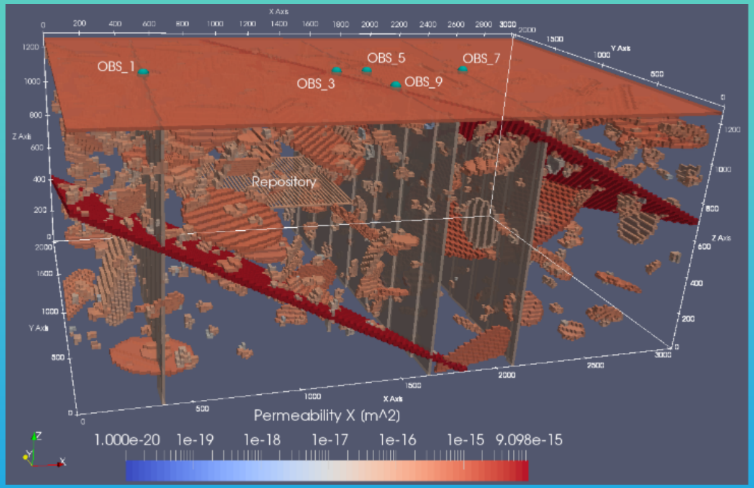
U.S. DEPARTMENT OF ENERGY | Office of NUCLEAR ENERGY

Spent Fuel and Waste Science and Technology (SFWST)

Clean. Reliable. Nuclear.



Understanding Stress Corrosion Cracking of SNF Dry Storage Canisters

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Sandia National Laboratories

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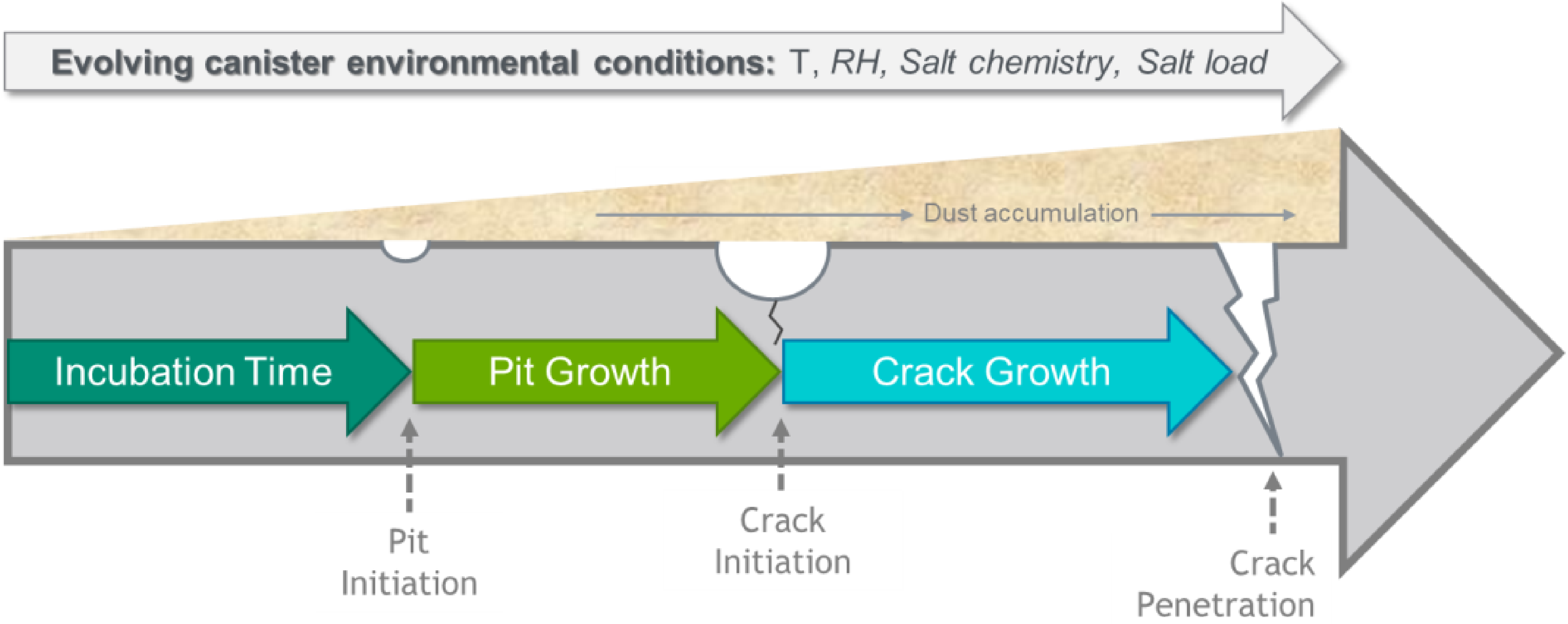
SAND2024-02166C

Timeline, SCC of SNF Dry Storage Canisters



Current Research:

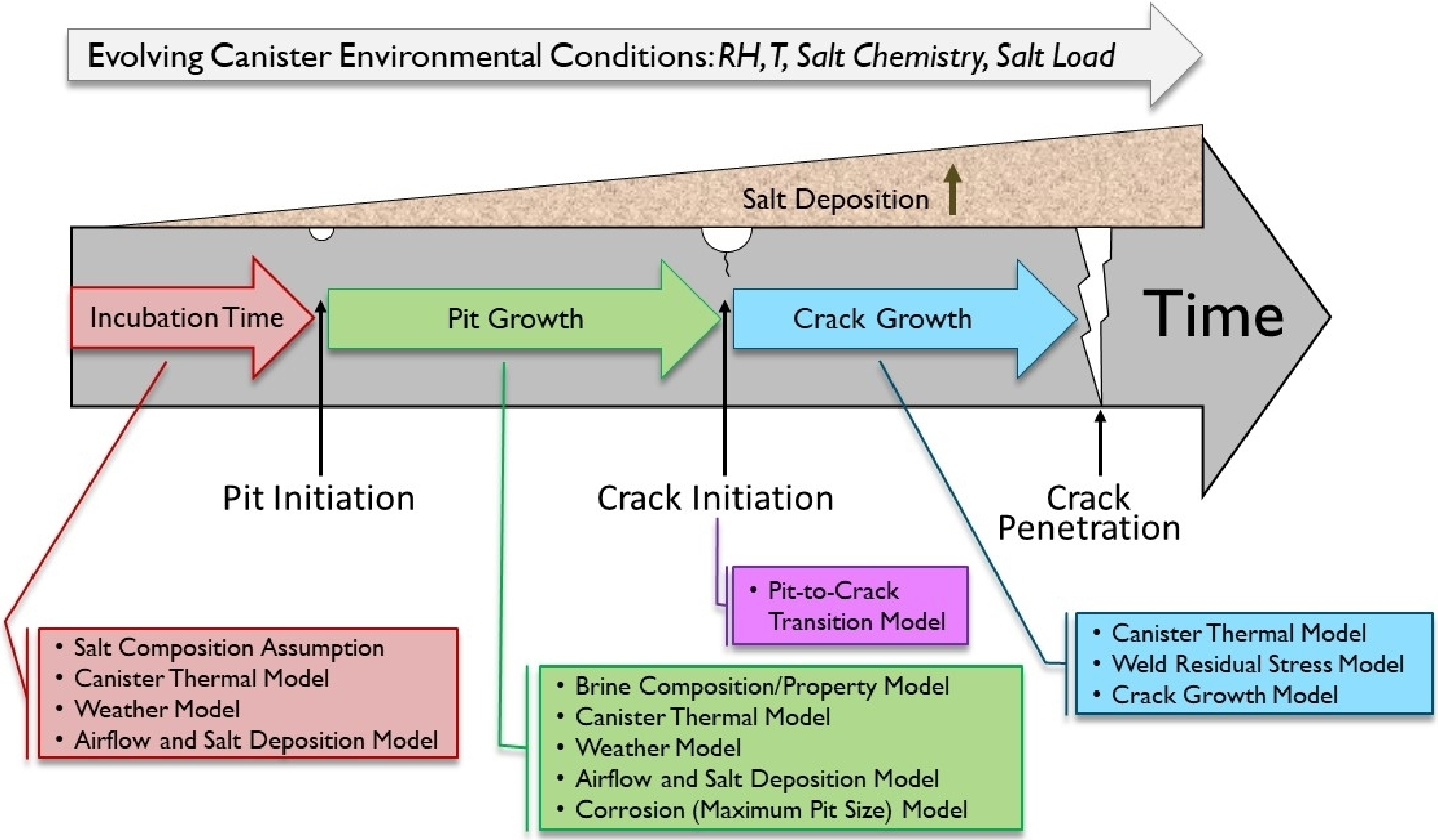
- **Defining the canister surface environment**
- **Influence of relevant canister environment on corrosion (pitting/SCC)**
- **Features and processes driving pit-to-crack transition**
- **Crack growth rate studies**

Probabilistic Model for Canister SCC

Provides the Framework for Experimental Studies

Evaluating timing of canister SCC initiation and penetration

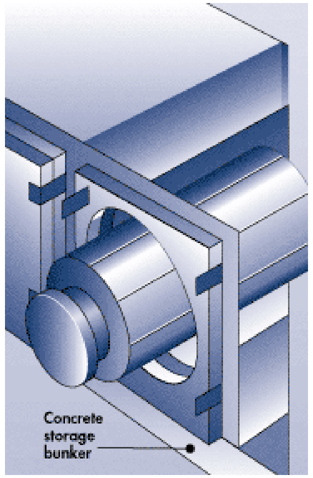
- Submodels for different features, events, and processes
- Used to evaluate model sensitivities, to focus research on reducing uncertainties



