

An Approach to Model Abstraction of Canister Corrosion Damage in Management of Spent Nuclear Fuel and High-Level Waste

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**Due to U.S. Government Shutdown, Dr. Ahn could not participate
at the Conference.**

An Approach to Model Abstraction of Canister Corrosion Damage in Management of Spent Nuclear Fuel and High-Level Waste

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Corrosion Damage in Nuclear Waste Systems**

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- Note:

The NRC staff views expressed herein are preliminary and do not constitute a final judgment or determination of the matters addressed or of the acceptability of any licensing action that may be under consideration at the NRC.

Outline

- **Radionuclide Confinement: Persistence of Passive Film (with respect to Low General Corrosion Rate under Anoxic Condition)**
- **Limited Radionuclide Release: Canister Opening Area by Localized (Pitting and Crevice) Corrosion and Stress Corrosion Cracking (SCC)**
- **Localized Corrosion: Initiation, Latent (Delayed, in comparison with transient during nucleation) Repassivation, and Active Dissolution**
- **Pit-Induced SCC**
- **Propagation of Single SCC Crack**
- **Multiple SCC Cracks: Possible Maximum Opening Area of Canister**

Introduction

- **Pits, crevice, and SCC corrosion is addressed in this presentation.**
- **Under certain conditions, following initiation of corrosion, a passive film develops and arrests the pit corrosion process area; a maximum crack opening of SCC may be limited.**
- **This presentation assumes no intervention in the process and conservatively evaluates potential consequences of pit, crevice and stress corrosion crack mechanisms.**

Example Material and Environment

- **Stainless steel and nickel-based alloys**
- **Chlorides**
- **Assumes no inspection and remediation**

Radionuclide Confinement

- **Persistence of passive film**
 - **required conditions for passive film development**
 - **steady-state thickness (Macdonald, et al., since 1981; Bataillon, et al., 2012; Ahn et al., 2008)**
 - **steady-state chemistry (Pensado, et al., 2002)**
 - **impurity effects (Ahn, et al., 2008; Jung and Ahn, 2011)**

(with respect to anoxic conditions for corrosion allowance metals)

Limited Radionuclide Release

- **Pit, crevice or SCC opening area could result in limited amount of radionuclide release.**



Radionuclide Source Term:
degradation of spent nuclear
fuel and cladding



Transport Path:
air, groundwater



Area Boundary:
dose

Localized Corrosion: Initiation and Latent Repassivation

- **Initiation: deterministic (electrochemical), stochastic, and mass transport**
- **Repassivation: latent (Macdonald and Urquidí-Macdonald, 1992)**

