SAND2018-5813PE

# SNF Storage Canister Pitting and SCC: Current Research at Sandia National Labs

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**Sandia National Laboratories**

**Spent Fuel and Waste Science & Technology Program**

**SFWST Meeting**

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# SNL Objectives

***When and where on the canister and across storage sites do we have greatest risk of developing cracks?***

- *Develop quantitative definition of brine properties on canister as function of environmental conditions to inform SCC models and laboratory studies*
- *Understand relationship between surface environment and damage distributions and rates*
- *Quantify impact of material and mechanical environment variability on corrosion and SCC processes*

# Canister SCC: Important Processes

Time

...th | Crack Growth

Begi... Storag... | Crack Initiation | Penetration

**(1) Role of surface environment on pitting damage distributions and pit-crack transition**

*SNL — SCC model...*

*SNL — Surface En...*

**(2) Relationship between surface environment and electrochemical processes driving damage**

*...nd growth, pit-*

*...rowth (stress,*

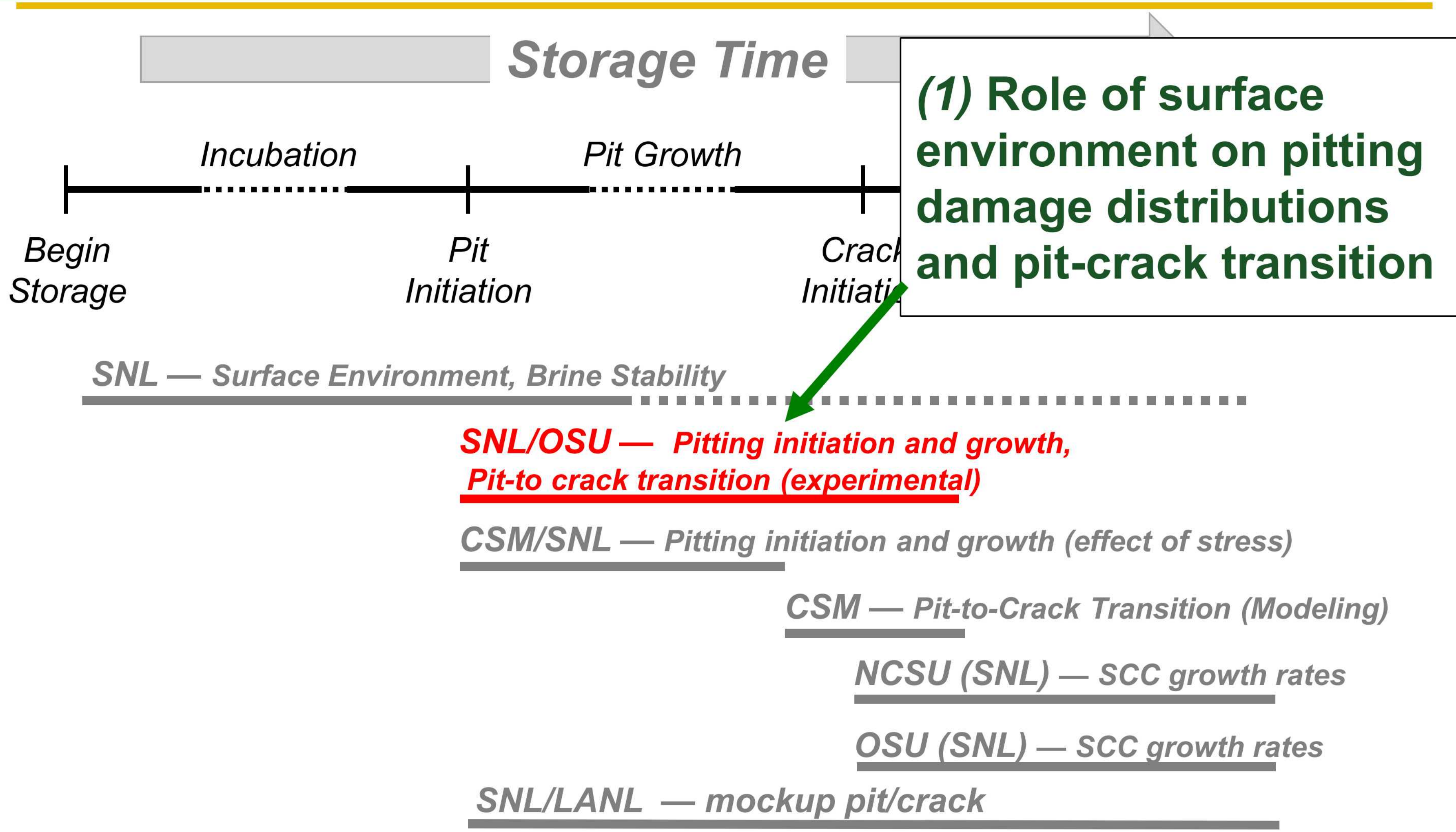
*...microstructu...*

*...g)*

**(3) Pitting/SCC initiation propensity versus canister material state (microstructure, surface finish, stress/strain)**

*SNL/LANL — ...*

# Canister SCC: Important Processes



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# Role of Surface Environment on Pitting Damage and Pit-Crack Transition

**Knowledge Gaps:**

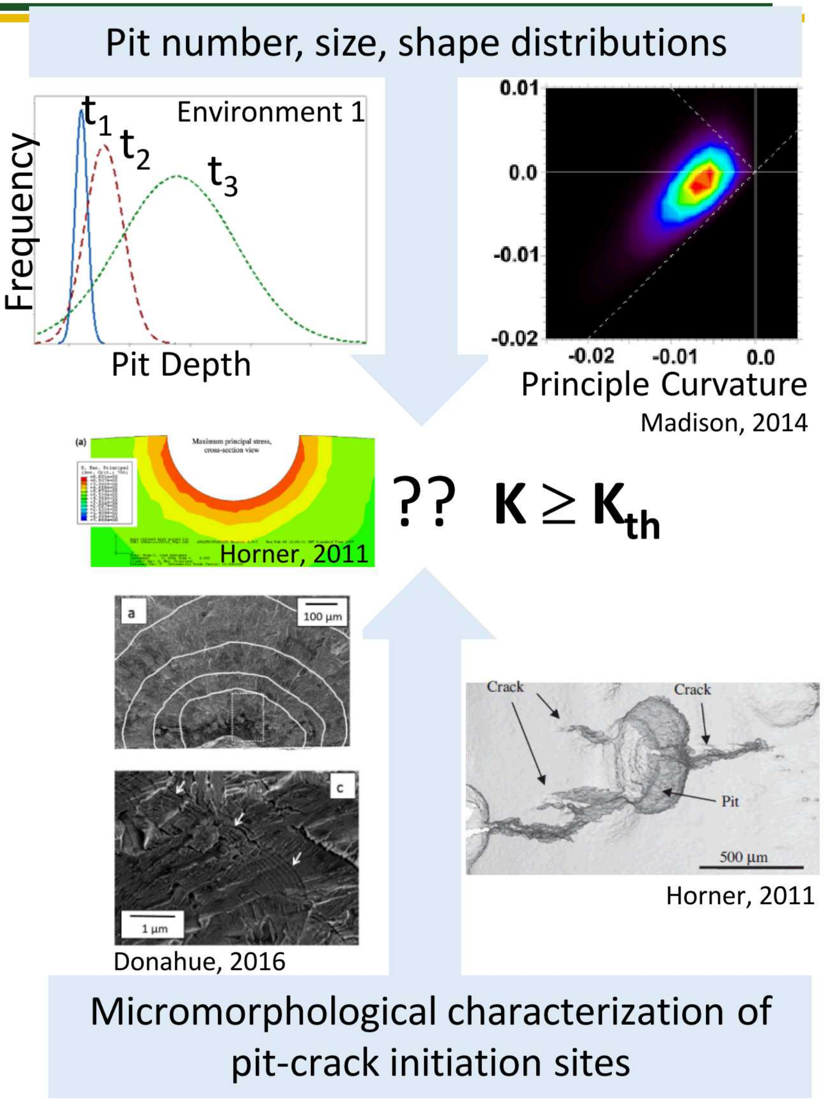
- Environmental condition (T, RH, salt load) affect pit kinetics and pit morphology?
- To what extent can we predict crack initiation based on pit characteristics (shape, size)?