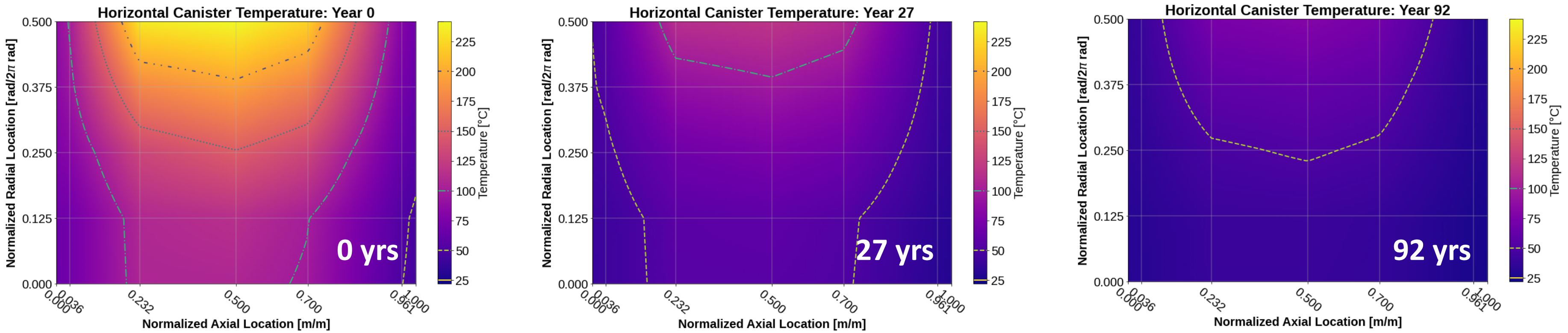
Canister Thermal Models

Based on PNNL thermal modeling

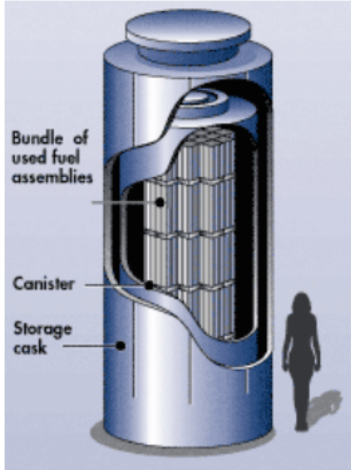
Horizontal Model



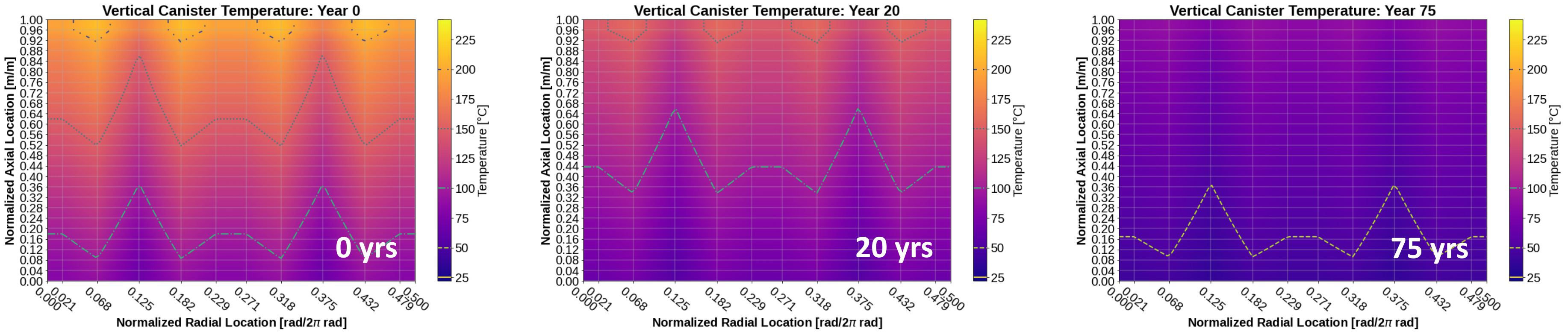
Based on modeling of Calvert Cliffs NUHOMS 24P — 8 decay heat loads, 24–2 kW, corresponding to 0-292 yrs out of the reactor



Vertical Model



Vertical canister (based on modeling of Diablo Canyon Holtec HI-STORM 100) — 8 decay heat loads, 30.2–5.6 kW, corresponding to 0-125 yrs out of the reactor



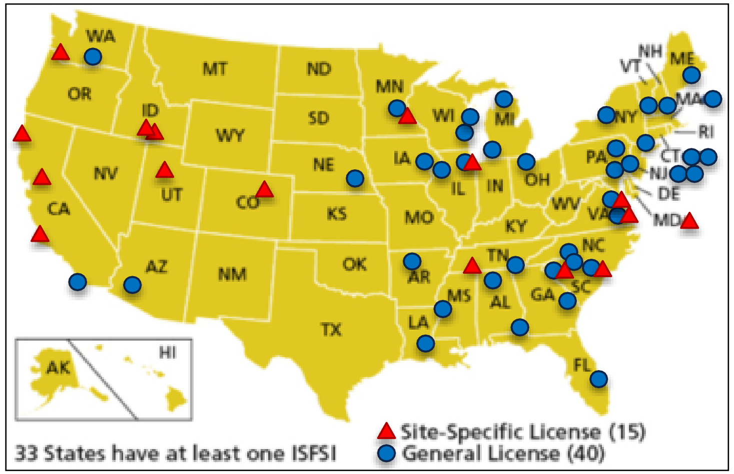
Suffield et al.(2012), Thermal Modeling of NUHOMS HSM15 Storage Module at Calvert Cliffs Nuclear Power Station ISFSI,PNNL-21788, 102 p.
Cuta and Adkins (2014), Preliminary Thermal Modeling of HI-STORM 100 Storage Modules at Diablo Canyon Power Plant ISFSI, PNNL-23298, 56 p.

Weather Model (Ambient T and AH)

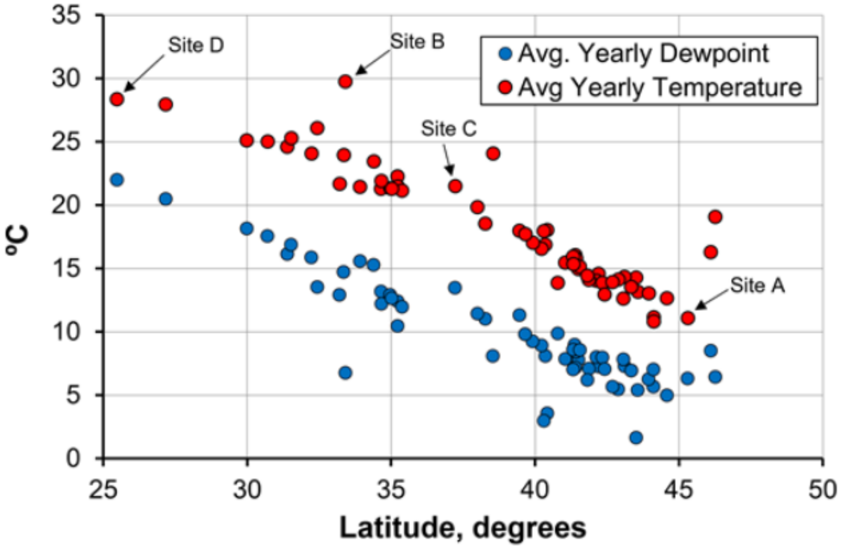
Weather Model—provides ambient T, absolute humidity (AH)

- Based on one year of hourly data for 2012 (National Weather Service)
- Nearest airports to each ISFSI (64 individual data sets)
- Fitted probabilistic model captures daily and seasonal variations in T and AH, with correlation

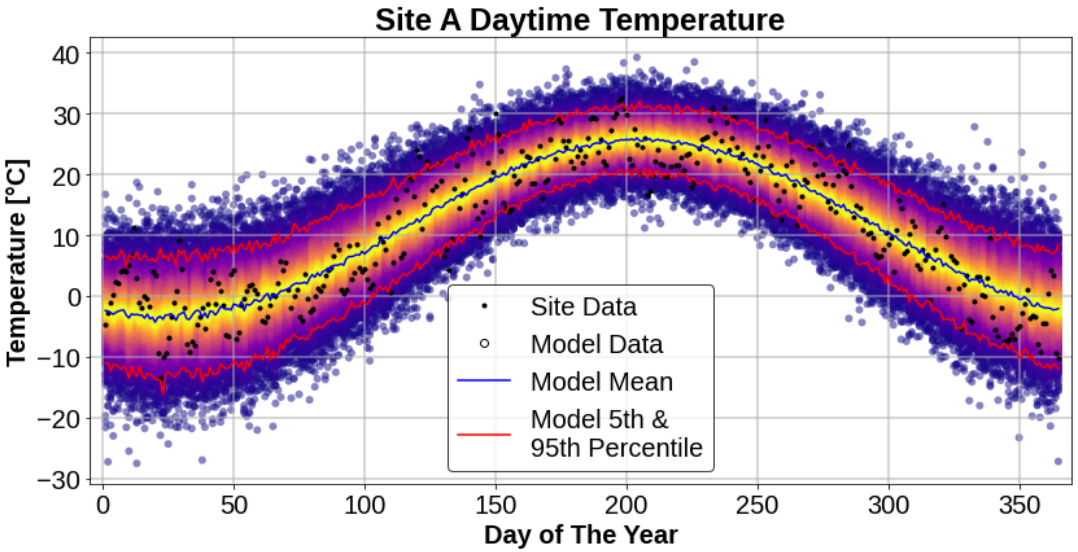
Independent Spent Fuel Storage Installations (ISFSIs) in the U.S.



Weather conditions vary widely from site to site



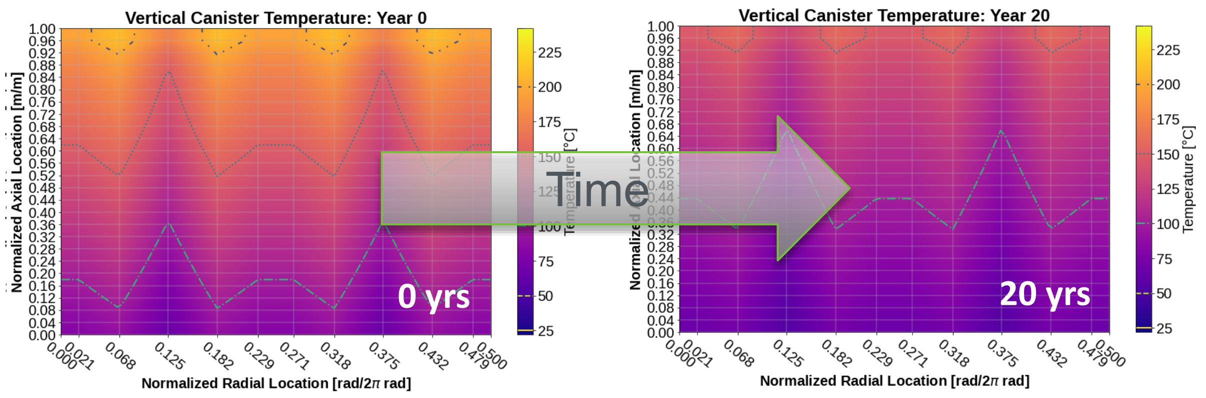
Predicted weather data (daily max T)



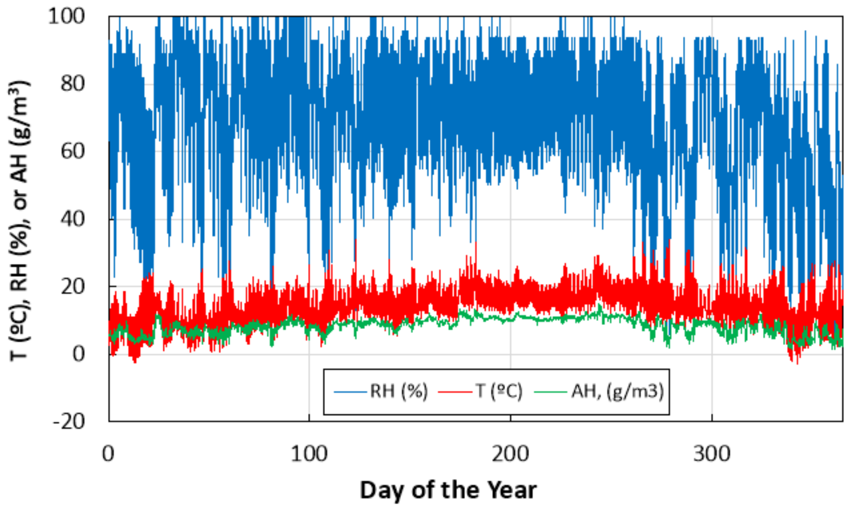
Predicting Canister Surface Temperature and RH

Canister thermal models + ISFSI site-specific weather data → Canister surface T and RH at any location on the canister surface