Pit survival rate $\sim \text{EXP} [-\lambda (t - t_{\text{ind}})]$

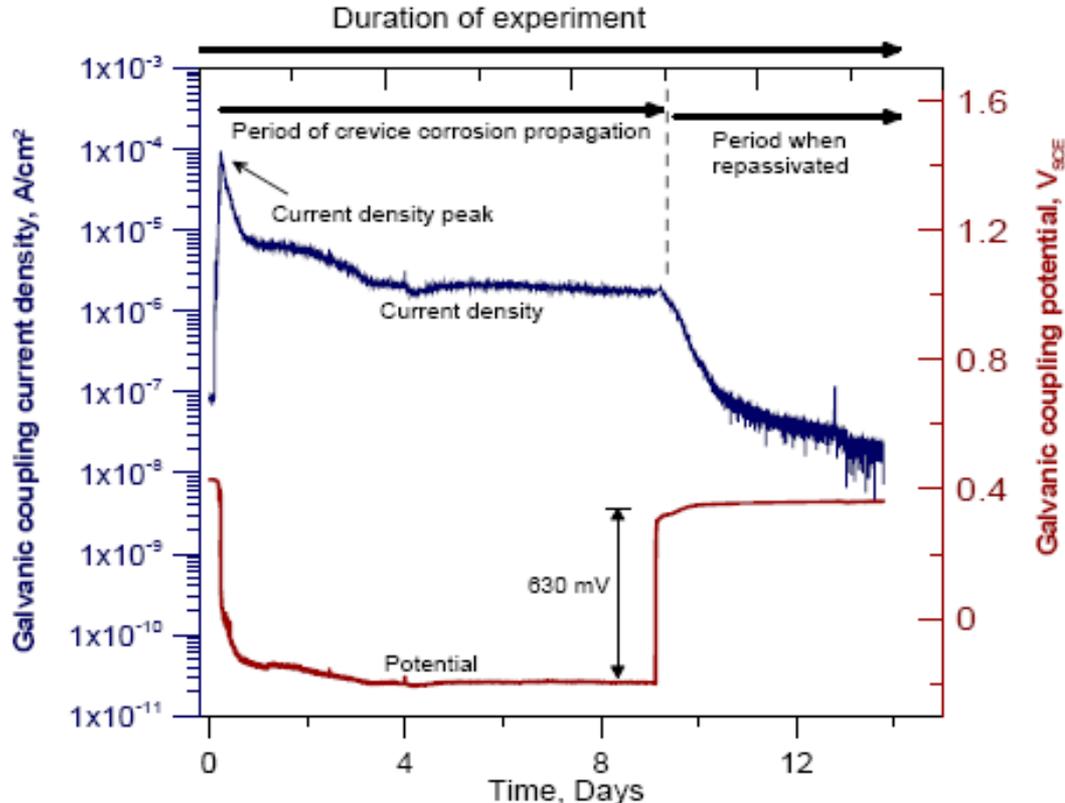
λ : latent repassivation constant

t : time

t_{ind} : pit induction time

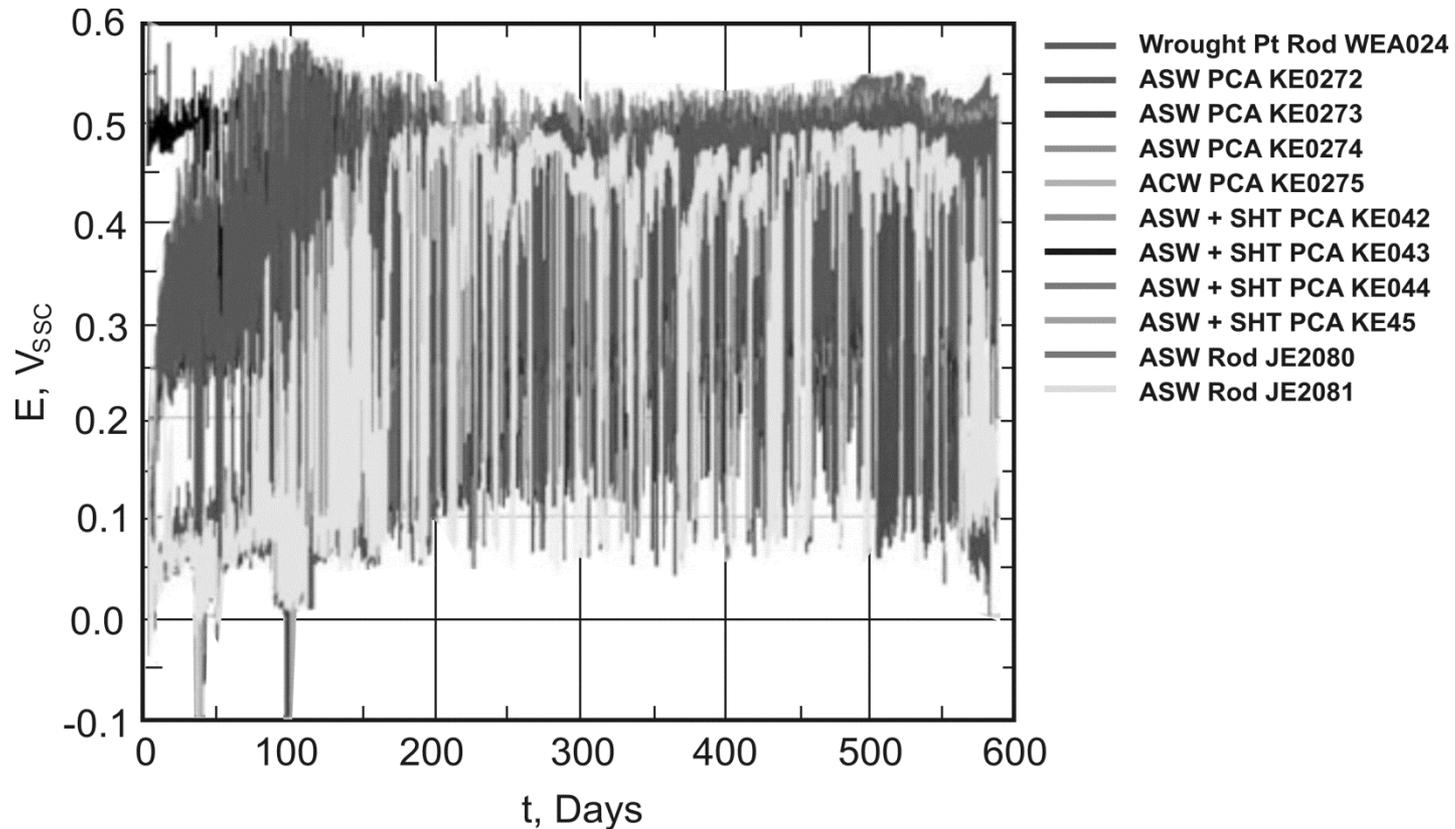
- **Cathode capacity (Shukla, et al., 2011; Cui, et al., 2005)**