**Impact:**

- Datasets for model development/validation
- Relevance and role of pitting stage as function of environment in SCC

**Approach:**

- Parametric coupon-level pitting experiments in ISFSI-relevant environments
- Constant load marker band SCC tests in same environments to determine corrosion features that act as crack initiation sites



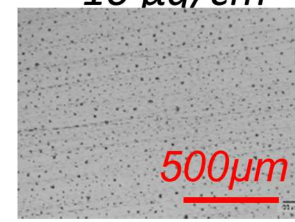
# High throughput approach for building parametric pitting damage datasets

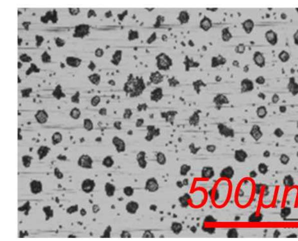
- *1 x 2"* coupons loaded with artificial sea salt and **exposed to fixed environmental conditions for up to 2 years**
  - *304H, mirror and 120 grit "mill" finish*
  - *10 and 300 µg/cm$^2$ artificial sea salt*

| %RH | Temperature (°C) | | | | |
|---|---|---|---|---|---|
| 75 | 35 | | | | 55 |
| 70 | 35 | | | | |
| 65 | 35 | | | | |
| 60 | 35 | | | | |
| 55 | 35 | 40 | | | |
| 50 | 35 | 40 | | | |
| 45 | 35 | 40 | 45 | | |
| 40 | 35 | 40 | 45 | | 55 |
| 35 | 35 | 40 | 45 | 50 | |
| 30 | 35 | 40 | 45 | 50 | |

