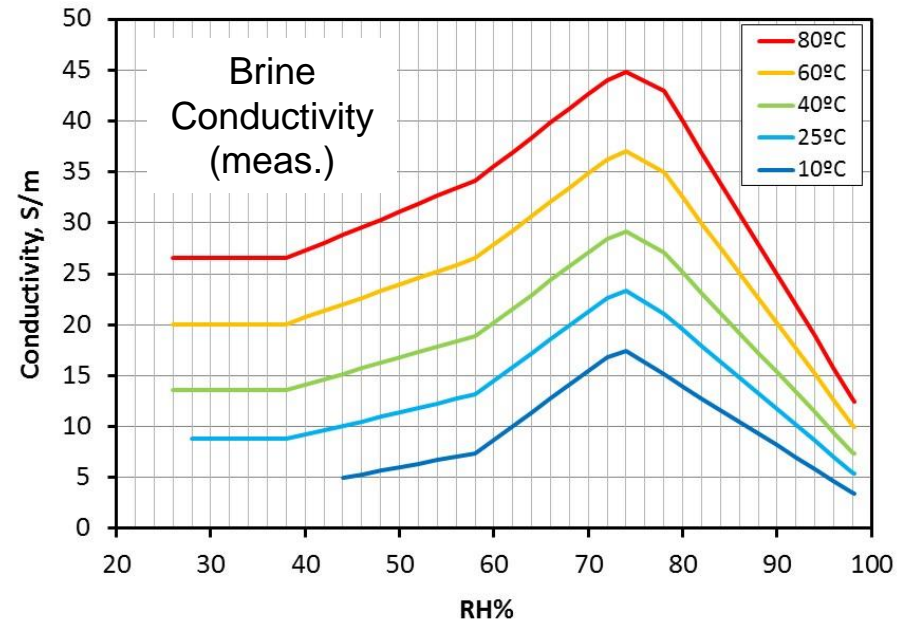
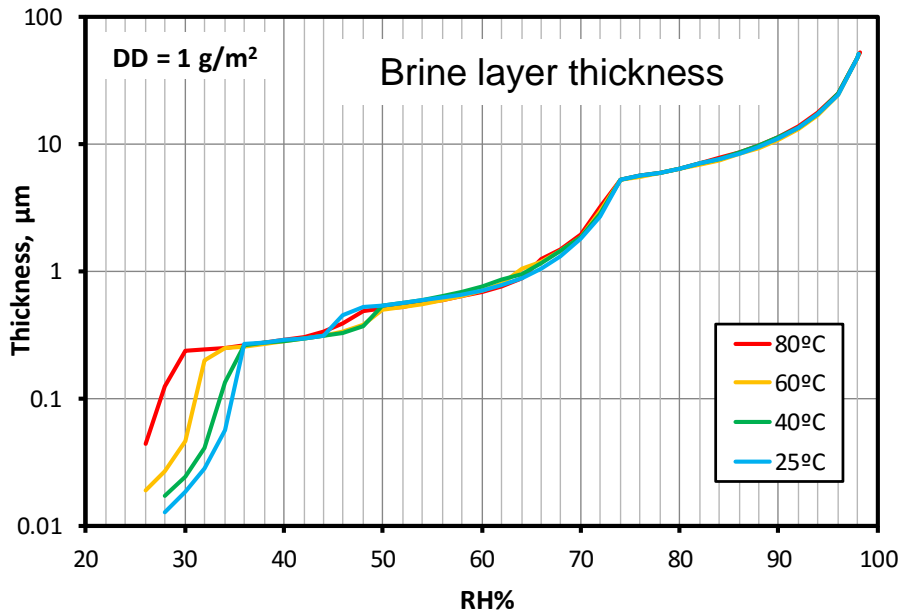
Original *Chen and Kelly* (2010) model: conductivities and brine volumes based on NaCl brines, valid for 75-100% RH.