Localized Corrosion: Pit Repassivation in Crevice



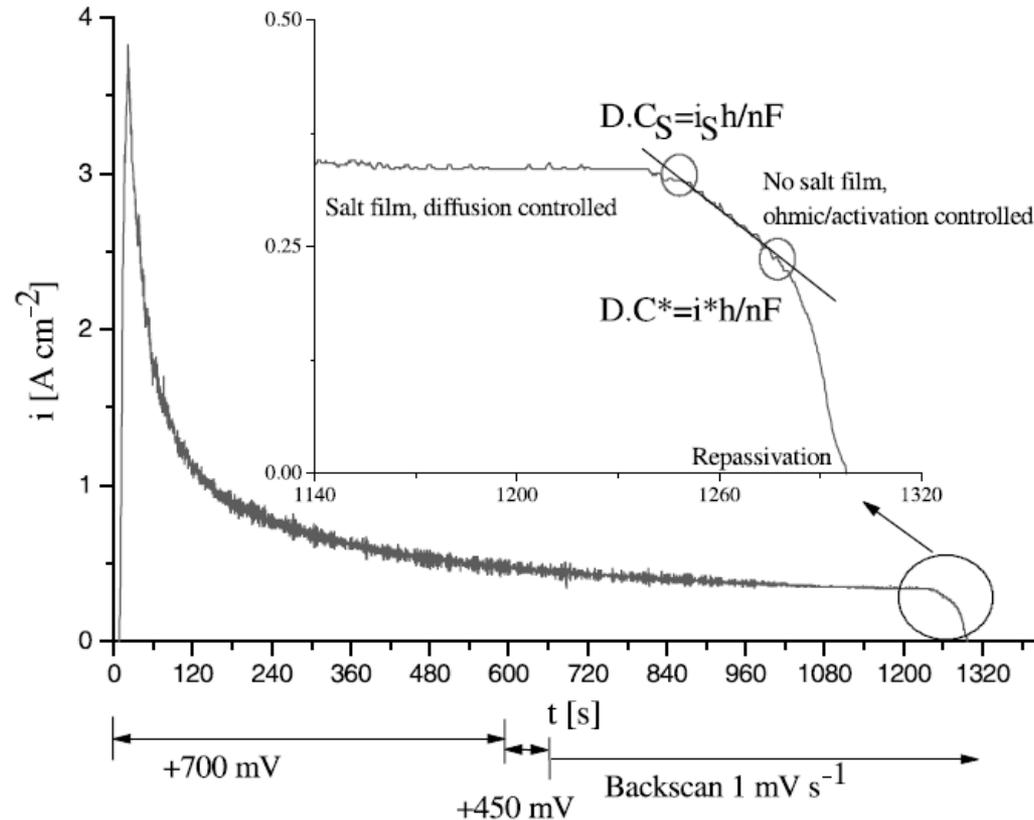
- Measured Current Density and Potential Using the Single Crevice Assembly for an Alloy 22 Cylindrical Specimen Galvanically Coupled to a Large Alloy 22 Plate in 5 M NaCl Solution with the Addition of $2 \times 10^{-4} \text{ M CuCl}_2$ at $95 \text{ }^\circ\text{C}$ [$203 \text{ }^\circ\text{F}$], mV(SCE) (He and Dunn, 2005)
- A large number of pits initially formed in the crevice were repassivated under open-circuit conditions