Inkjet printing for high-throughput salt loading

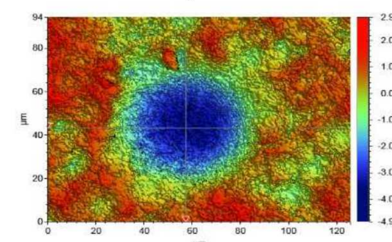
*10 µg/cm$^2$*

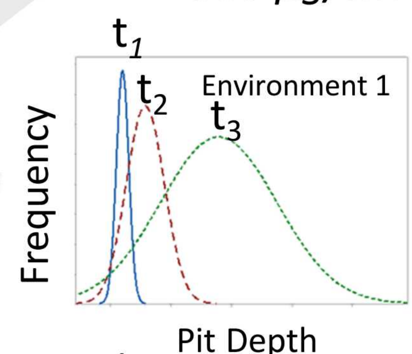



*300 µg/cm$^2$*

*J. Locke, OSU*



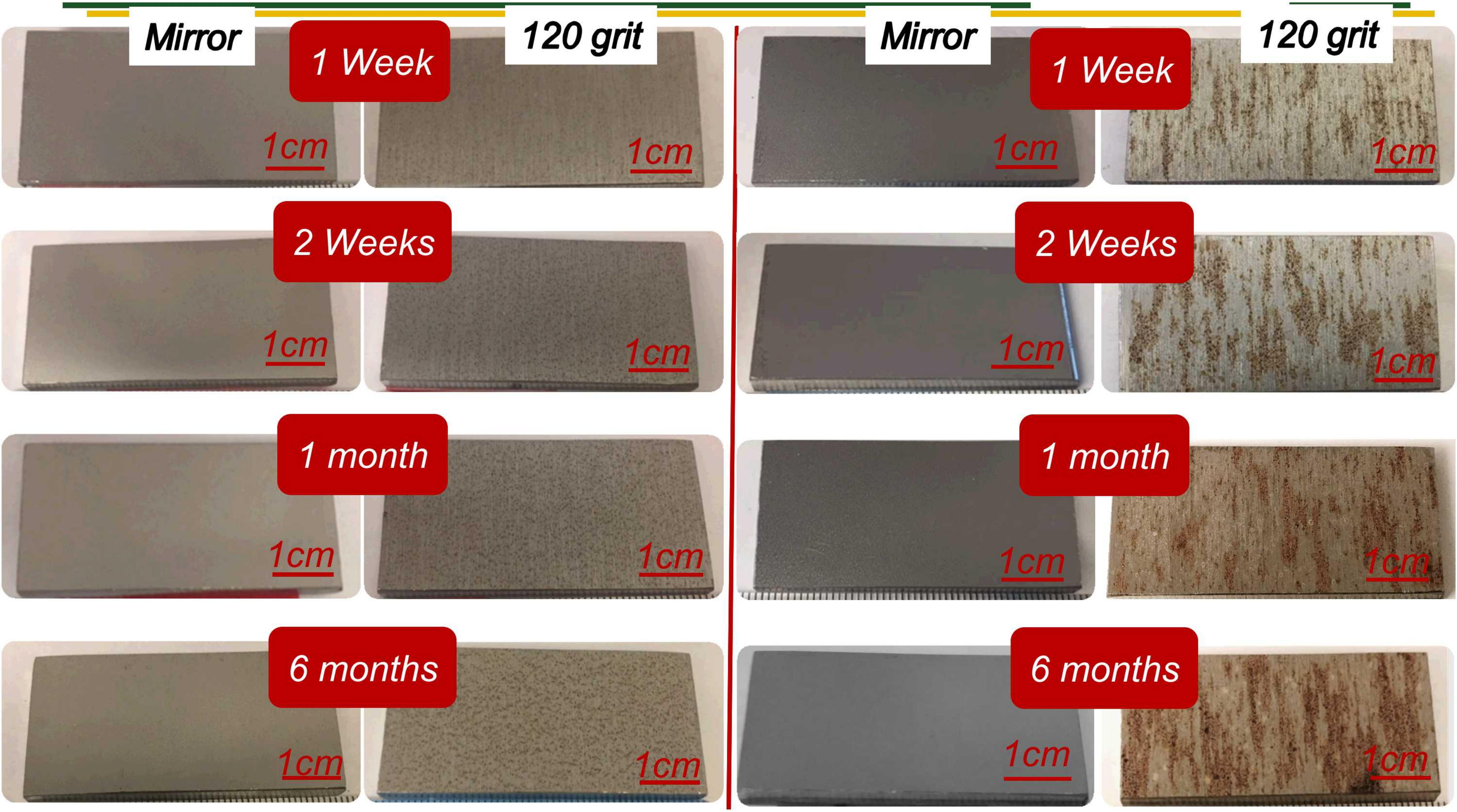



Pit morphology analysis w/ optical profilometry

# 300μg/cm$^2$ salt loading density samples after environmental exposure

35°C, **40%RH**

35°C, **75%RH**



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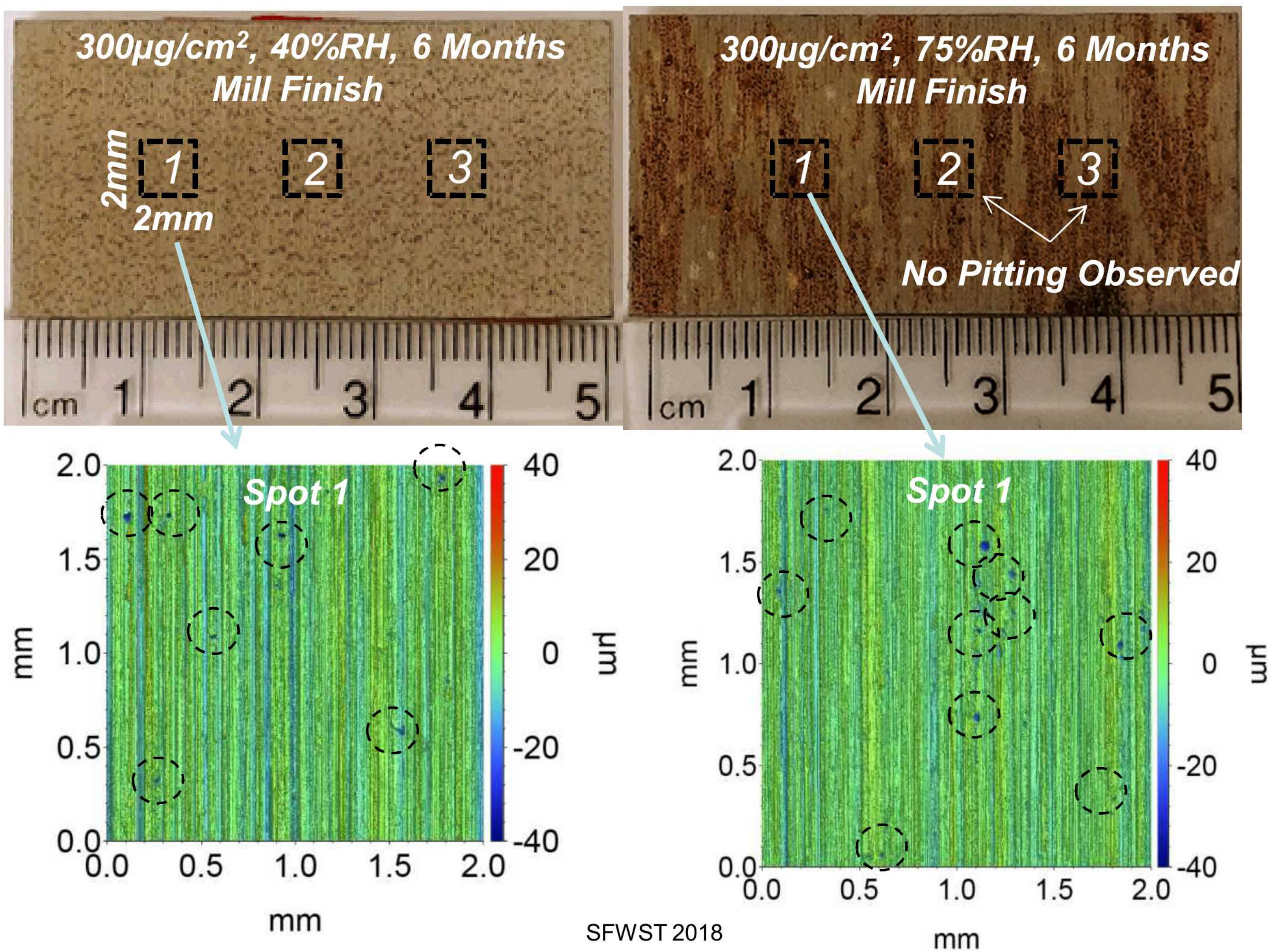
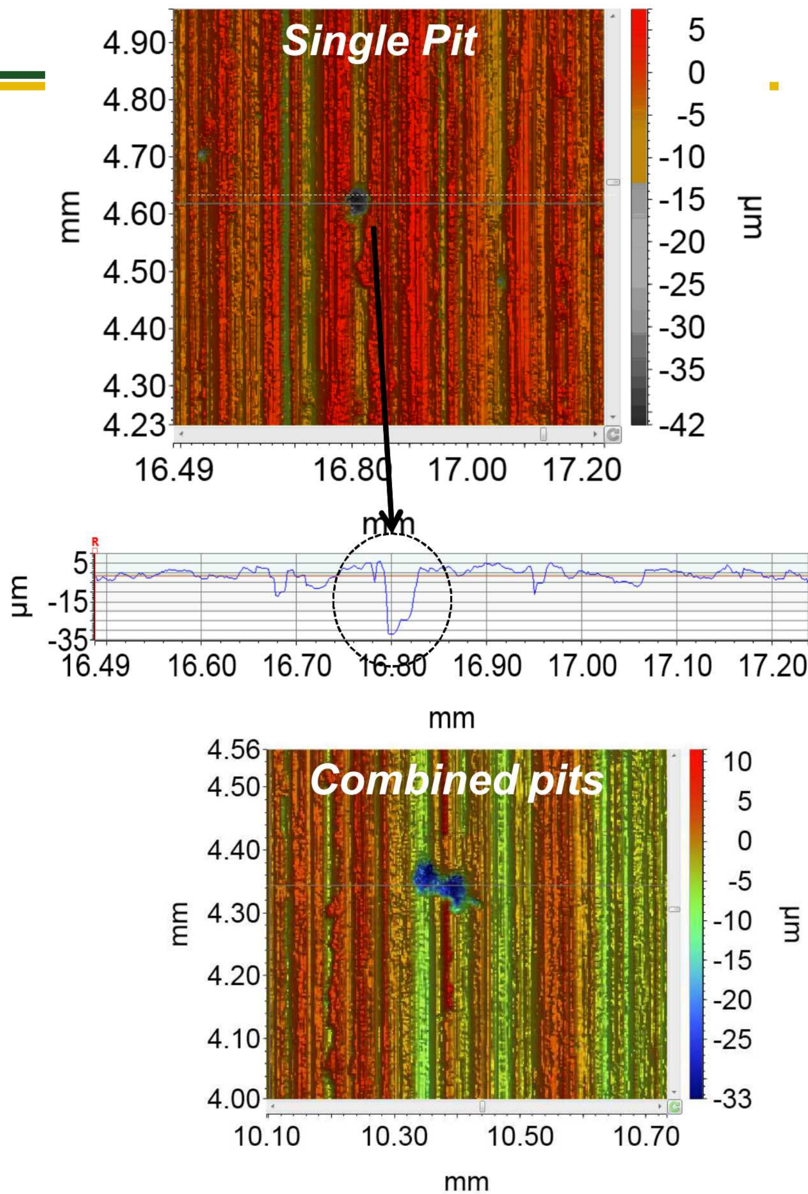
# Small area scans used to measure pit distributions across surface

- Threshold condition used to find pits: Area > 75µm$^2$ , Depth > 8µm
- Individual pits manually filtered from damage sites.