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



# 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).

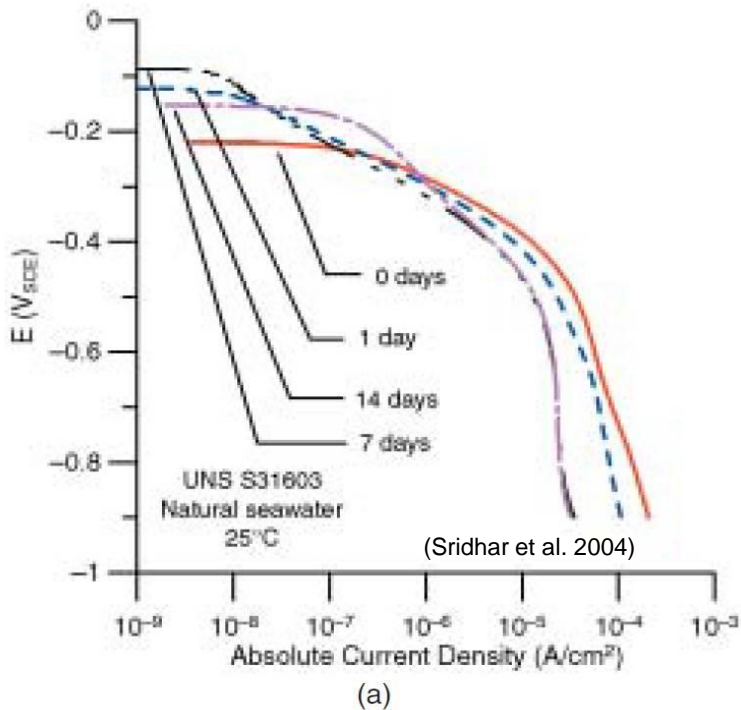


Original *Chen and Kelly* (2010) model:  
Based on NaCl brines  
(dry out at ~75% RH).



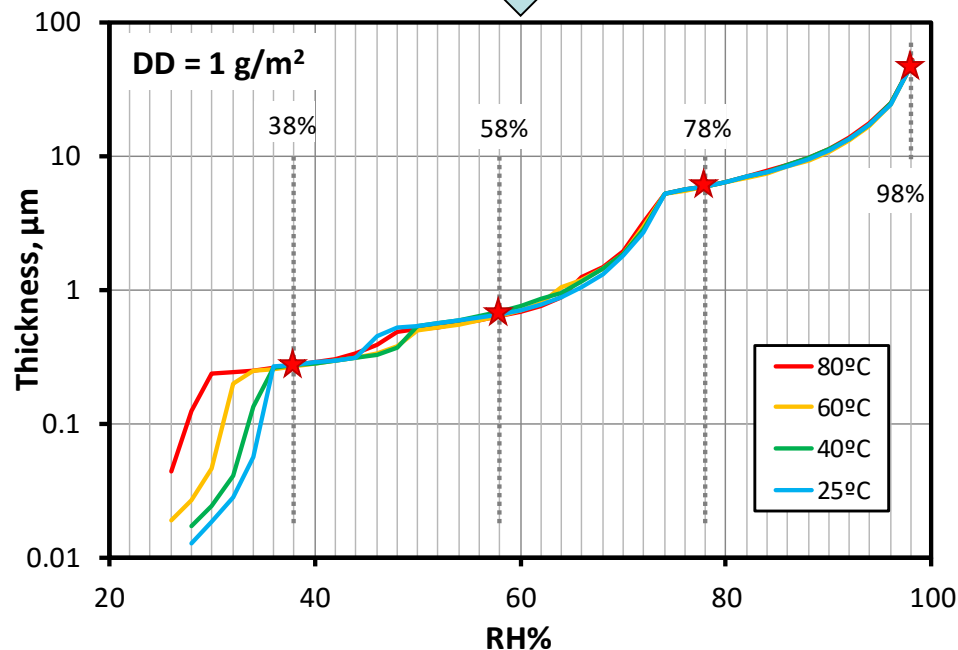
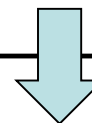
# Other Needed Data: Cathodic Polarization Curves and electrochemical parameters for concentrated brines

Currently, limited available data:  
Cathodic polarization curve for seawater  
at 25°C, 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:

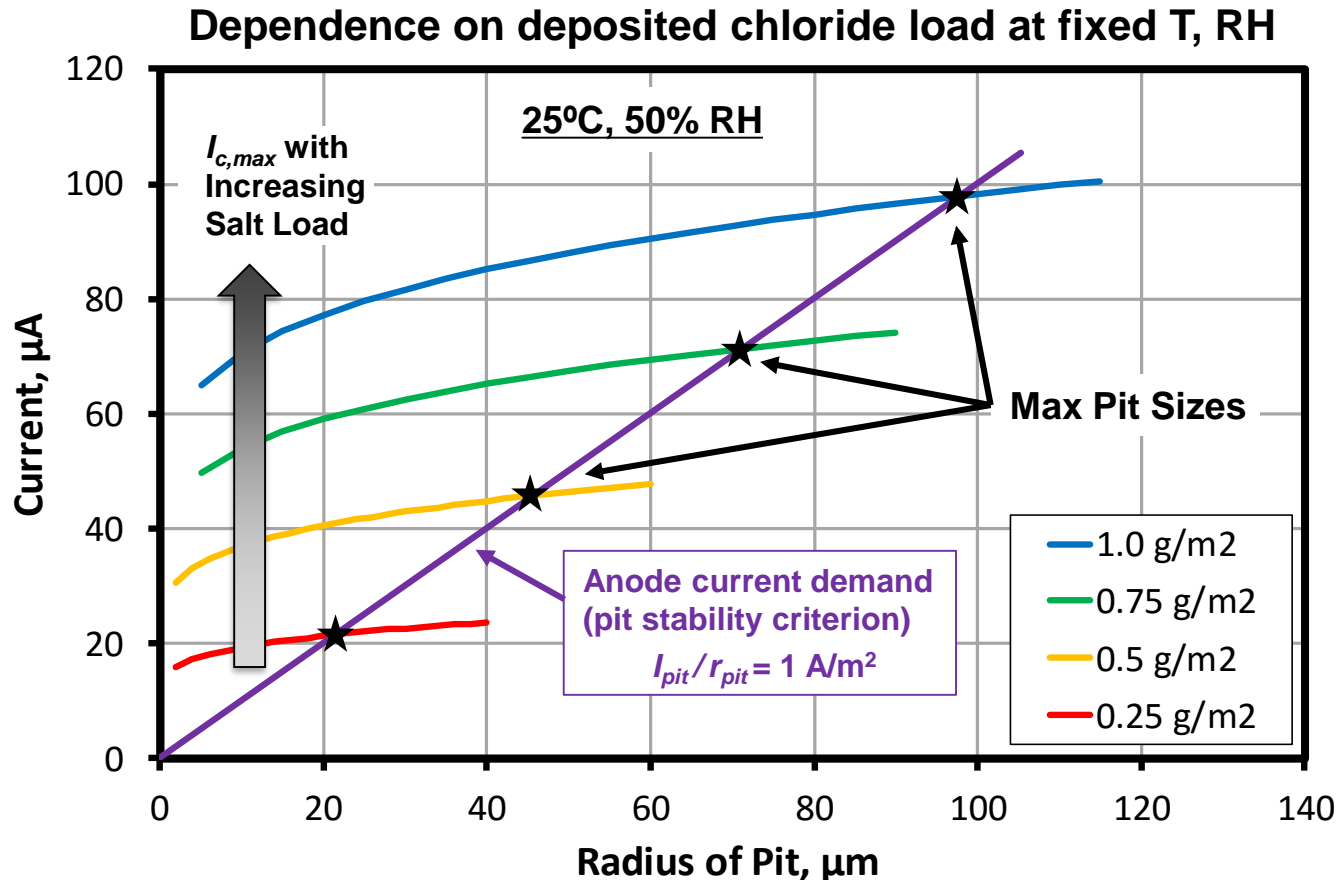
- Unevaporated Seawater (98% RH)
- Evap. to 78% RH
- Evap. to 58% RH
- Evap. to 38% RH