Canister thermal data



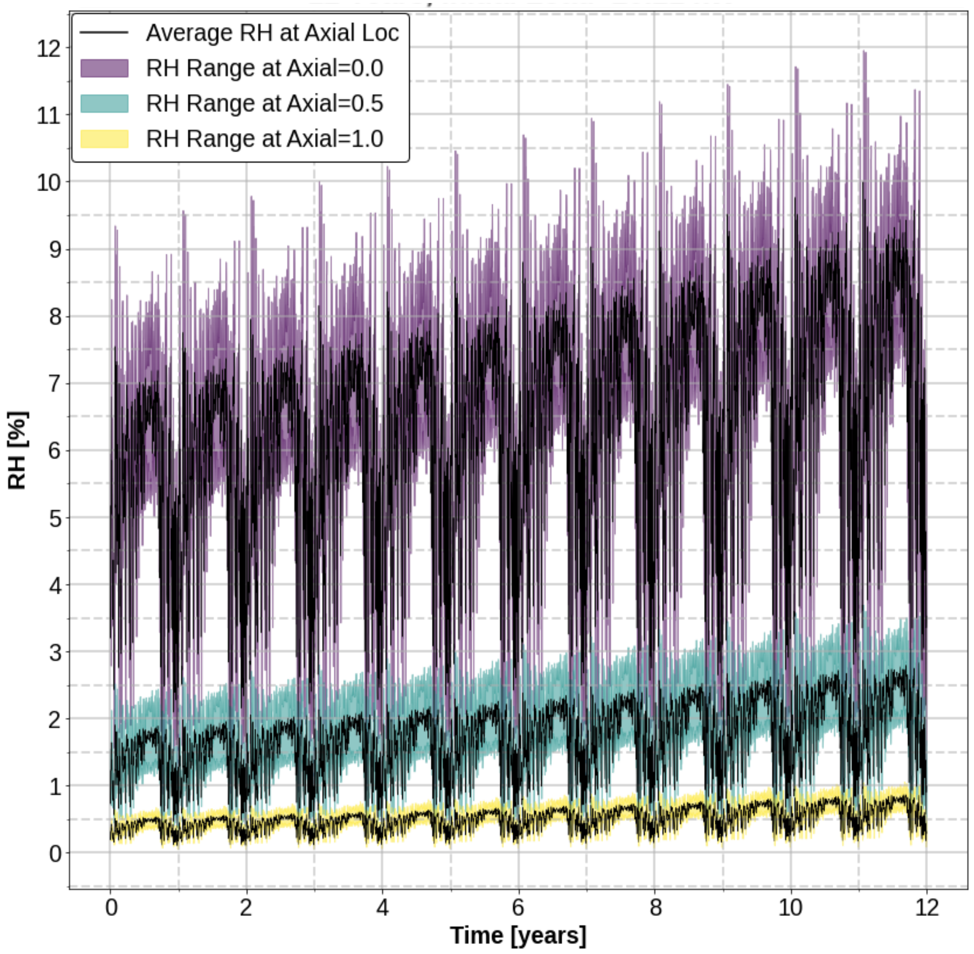
Weather data



Base-case thermal models for ambient temperature, 15.5°C (60°F)

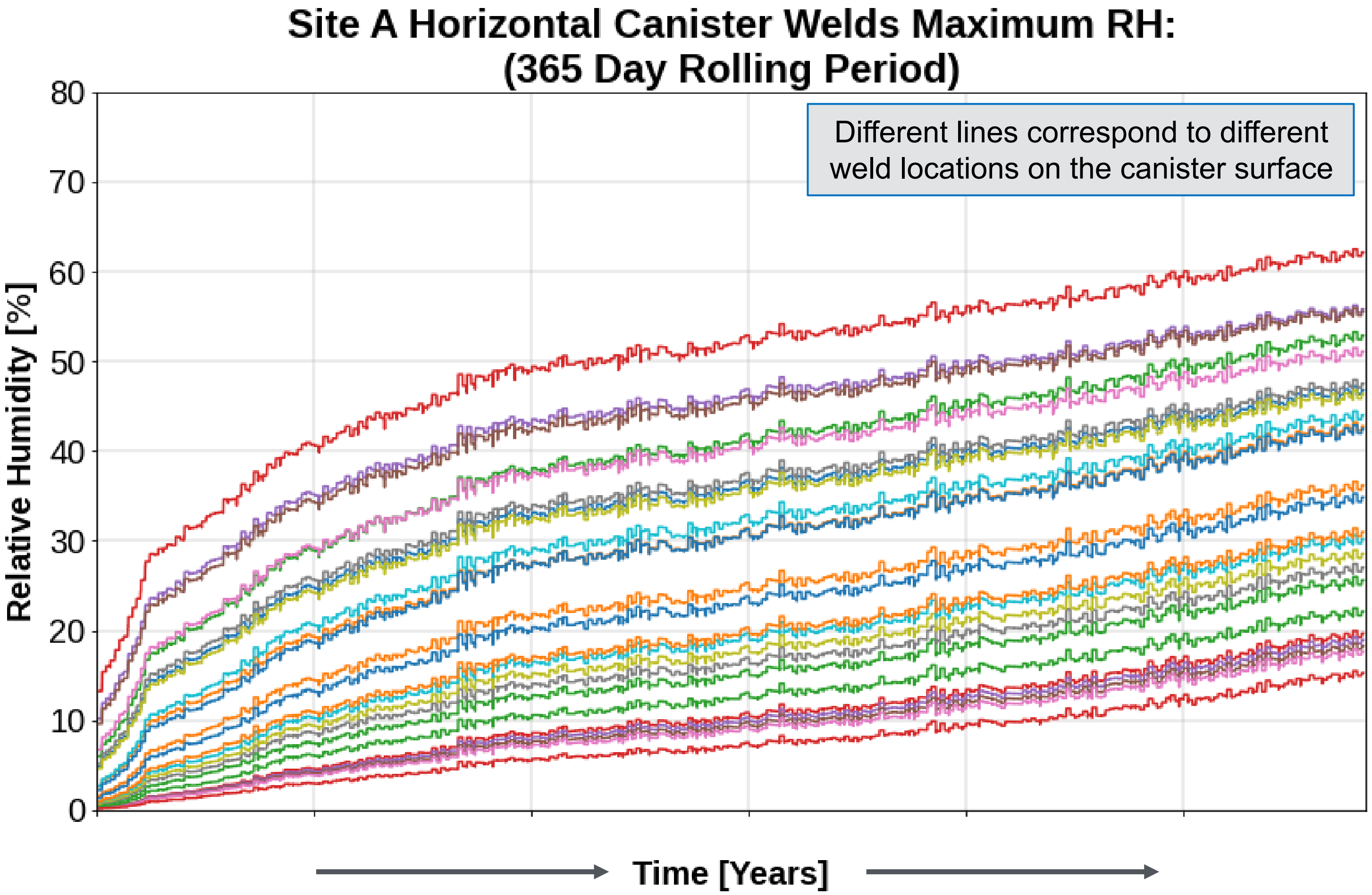
Implement different ambient temperatures as a delta to the base-case model

Vertical Canister Predictions



Predicting Canister Surface Temperature and RH

Canister thermal models + ISFSI site-specific weather data →
Canister surface T and RH at any location on the canister surface



When can brine formation and corrosive conditions occur?

Depends on the composition of salts present

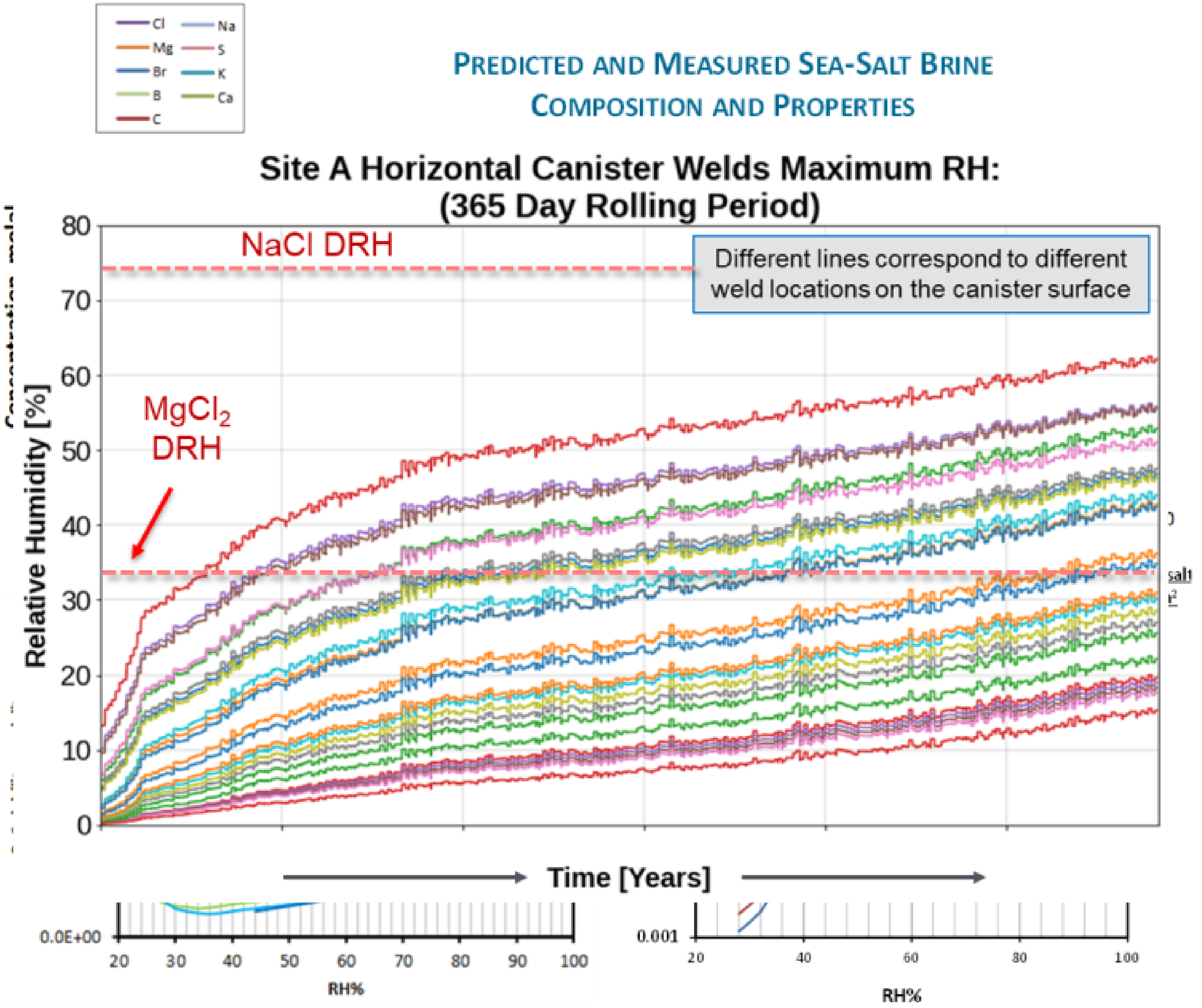
Salt Composition Assumption

Salt compositions: For near-marine sites, assume chloride is deposited as sea-salt aerosols

- Brine composition/properties: calculated/measured brine properties as *f*(RH,T) used for modeling
- Compositions affect timing of deliquescence and potential corrosion initiation

Importance of $MgCl_2$ brines:

For SNF dry storage canisters, $MgCl_2$ brines will form first and will persist for hundreds of years