# Small area scans used to measure pit distributions: 40% RH, $35^{o}$C

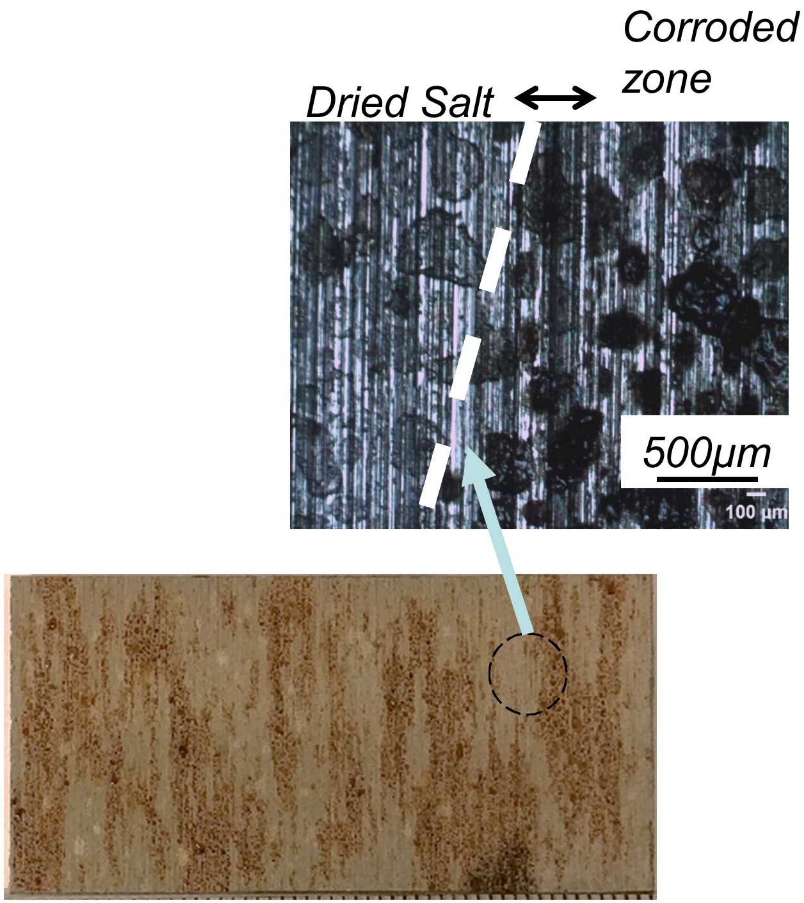
| | | Pit Depth | | Diameter | |
|---|---|---|---|---|---|
| Time | # of Pits | Avg. (µm) | Max (µm) | Avg. (µm) | Max (µm) |
| 1 Week | 4 | 15 | 21 | 24 | 35 |
| 2 Weeks | 12 | 15 | 19 | 23 | 40 |
| 1 Month | 16 | 15 | 22 | 29 | 52 |
| 6 Month | 53 | 22 | 44 | 32 | 89 |



# *Small area scans used to measure pit distributions: 75% RH, 35°C*

| | | Pit Depth | | Diameter | |
|---|---|---|---|---|---|
| Time | # of Pits | Avg. (μm) | Max (μm) | Avg. (μm) | Max (μm) |
| 1 Week | 0 | < 8 | < 8 | - | - |
| 2 Weeks | 0 | < 8 | < 8 | - | - |
| 1 Month | 0 | < 8 | < 8 | - | - |
| 6 Month | 29 | 20 | 41 | 24 | 82 |

Larger area scans needed for a statistically accurate damage distribution



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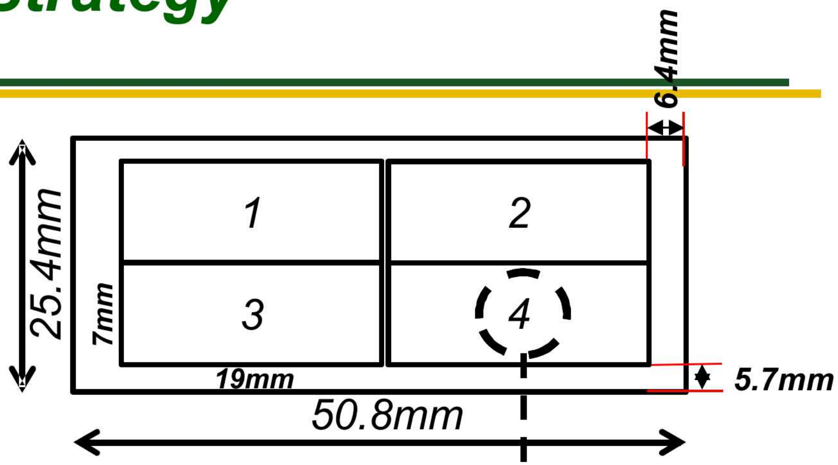
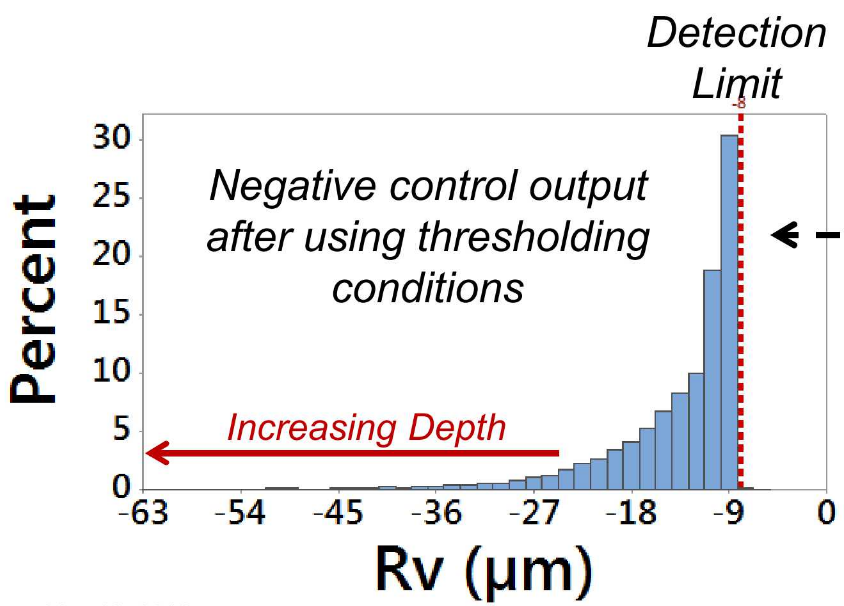
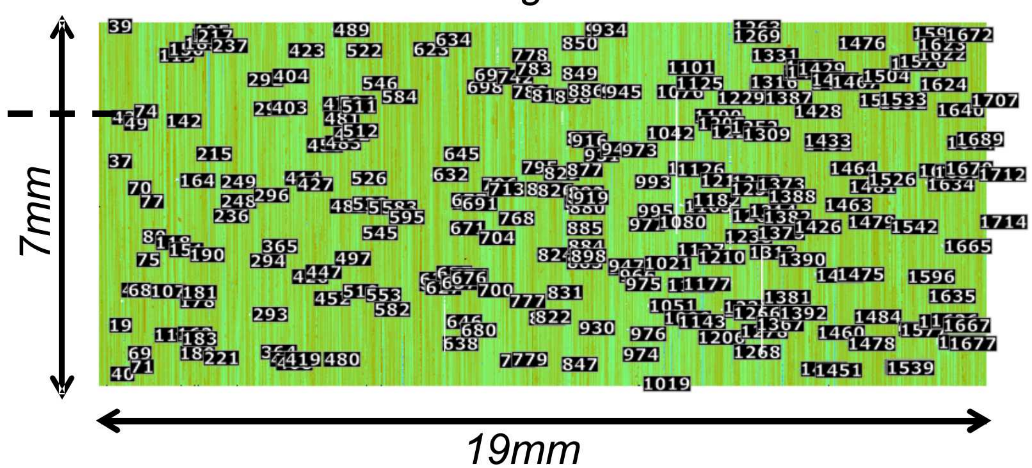
# Currently Developing Large Area Analysis Strategy

- *Samples split into four sections and measurements made across each quadrant*
- *Data filtering parameters:*
  - *Depths > 8µm below zero*
  - *Areas > 75µm²*
- *Damage sites are the same depth as pits*




*1 quadrant showing output from image analysis software – Negative Control*



# Role of Surface Environment on Pitting Damage and Pit-Crack Transition

**Knowledge Gaps:**

- Environmental condition (T, RH, salt load) affect pit kinetics and pit morphology?
- To what extent can we predict crack initiation based on pit characteristics (shape, size)?