# Determining Maximum Pit Size

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





# 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.

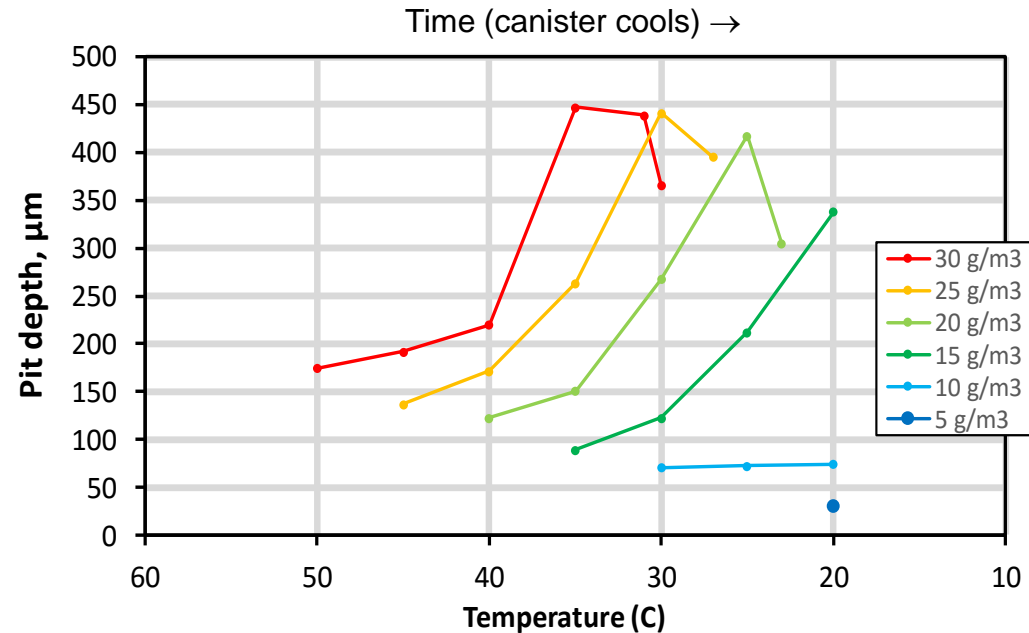
■ **Largest pits possible at temperatures corresponding 75-85% RH**

- Lower T = higher RH, more dilute solutions.
- Higher T = lower RH, thinner brine films
- Maximum pit depths at any AH decrease with increasing T.
- Maximum pit depths at any T increase with increasing AH.

■ **Overall trend:**

- 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.**

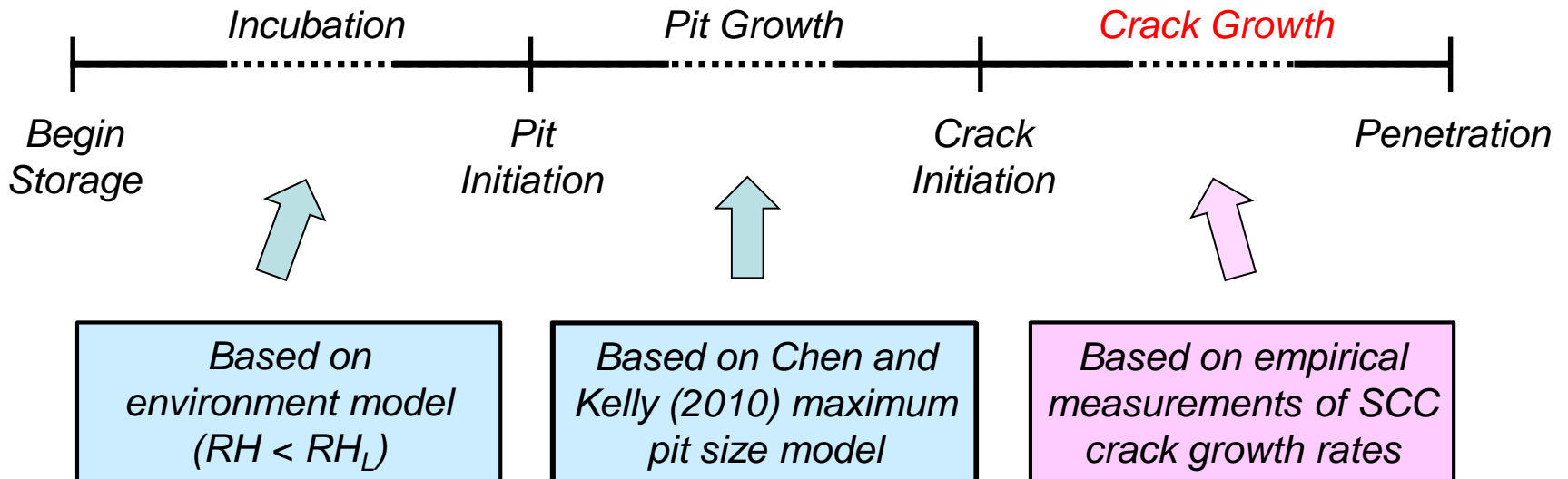


Caveat: Electrochemical terms are held constant. Realistically, they vary with brine composition and temperature.