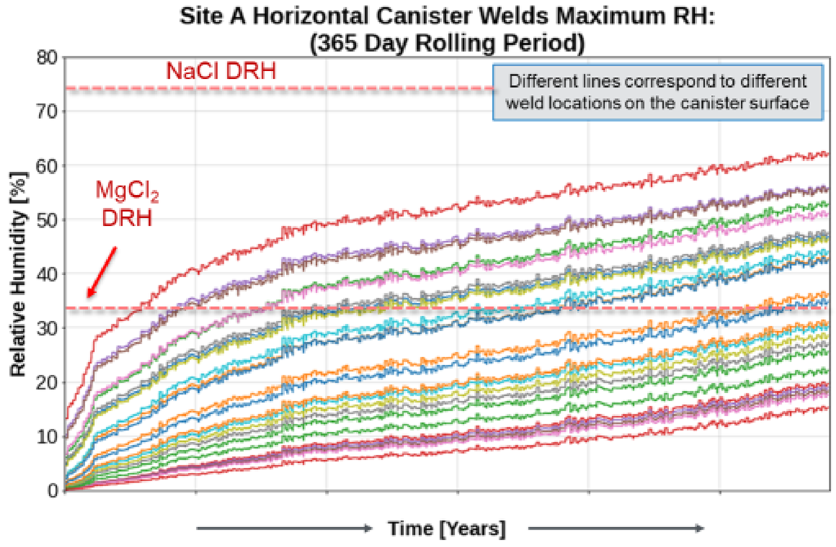
Canister Surface Environment

Mg-Chloride Brine Stability

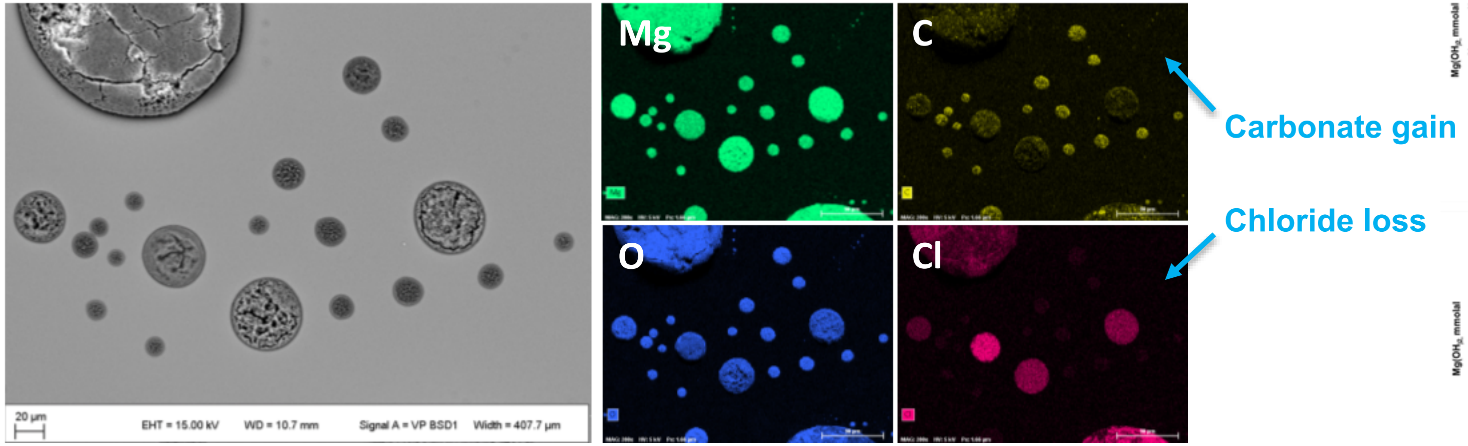
Mg-chloride brines are not stable at elevated temperatures. **Why Important?** Impacts:

- Timing of corrosion initiation on canisters
- Brine volumes and corrosion extent/evolution
- Corrosion morphology, pit-to-crack transition
- Interpretation of experimental results
- Extrapolation to field conditions

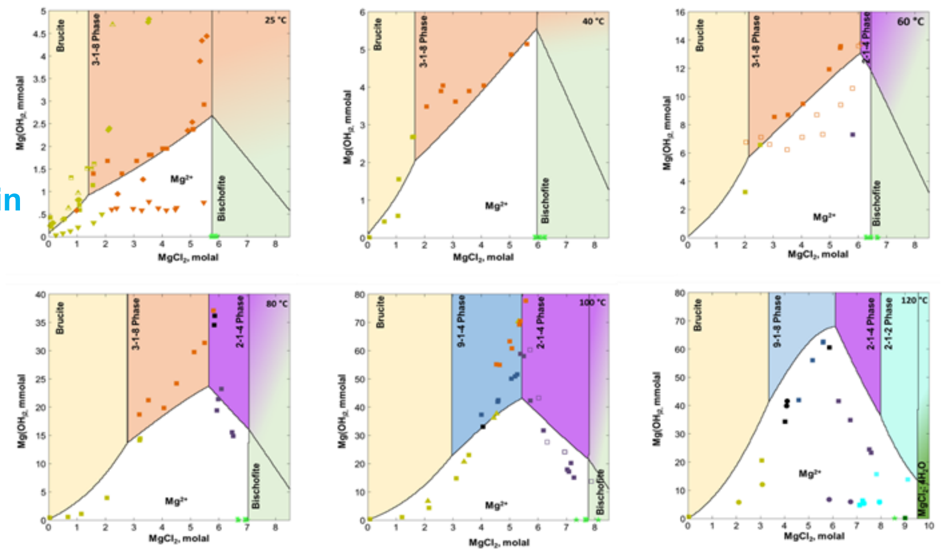
Evaluated via experiments and modeling



MgCl$_2$ brine degassing experiments



Thermodynamic model for Mg-Cl-(OH)-H$_2$O system



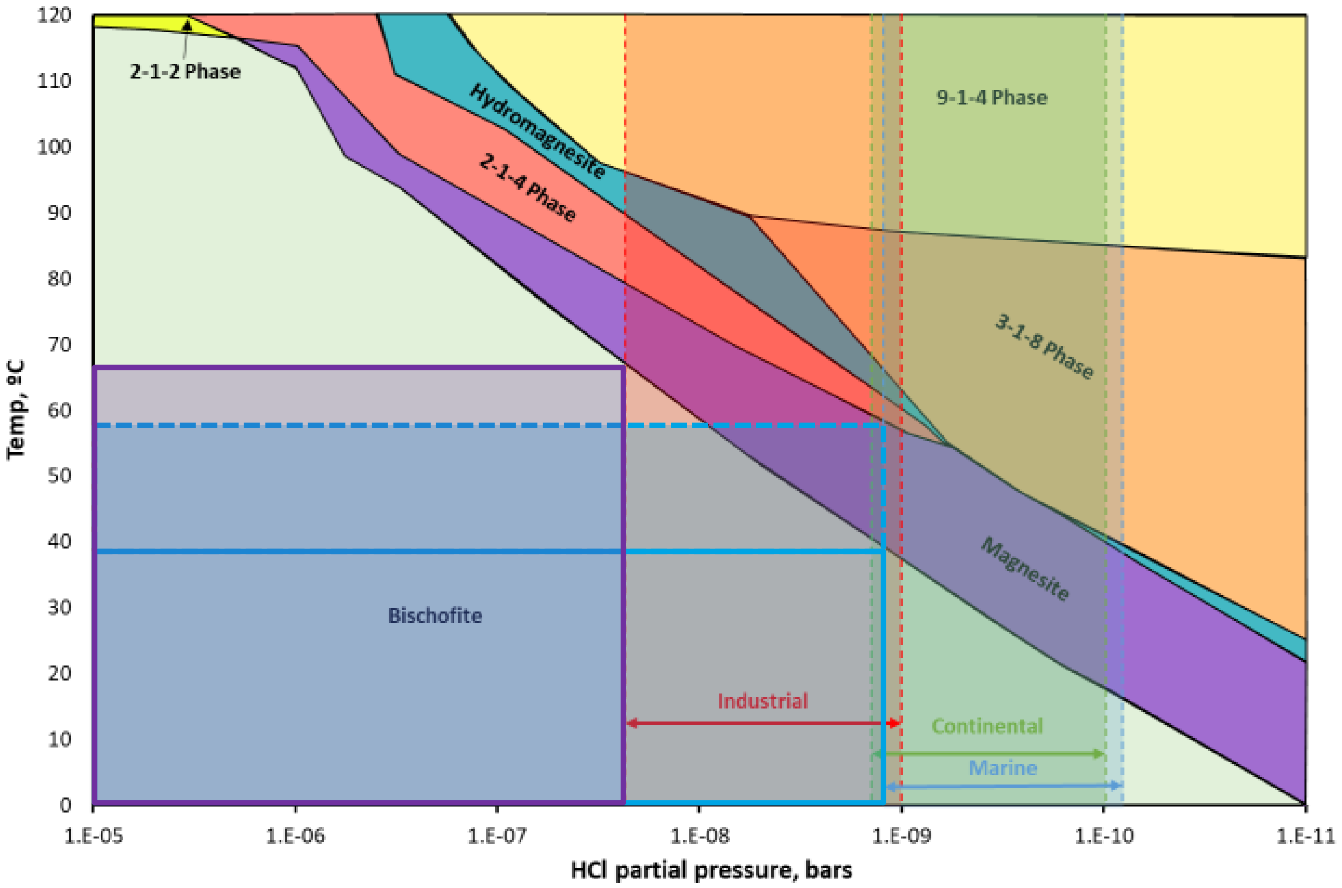
Canister Surface Environment

Mg-Chloride Brine Stability

Stable conditions for $MgCl_2$ brines:

- In near-marine settings, ~38ºC to ~57ºC, depending upon buffering phase (carbonate or hydroxychloride).
- Industrial settings—potentially higher temperatures

$MgCl_2$ brine degassing/conversion to non-deliquescent phases explains many experimental results



Corrosion Modeling

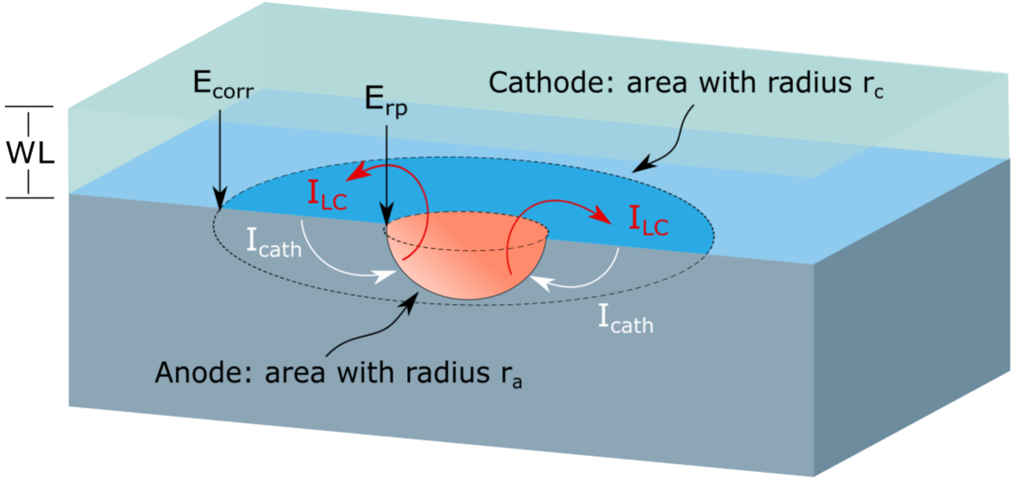
Maximum Pit Size Model

Maximum pit size model

- Pit (anode) must be supported by cathodic reduction reaction forming an inherent galvanic couple
- In finite-thickness water layers, cathode limited by ohmic drop