**Impact:**

- Datasets for model development/validation
- Relevance and role of pitting stage as function of environment in SCC

**Approach:**

- Parametric coupon-level pitting experiments in ISFSI-relevant environments
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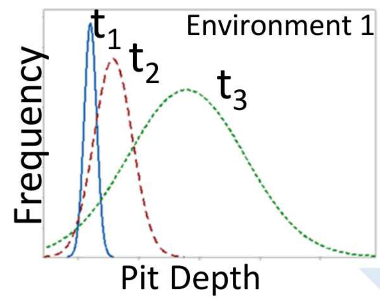
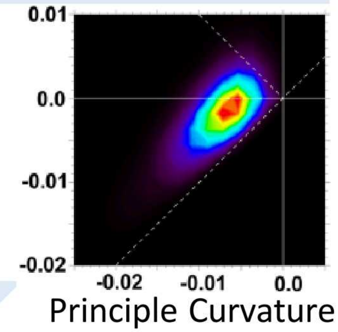
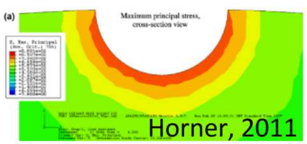
Pit number, size, shape distributions



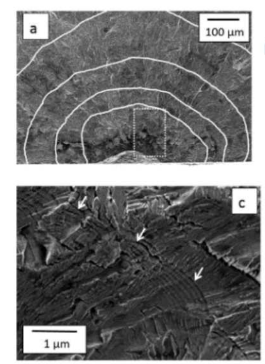




Madison, 2014

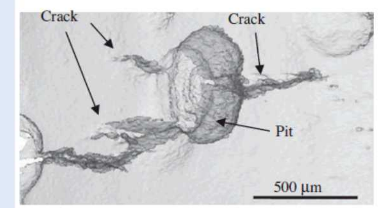



Horner, 2011

?? $K \geq K_{th}$




Donahue, 2016




Horner, 2011

Micromorphological characterization of pit-crack initiation sites

# Understand the pit morphology leading to SCC initiation

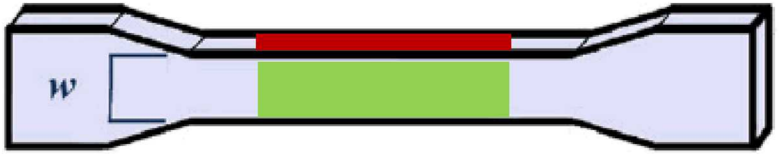
*J. Locke, T. Weirich, OSU*

Variables:

- *Atmospheric exposure parameters and salt loading density*
- *Naturally occurring corrosion morphology*

Method: SCC testing

- *Gauge length of longitudinal tensile bars will be loaded with salt and corroded in a humidity controlled chamber.*
  - *Load salt and corrode side of coupons (red).*
  - *Remove from humidity chamber and print salt on face of coupon (green).*
  - *Extra salt on face will contribute electrolyte to the crack tip during propagation.*
- *Constant load with intermittent high R ripple fatigue loads during SCC tests to determine corrosion features that act as crack initiation sites.*

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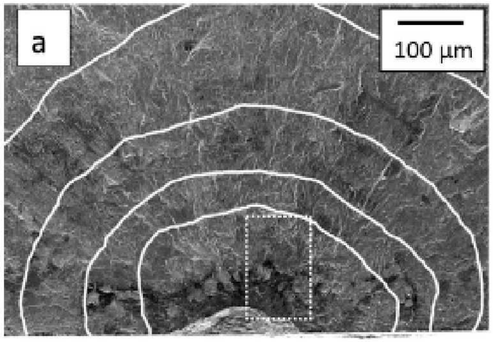
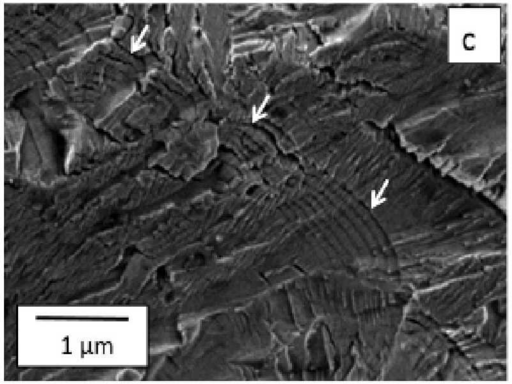
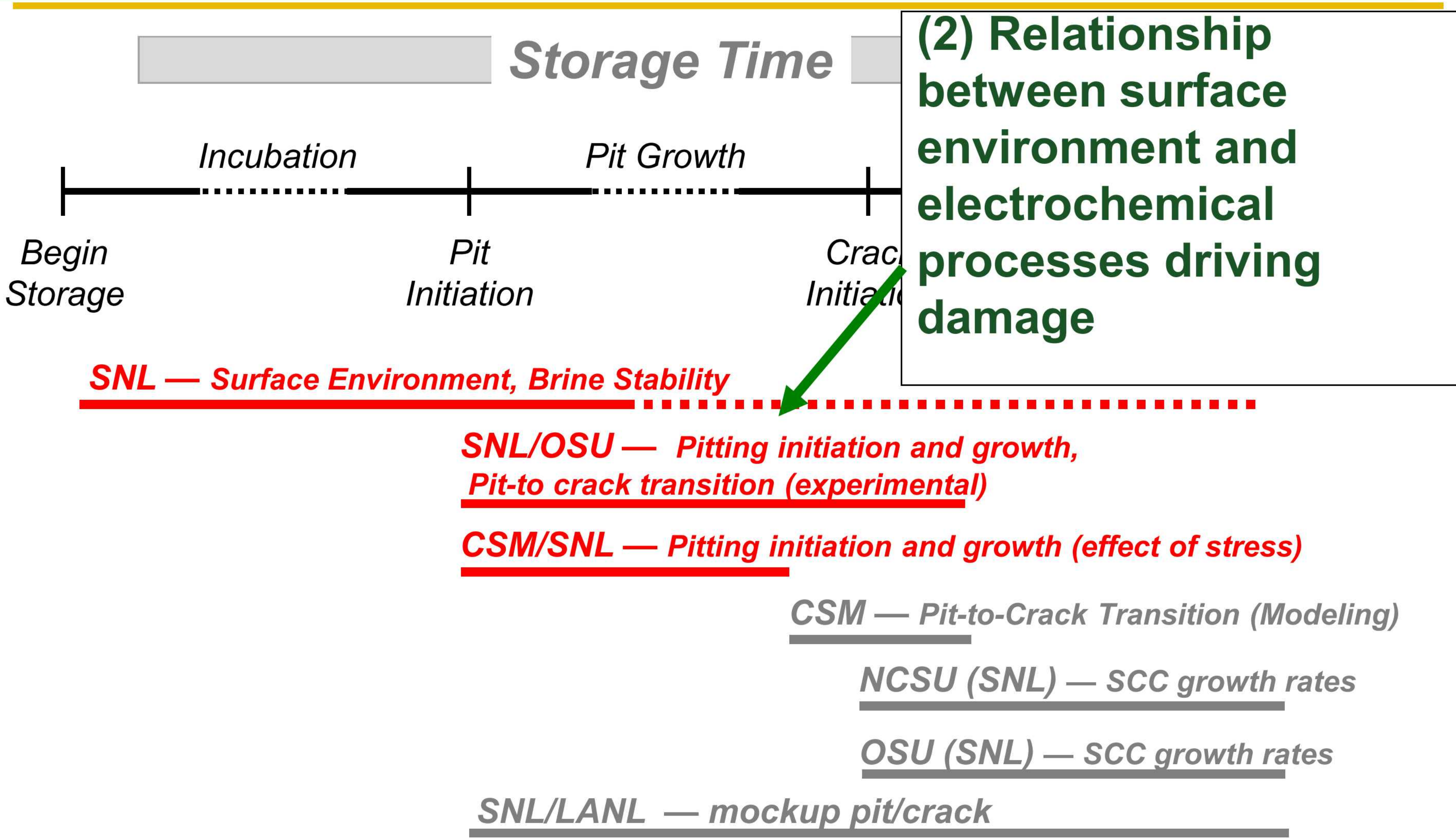
*Fracture surface of Custom 465-H950*




*J. R. Donahue and J. T. Burns, International Journal of Fatigue 91 (2016), 79-99.*

# Canister SCC: Important Processes



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# How do Surface Conditions Impact Electrochemical Processes Driving Corrosion/SCC?