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



## 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

$\alpha_{crack}$  = 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.

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

$\sigma_{applied}$  = tensile stress

$Y$  = shape factor

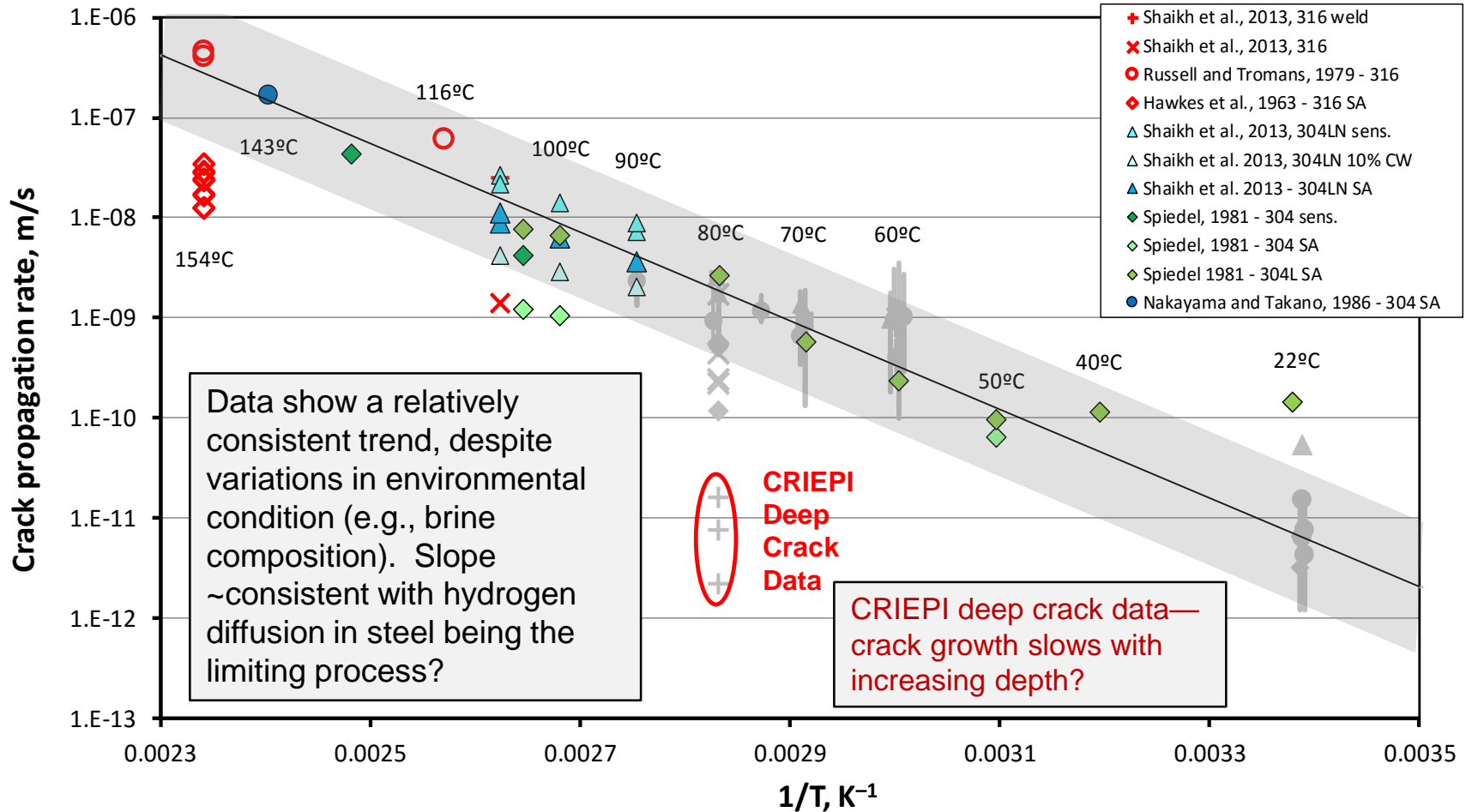
$x_{crack}$  = depth



# Literature Crack Growth Rates

## Data for 304 SS and 316 SS

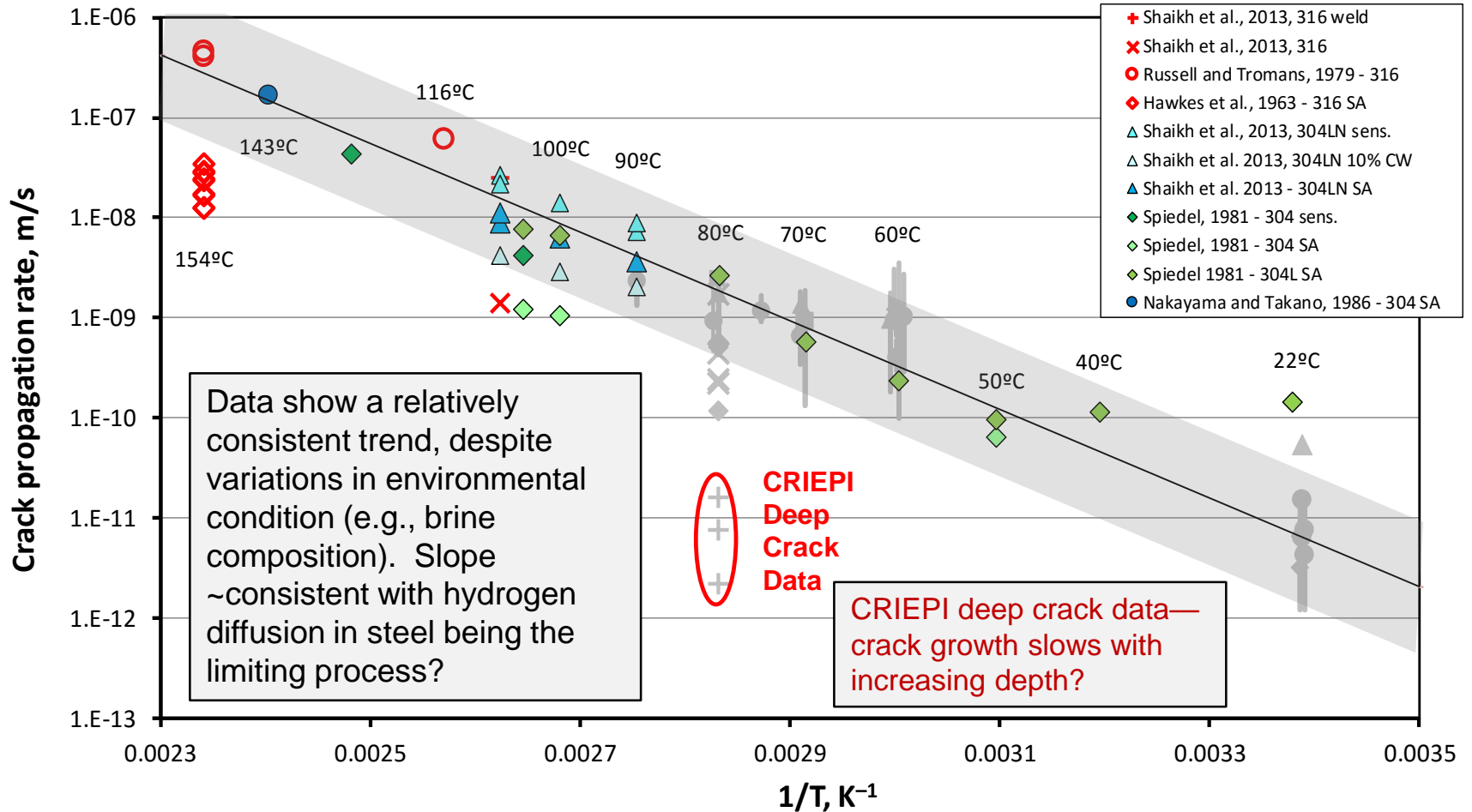
### Add Data from Immersed Experiments





# Literature Crack Growth Rates

## Data for 304 SS and 316 SS

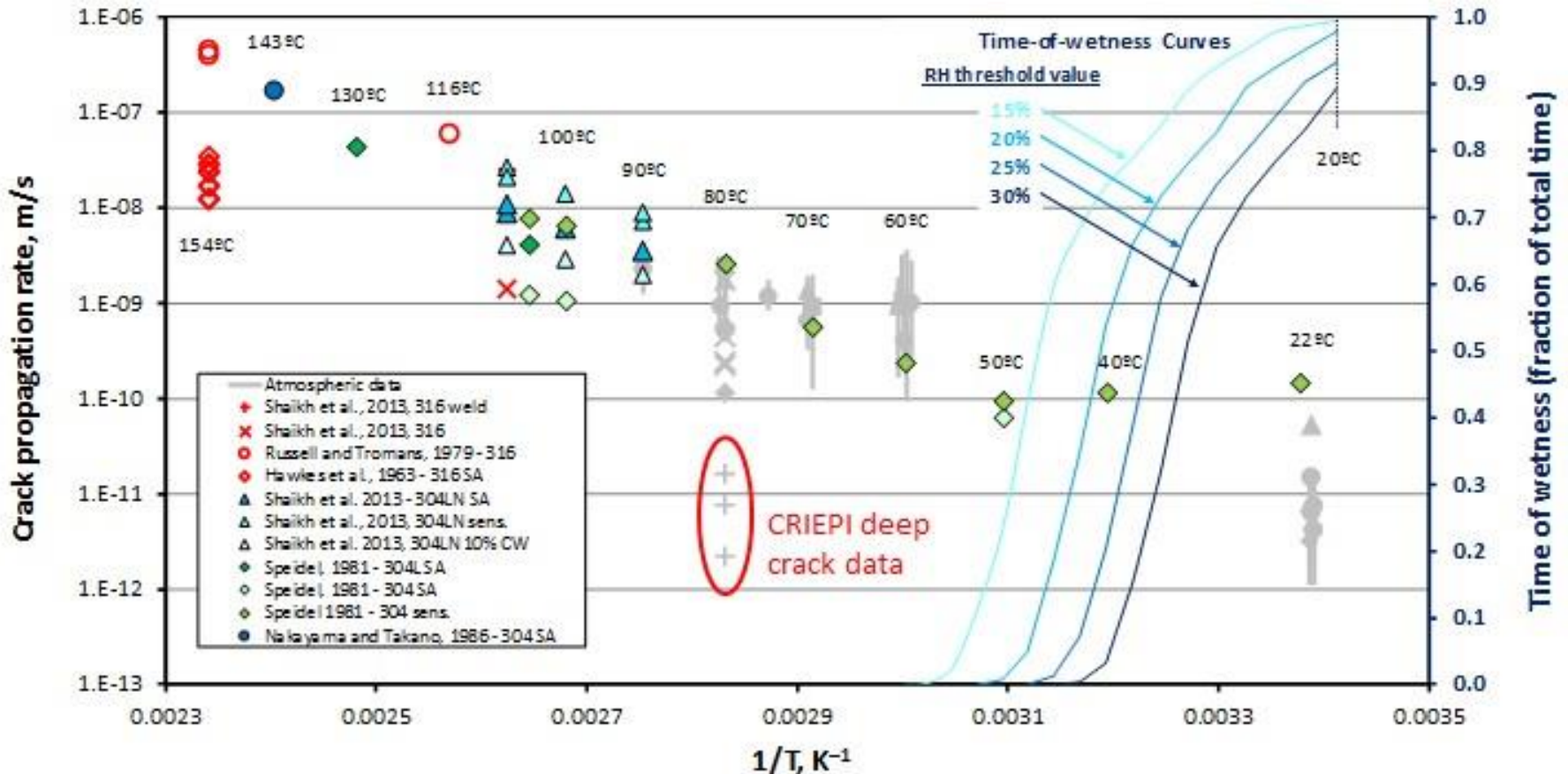




# Role of Time-of-Wetness

## Nuclear Energy

Time-of-wetness plays an important role in decreasing overall crack penetration rates



## Intended Model Uses

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- **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



# 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?