Finite cathode → Finite anode → Finite pit

Pitting under thin electrolyte layer



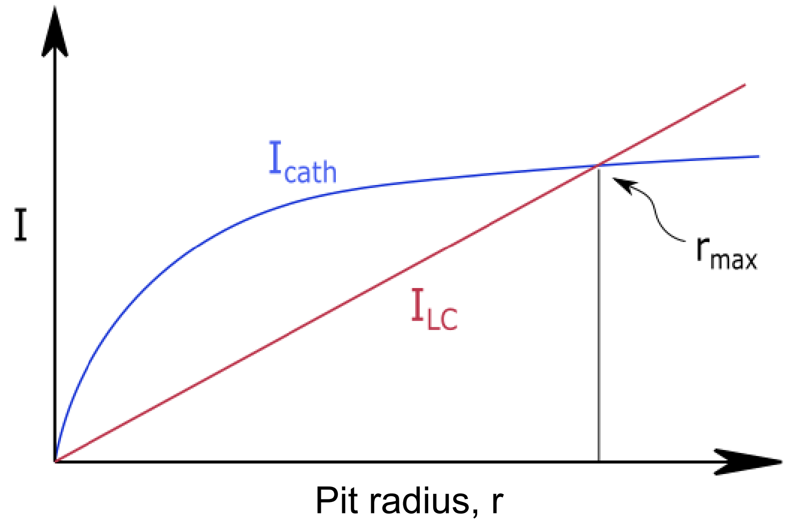
Max. cathode current

Brine conductivity

Brine layer thickness

Electrochemical term (from cathodic polarization curve)

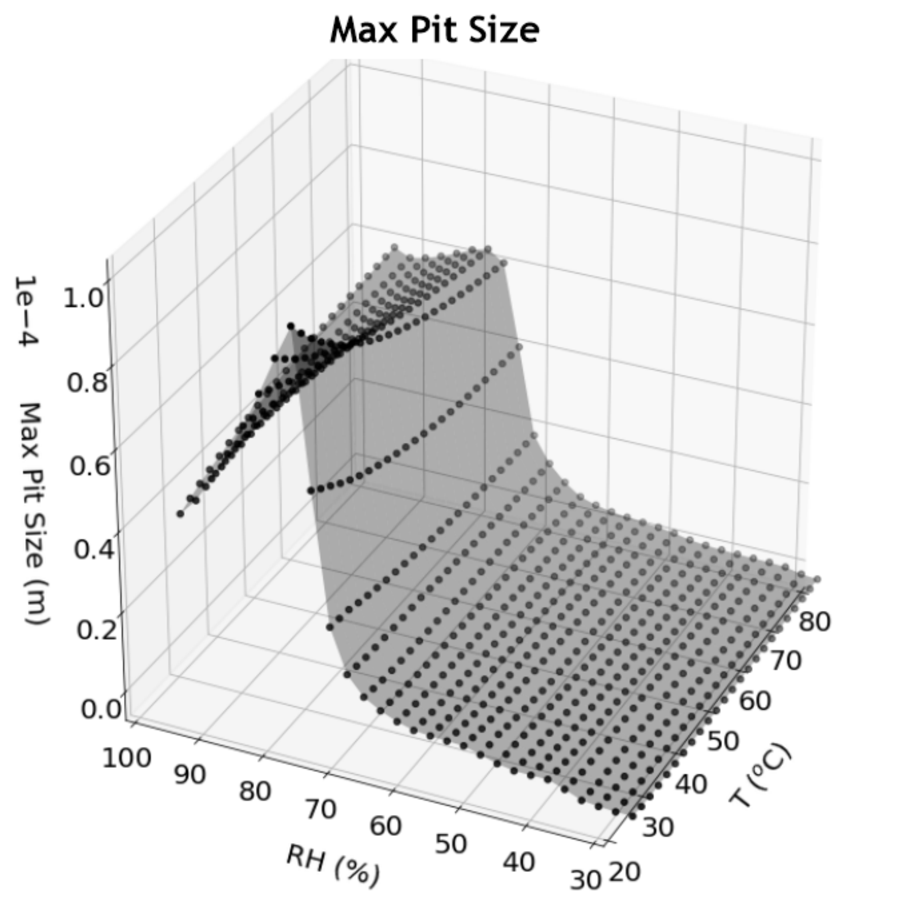
$$\ln I_{c,max} = \frac{4\pi k W_L \Delta E_{max}}{I_{c,max}} + ln\left[\frac{\pi e r_a^2 \int_{E_{corr}}^{E_{rp}} (I_c - I_p)\, dE}{\Delta E_{max}}\right]$$

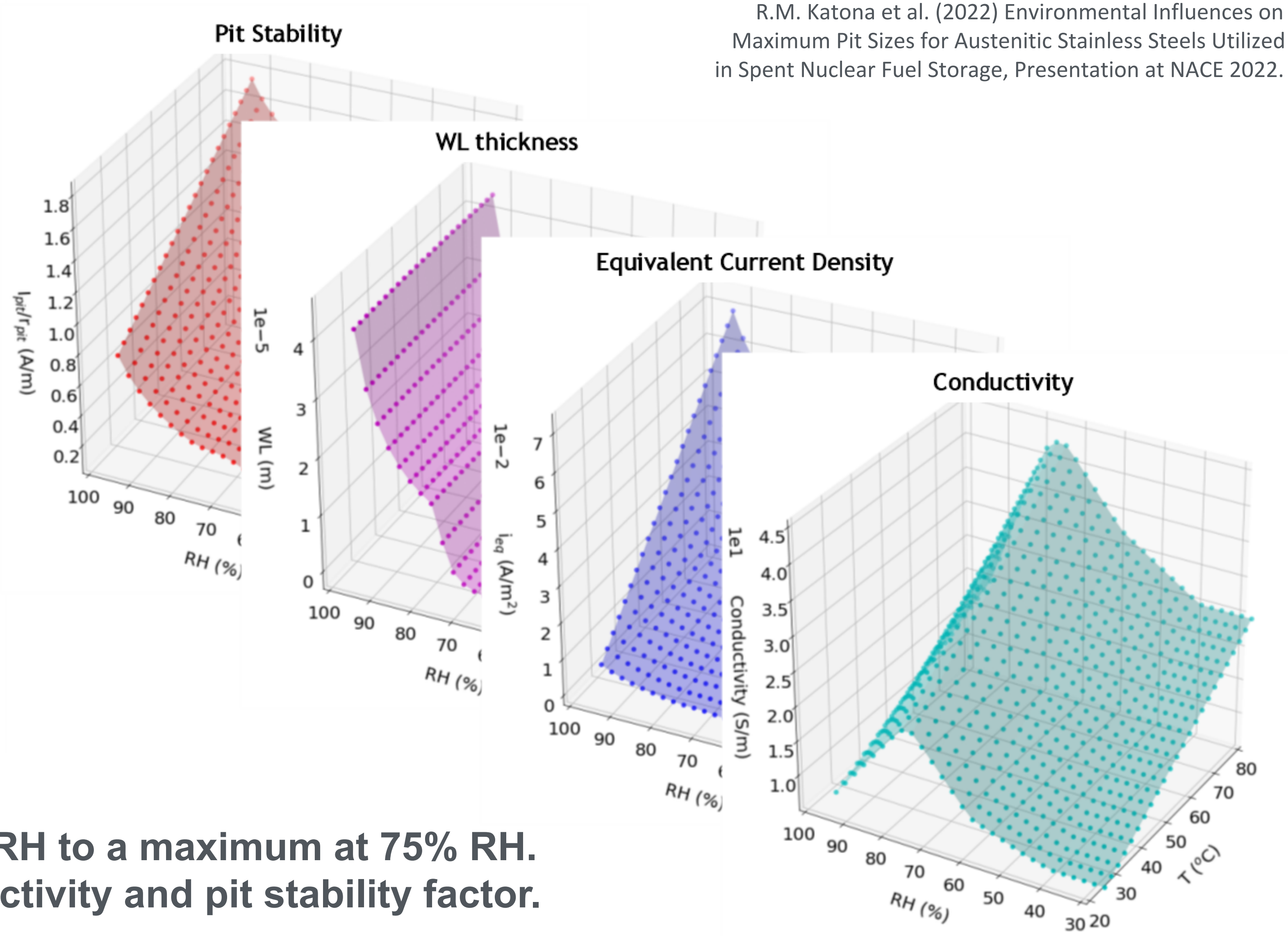


Chen, Z. Y., & Kelly, R. G. (2009). Computational modeling of bounding conditions for pit size on stainless steel in atmospheric environments. *Journal of the Electrochemical Society*, *157*(2), C69.

Prediction of Maximum Pit Sizes
Parameterization of the model

R.M. Katona et al. (2022) Environmental Influences on Maximum Pit Sizes for Austenitic Stainless Steels Utilized in Spent Nuclear Fuel Storage, Presentation at NACE 2022.



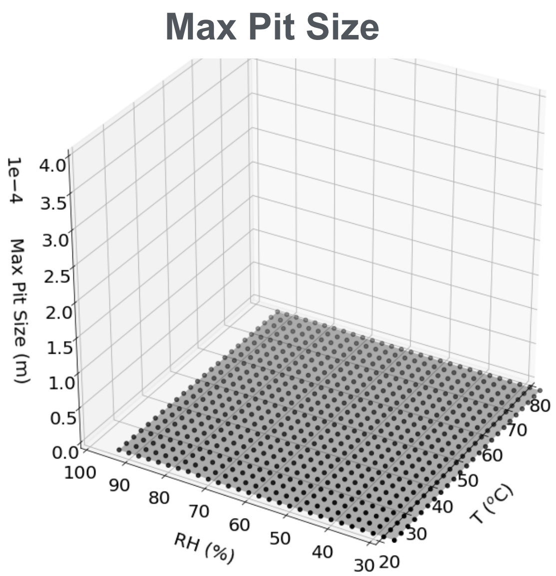


Max pit size increases with increasing RH to a maximum at 75% RH. Peak at 75% RH due to trends in conductivity and pit stability factor.

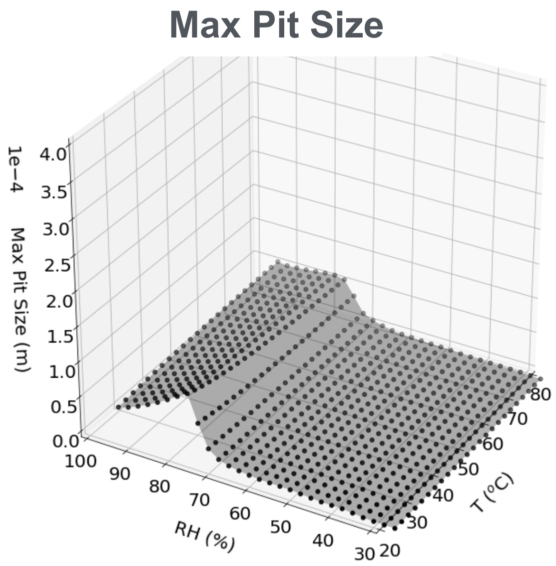
Prediction of Maximum Pit Sizes

Parameterization of the model

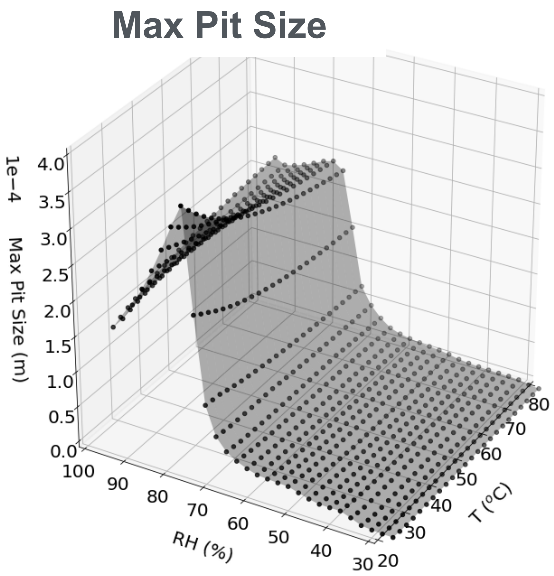
Increasing Salt Load (LD)



LD = 0.1 g/m²



LD = 3 g/m²



LD = 10 g/m²

Environmental influences on corrosion damage (maximum pit size)

- ***Decreasing RH increases maximum pit sizes to a maximum at ~ 75 % RH***
- ***Increasing temperature slightly decreases maximum pit size***
- ***Increasing salt load increases maximum pit size***

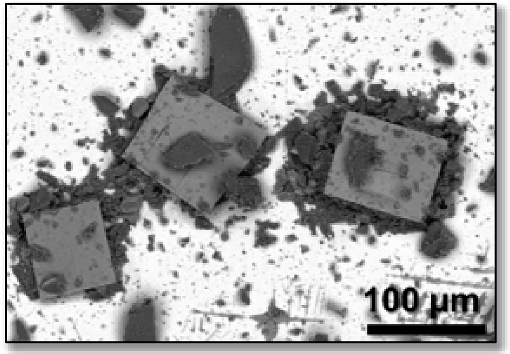
What about other factors?