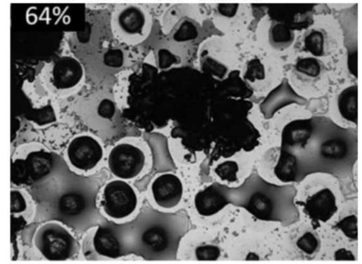
## Impact:

1) Relevance and accessible limits of existing deterministic damage models
2) Conditions under which initial salt chemistry, RH and T can be used to predict kinetics and damage
3) Inform laboratory tests- salt loading, time

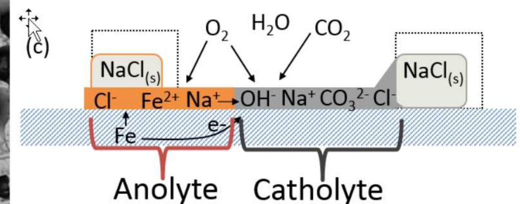
## *Approach:*

- Define physical/chemical electrolyte properties during corrosion
- Quantify impact on electrochemical corrosion processes controlling rate

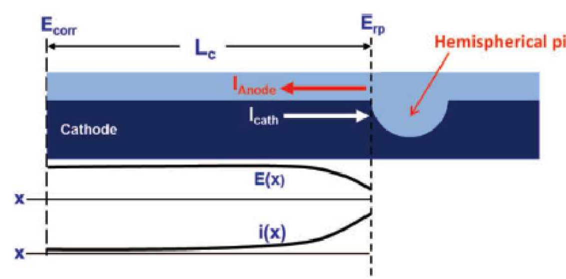
*NaCl on steel*



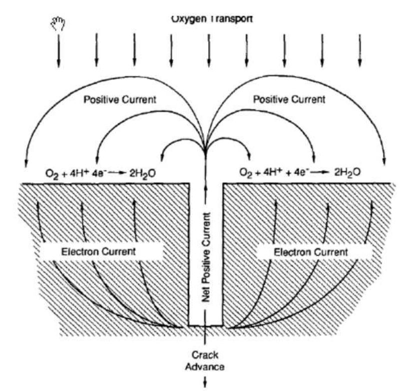

*Schindelholz, 2014*




*maximum pit size* *crack growth rate (?)*




*Chen et al. 2008*




*MacDonald, 1991*

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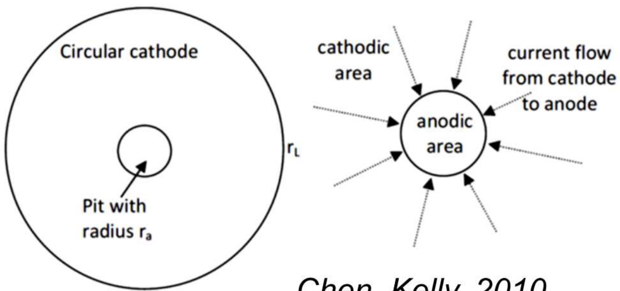
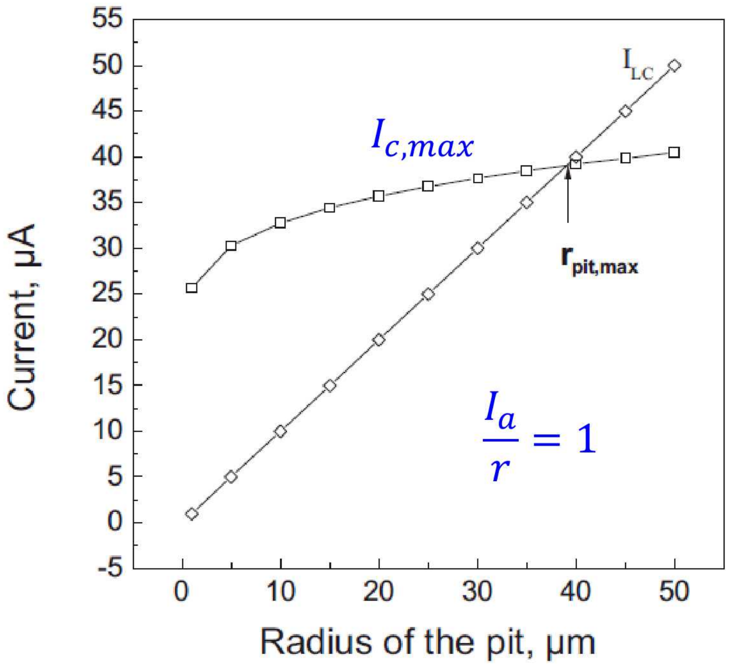
# Prediction of Maximum Pit Size from Brine Characteristics and Electrochemical Kinetics




Chen, Kelly, 2010




Max. cathode current — Brine conductivity — Brine layer thickness — Cathodic Kinetics

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

**Challenge:**

Information on electrochemical parameters lacking for expected canister brine conditions (electrolyte thickness and chemistry)

**Assumptions:**

1. Continuous brine layer
2. Hemispherical pit
3. Cathodic and anodic kinetics independent of time (fixed electrolyte conditions)

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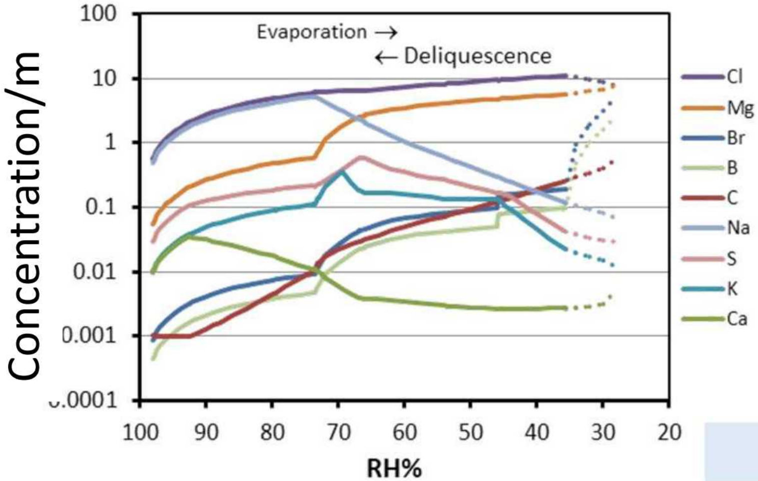
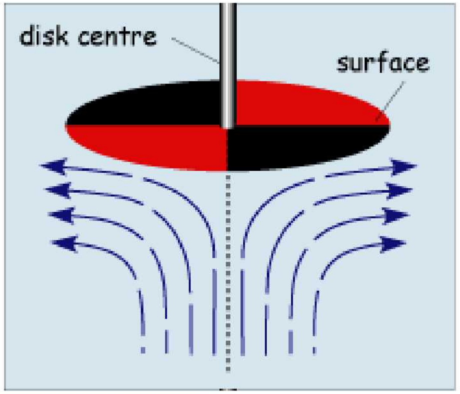
# Measurement of Electrochemical Parameters in Analog Electrolyte Brines as f(T,RH, salt load, chemistry)

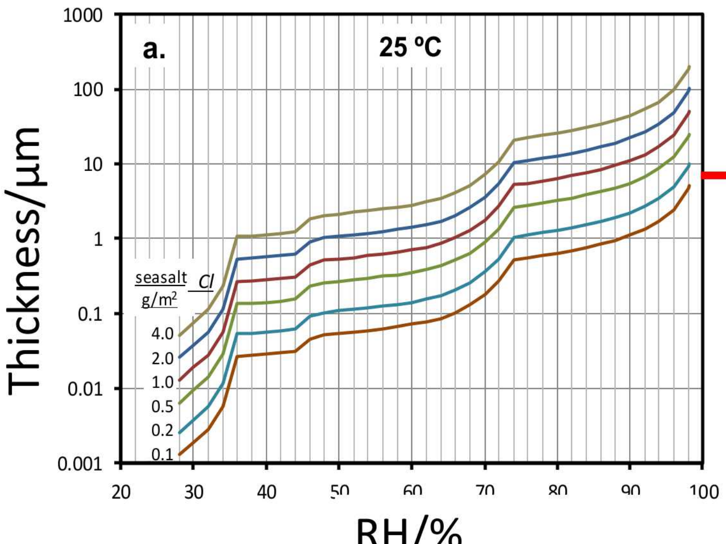



*SNL/UVA*

Rotating disc electrode to simulate brine layer thickness > 1 µm

Cathodic polarization curves



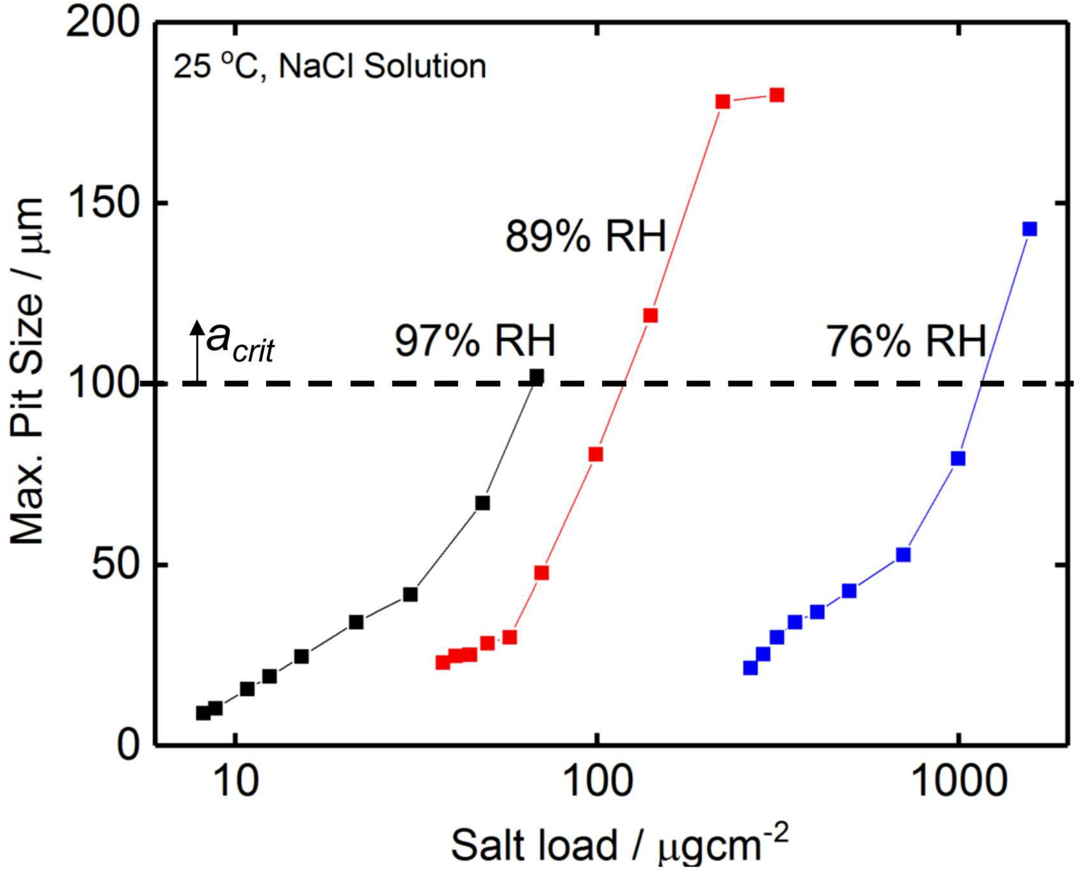

$$\ln I_{c,max} = \frac{4\pi k W_L \Delta E_{max}}{I_{c,max}} + ln\left[\frac{\pi e r_a^2 \int_{E_{corr}}^{E_{rp}} (I_c - I_p)dE}{\Delta E_{max}}\right]$$

# Cracking Susceptibility Based on Maximum Pit Size Predictions

*Conceptual* calculations of **hemispherical** maximum pit size derived from cathodic kinetics measured in NaCl brines

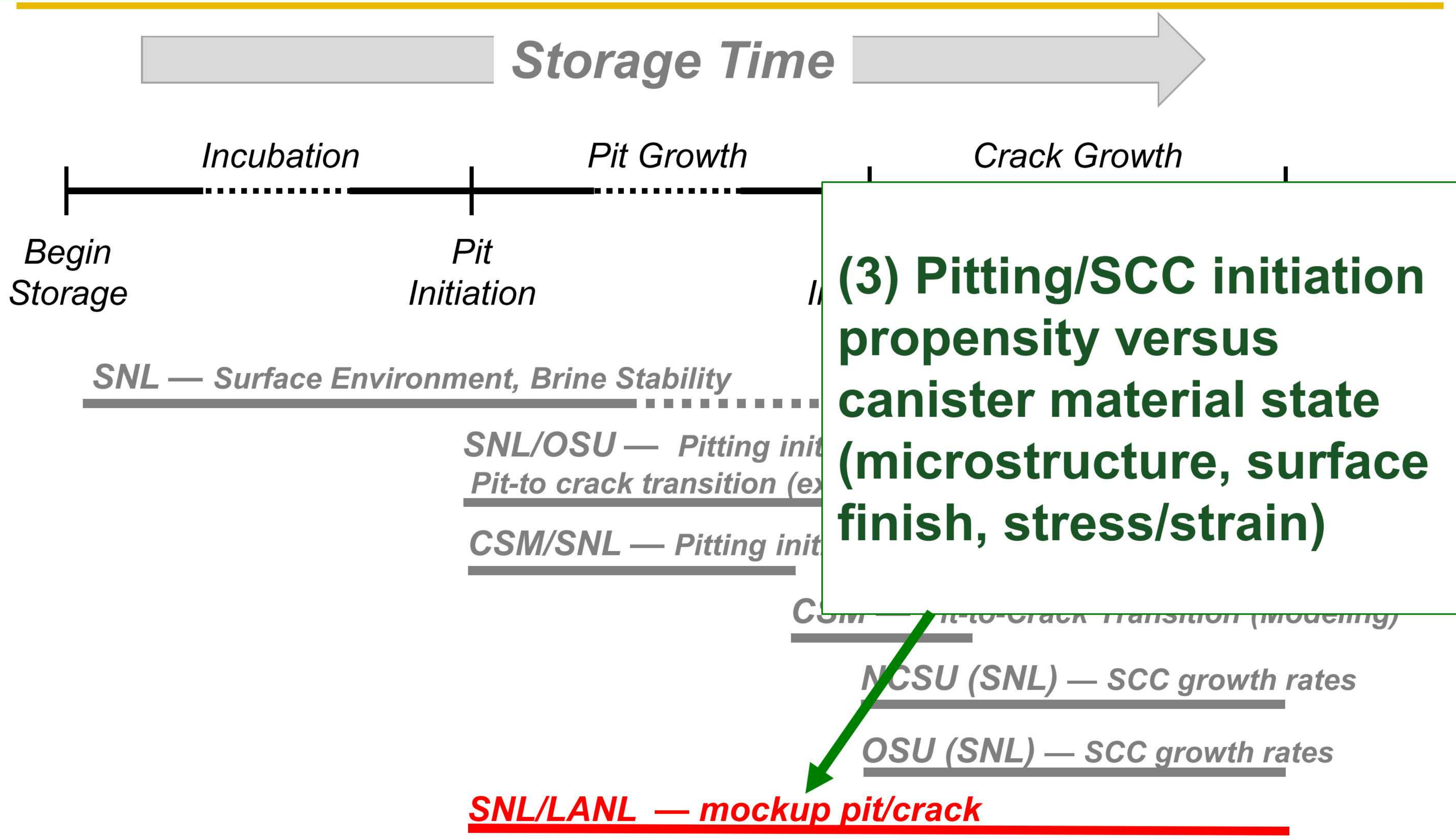
**Need to understand relevant limits of numerous assumptions including**:

- lab = field $E_{chem}$ parameters?
- Electrolyte and surface attributes are constant or changes due to corrosion or other processes are inconsequential
- Effects of Material features (microstructure) and stress/strain are negligible




*Example: $K_{ISCC}$ = 5 Mpa·m$^{1/2}$, σ= 500 MPa*

# Canister SCC: Important Processes



# What is pitting/SCC initiation propensity given materials features/state?

**Knowledge Gap:**

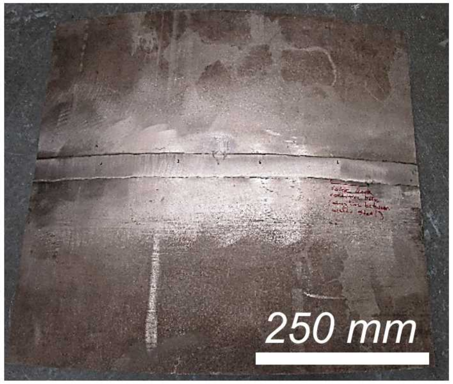
Relationship between canister-relevant material characteristics (microstructure, stress/strain) and relative pit/crack susceptibility

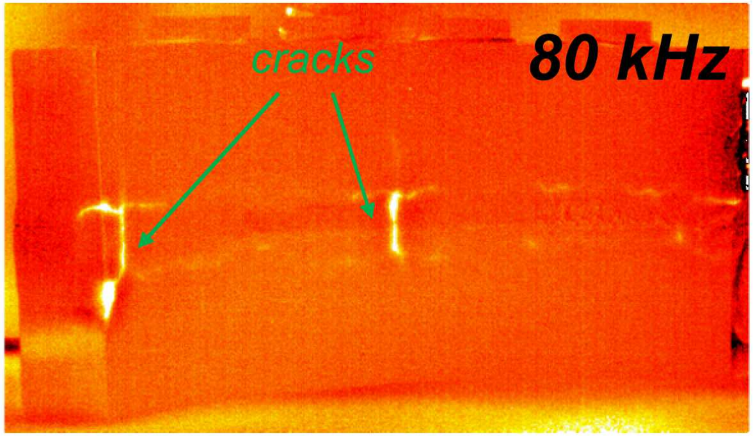
**Impact:**

1) Knowledge of canister-relevant extreme case corrosion/SCC behavior
2) Inform laboratory experiment design and extrapolation of lab data to field conditions
3) Benchmark, inform SCC models

**Approach:**

- Exposing mockup plates loaded with $MgCl_2$ (> 400 µg/cm$^2$) to 4 % RH, 80$^O$ C
- Characterize pit and crack distribution over course of exposure
- Postmortem characterization of pit and crack geometry in relation to stress and material




*Vibrothermography crack detection method -courtesy M. Remillieux, LANL*

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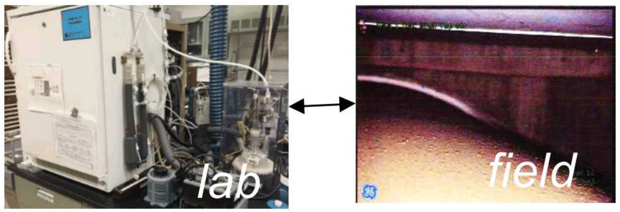
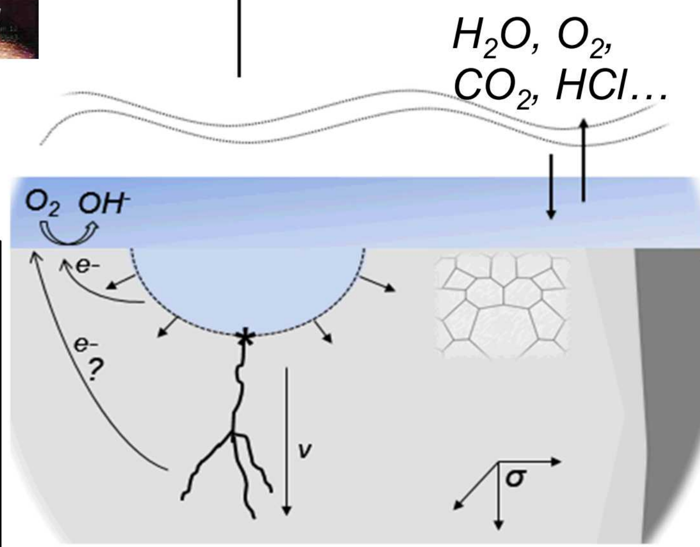
# When and where on the canister and across storage sites do we have greatest risk of developing cracks?

## How representative are lab conditions?




*Surface/atmospheric chemistry, RH variation*



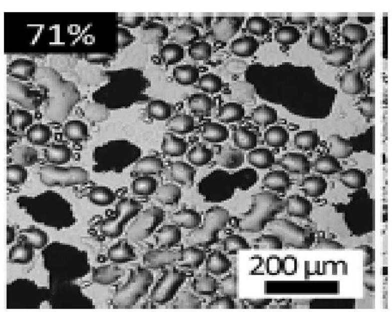

## What are relevant and accessible model limits?

$$\frac{da}{dt} = \begin{cases} \alpha \exp\left[-\frac{Q_g}{R}\left(\frac{1}{T} - \frac{1}{T_{ref}}\right)\right] & \text{for } RH \geq DRH \text{ and } K_I > 0 \\ 0 & \text{for } RH < DRH \text{ or } K_I \leq 0 \end{cases}$$

EPRI, 2017

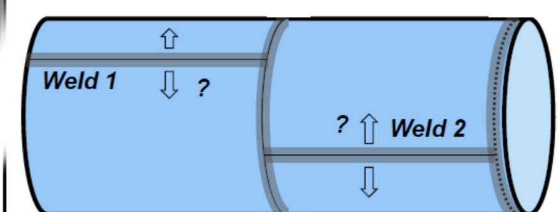
*Benchmarking datasets, bounding limits, test assumptions, model confidence*

## Environmental control of damage distribution and rates?




*Salt loads and distributions, temperature, RH*

## Where and when to focus inspection?




*Variations in canister surface environment, material properties, and stress*