More realistic canister surface environments

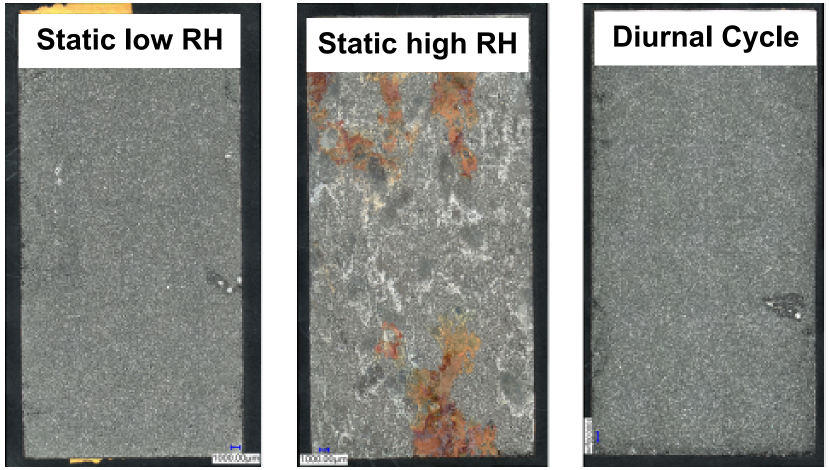
Dust Exposures

- Atmospheric Exposure – 3 conditions

74 µm dust deposited with seawater

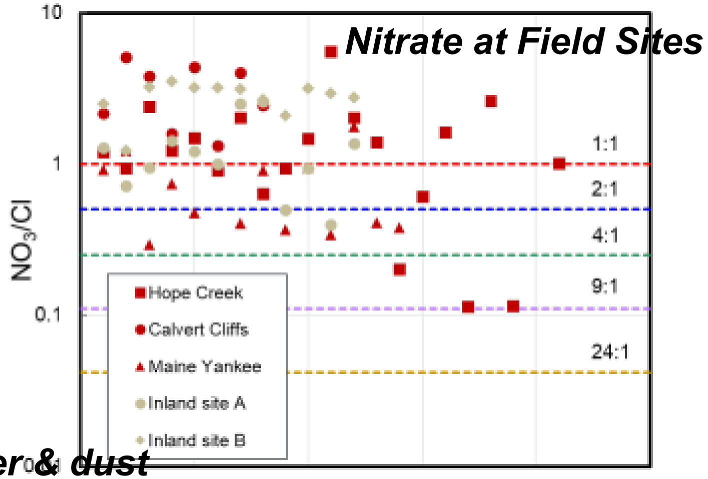


1 month exposure – 304 coupons with seawater & dust

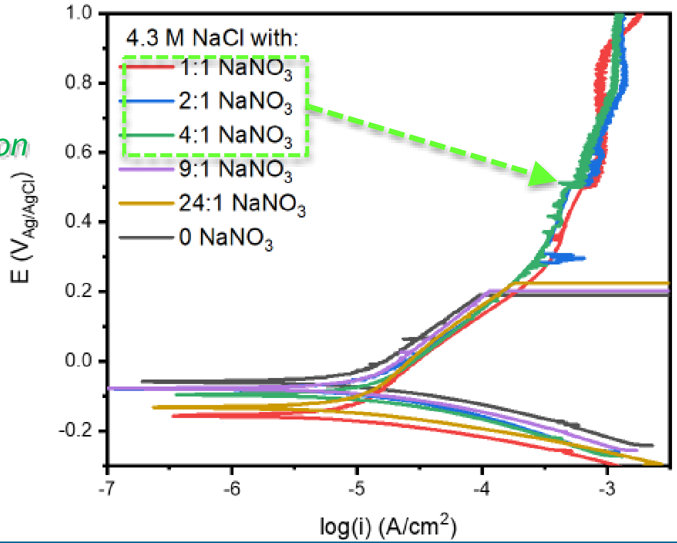


Chemistry

- Immersed scoping measurements

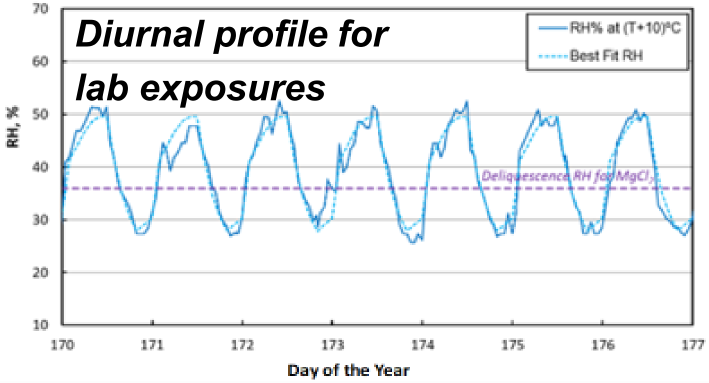


Passivating effects of Nitrates: Concentration Dependent

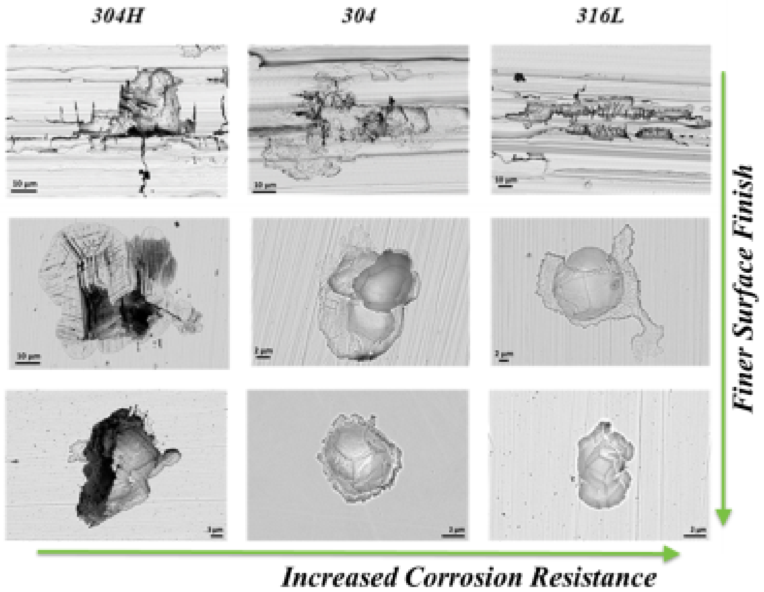


Cyclic Exposures

- Atmospheric Exposure – diurnal cycle



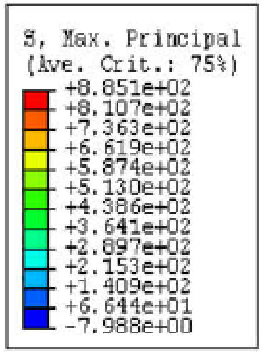
Post Exposure Corrosion Damage



Pit-to-Crack Transition

Effect of Maximum Pit Size

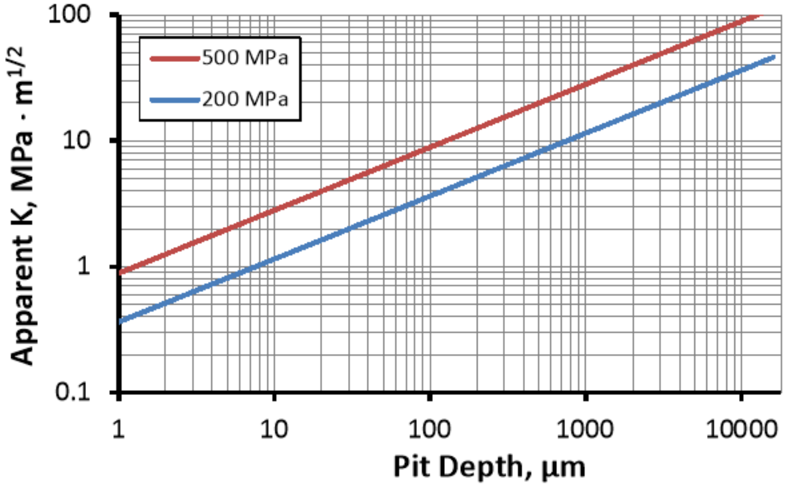
Why is pit size important?
Contributes to pit-to-crack transition



Pits act to focus tensile stresses

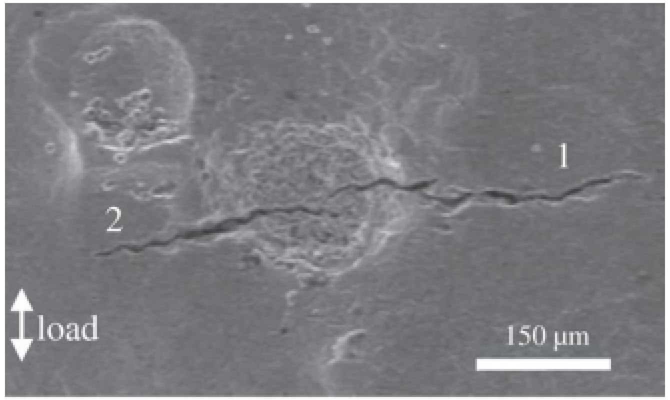
Turnbull, A., Wright, L., & Crocker, L. (2010). *Corrosion Science*, *52*(4), 1492-1498.

As pits grow, tensile stresses increase; ultimately a SCC crack can spawn from the pit (Kondo* criterion)



Apparent crack tip stress intensity factor (K) increases with pit depth; once a threshold value is reached, a SCC crack initiates

Turnbull, A. (2014). *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences*, *470*(2169).



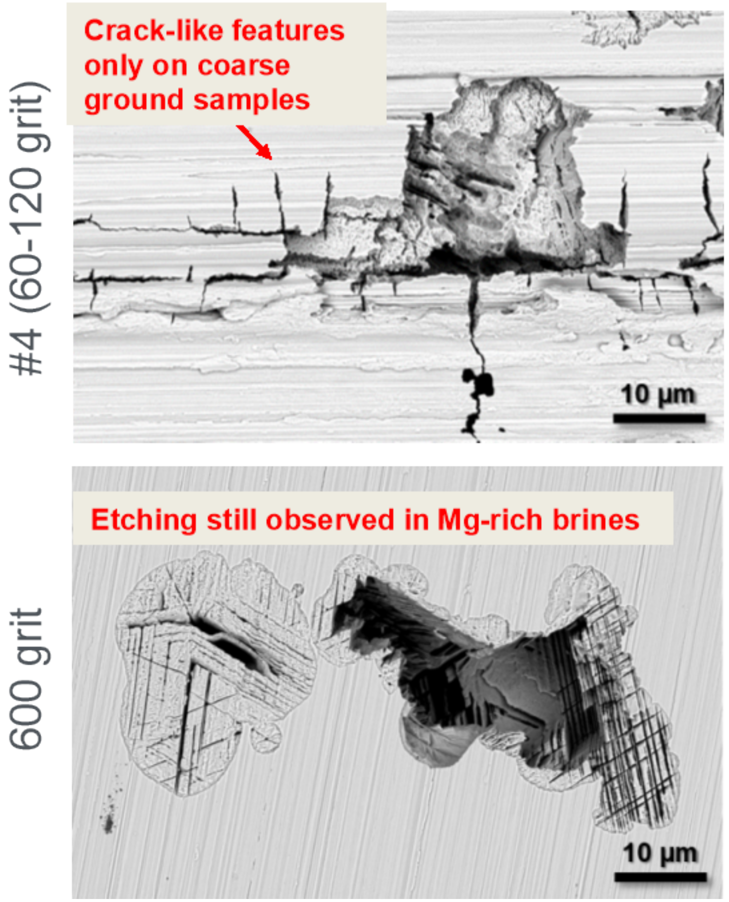
However, pit shape varies widely with brine composition, material, surface finish. How does this affect pit-to crack transition?

*Kondo, Y. (1989). Prediction of fatigue crack initiation life based on pit growth. *Corrosion*, *45*(1), 7-11.

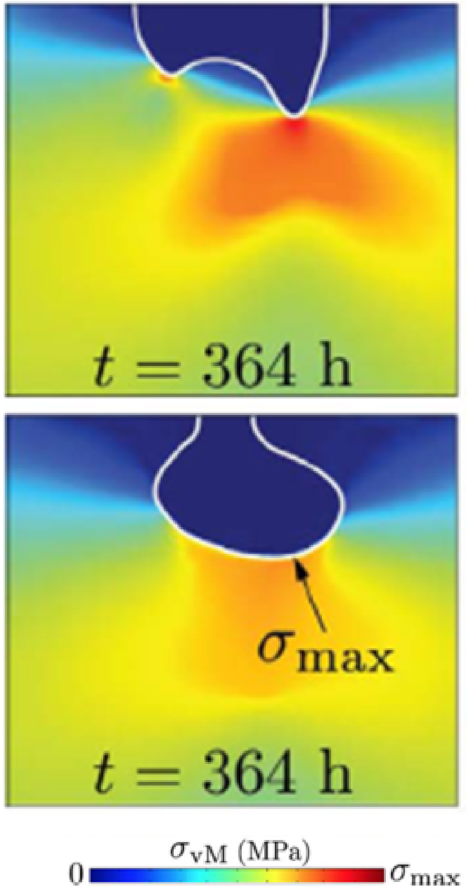
Pit-to-Crack Transition

Effect of brine composition and other factors?

$MgCl_2$-rich brines: etching and irregular pit shapes



Irregular pit shapes = more focusing of stresses



Mai, W., & Soghrati, S. (2017). A phase field model for simulating the stress corrosion cracking initiated from pits. *Corrosion Science*, *125*, 87-98.

Extent and morphology of corrosion pits strongly related to

- *brine composition*
- *surface finish*
- *material composition*

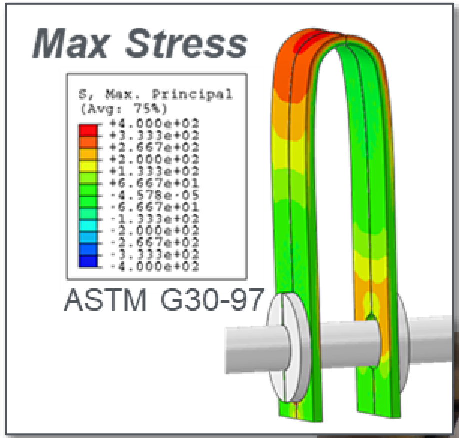
Does pit shape (and hence the above parameters) affect pit-to-crack transition?

Experimental testing of pit-to-crack transition is in progress

Pit-to-Crack Transition

Experimental testing

Large Scale Exposure Testing: U-bend coupons to examine pit-to-crack transition



Example stress modeling

Salt Deposition

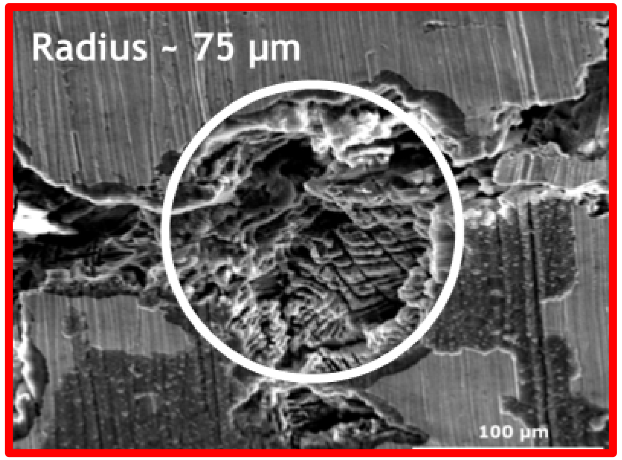
Tensile bar testing in immersed conditions

$MgCl_2$ NaCl

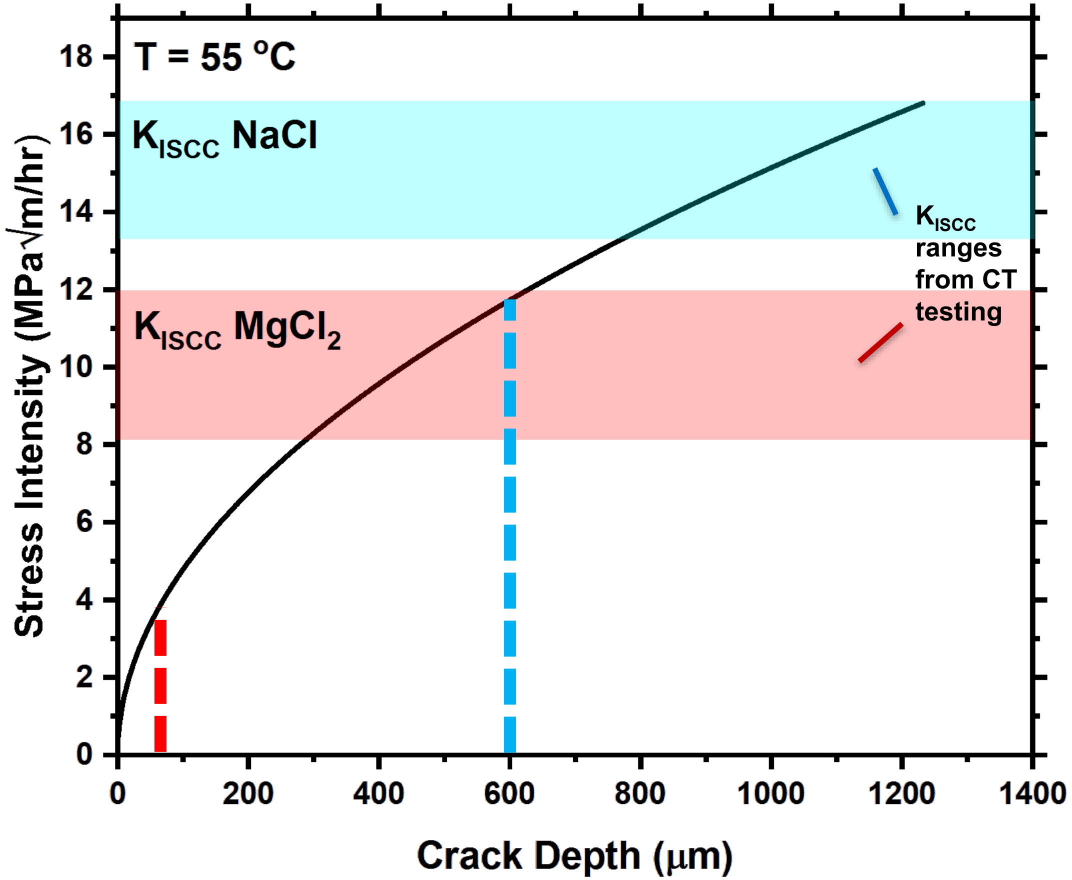
- Numerous cracks observed when exposed to $MgCl_2$
  - No cracks in NaCl even at longer time periods
- Initiating features difficult to identify in $MgCl_2$ solutions

Pit-to-Crack Transition

Tensile testing



$MgCl_2$

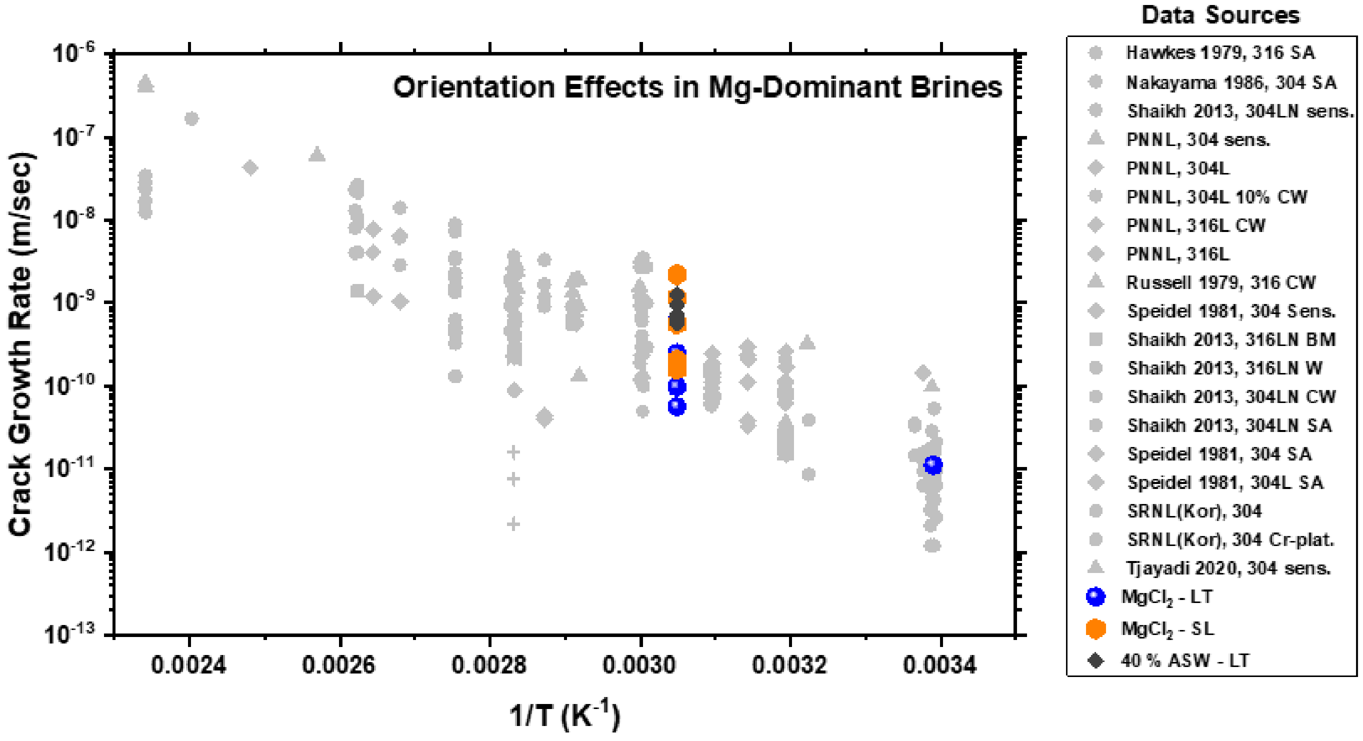


NaCl

Macroscopic Flaw Size does not Dictate Transition in All Environments

Crack Growth Rates in Relevant Brine Environments

Compilation of Literature SCC Crack Growth Rate Data



Available data are highly scattered, mostly from immersed testing. ***Work at PNNL and SNL to reduce uncertainties, and to develop an understanding of CGR in atmospheric SCC.***

Model for crack growth:

- Implement a model that varies T and K:

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

Constants:

α_{crack} = crack growth at T_{ref}

Q = activation energy for crack growth

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

T_{ref} = reference temperature (K) at which α was derived

K_{th} = threshold stress for SCC

β_{crack} = stress intensity factor exponent

Model Inputs:

T = temperature (K) of interest

K = crack tip stress intensity factor

Model Output:

dx_{crack}/dt = crack growth rate

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

$\sigma_{applied}$ = tensile stress

Y = shape factor

x_{crack} = depth

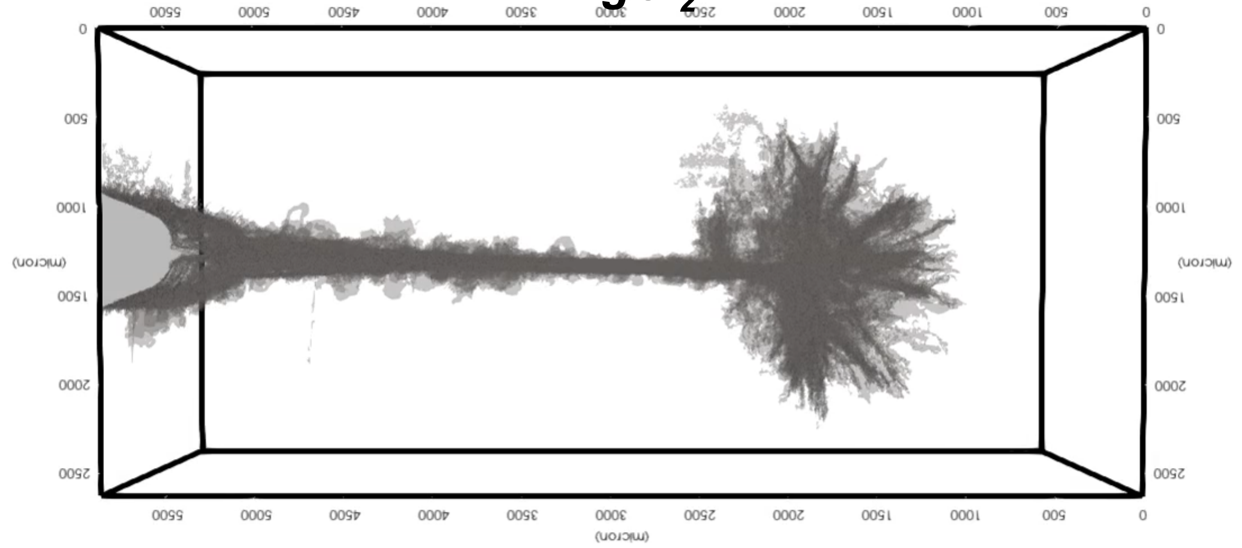
Crack Growth in Relevant Brine Environments

Fractography

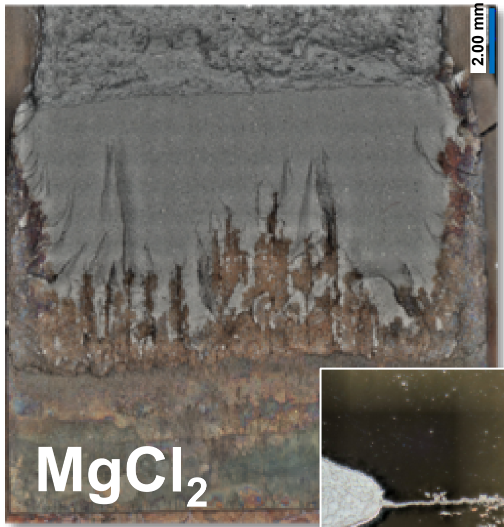
Why important? Crack morphology influences crack blunting (stifling), crack tip stress intensity (K) values, and potentially crack growth rates.

Hydrogen embrittlement in $MgCl_2$ brines?

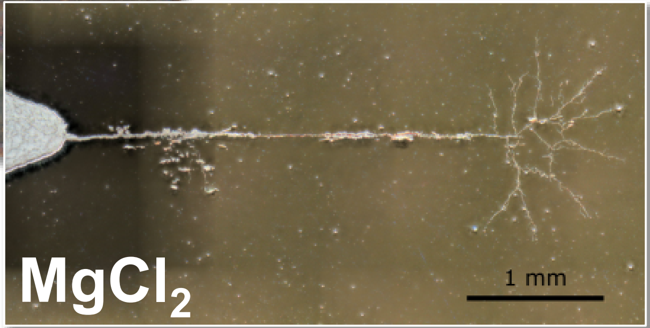
3D image of SCC crack formed in $MgCl_2$

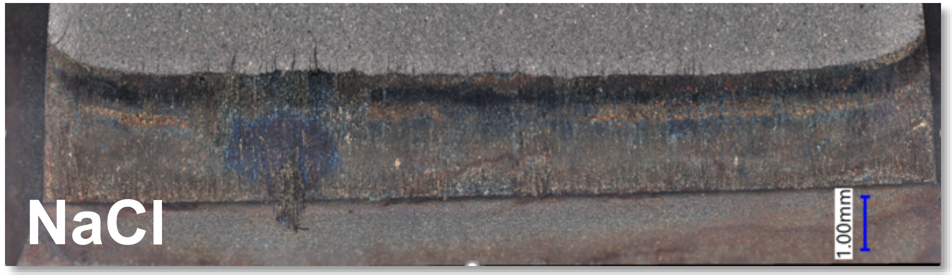


Developing an understanding of DCPD and fractography in saturated salt solutions



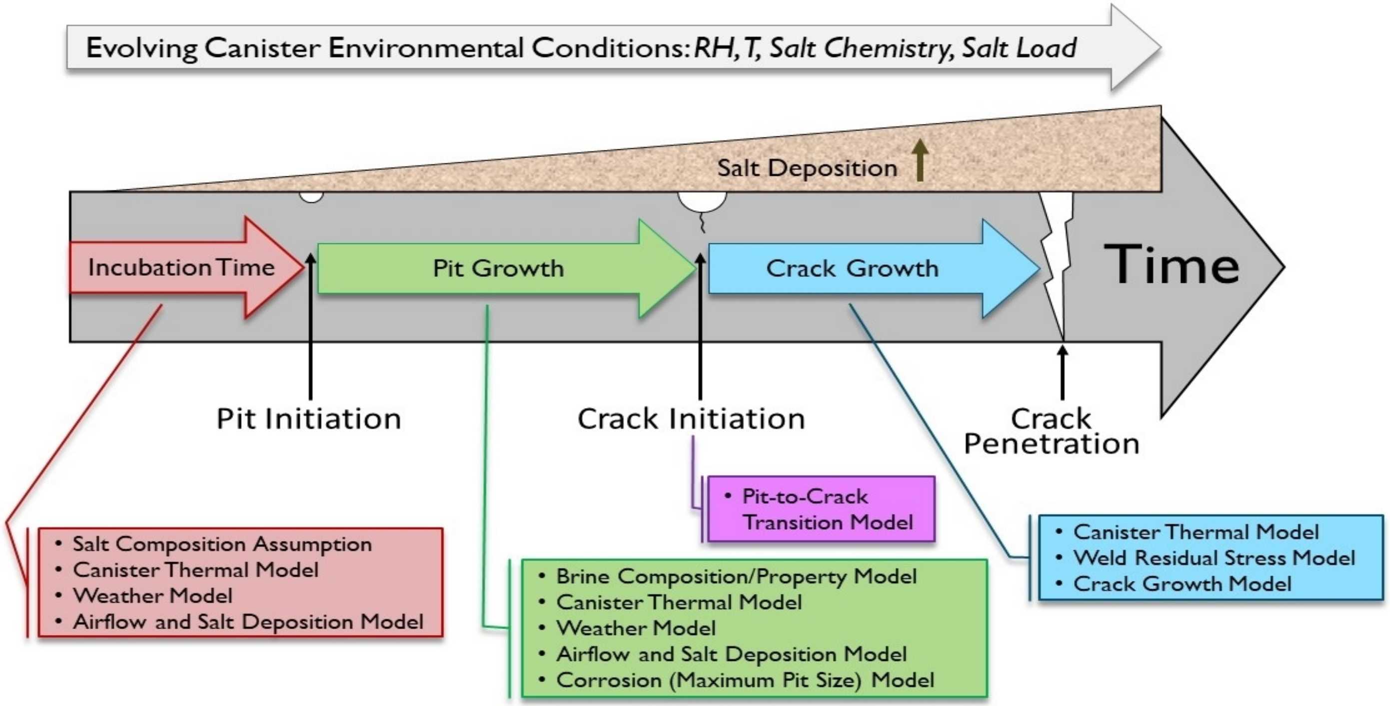
Example: Crack fronts in saturated $MgCl_2$ tests compared to NaCl

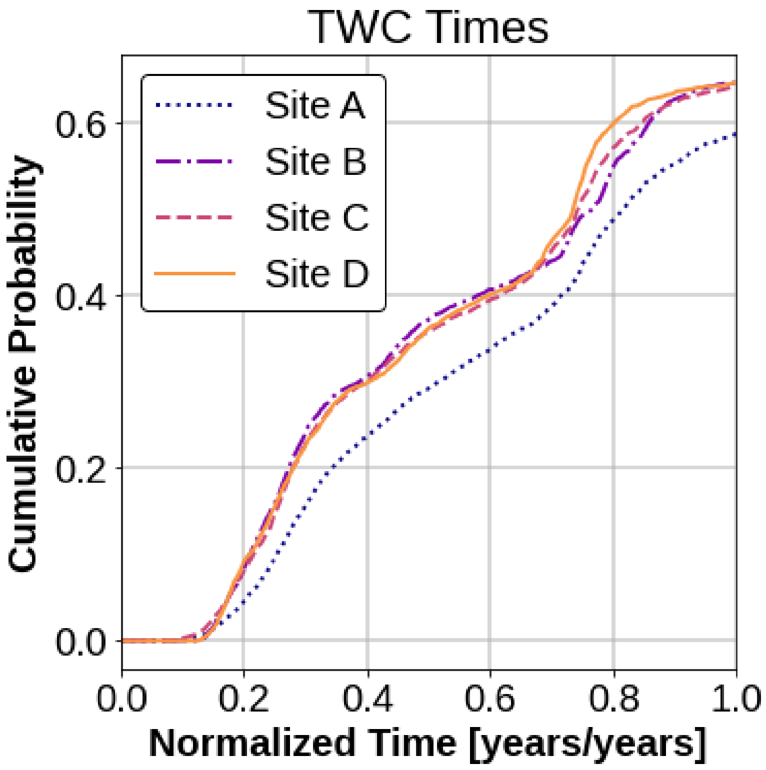




Probabilistic Model for Canister SCC

Major uncertainties that remain in the model





<u>Major uncertainties remaining in model:</u> Environmental controls (brine chemistry, atmospheric vs. immersed) on:

- Timing of brine formation/corrosion (nitrate/chloride mixtures)
- Pit size/morphology and effects on pit-to-crack transition
- K_{th} for SCC
- Crack growth rates under atmospheric conditions

<u>Example simulation</u>:

- 4 sites, 27 cases, varying:
  - Threshold stress intensity factor for SCC (K_{th})
  - Salt deposition rate
  - Limiting RH for corrosion
- 400 realizations per case (43,200 total)

Acknowledgements

DOE and National Labs

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Dr. Jimmy Burns

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