Localized Corrosion: Pit Repassivation outside Crevice



Corrosion potential as a function of immersion days for Alloy 22 specimens in 18 M CaCl₂ + 0.9 M Ca(NO₃)₂ (Rodriguez et al., 2007) (reprinted by permission from NACE International)

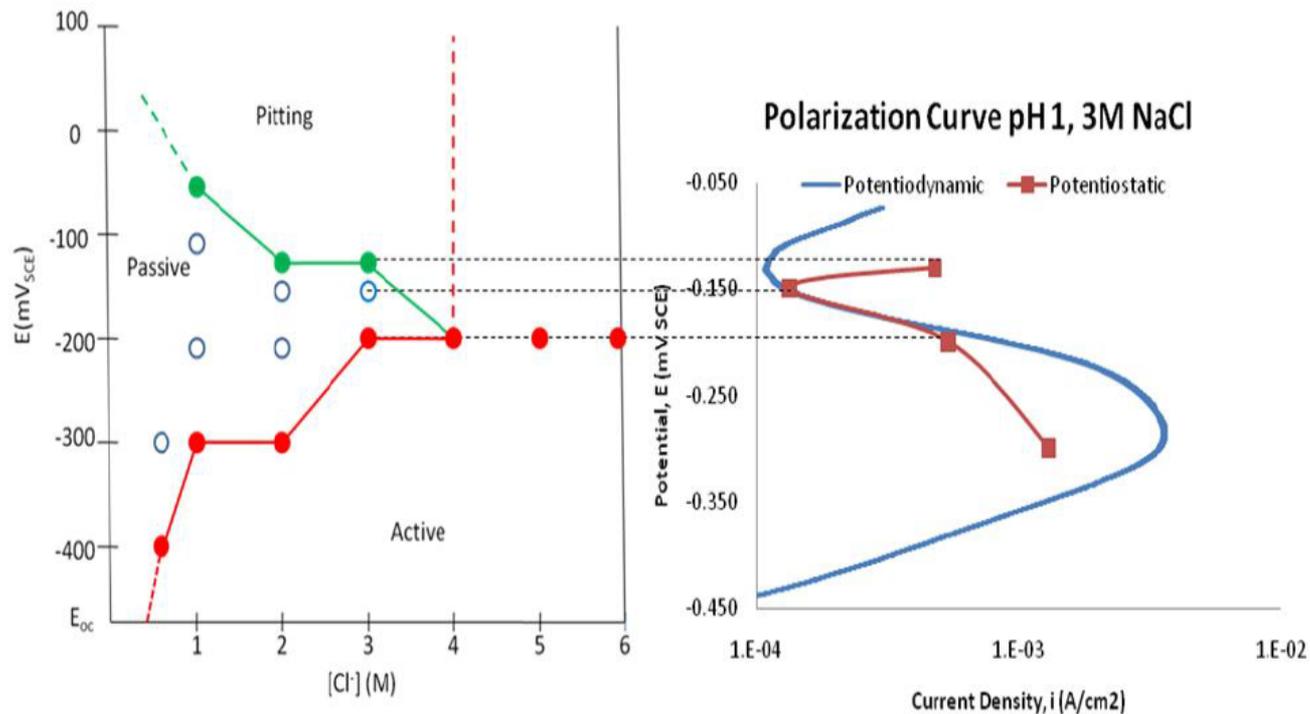
Localized Corrosion: Repassivation of Pitting Corrosion



Current –time response for a pencil electrode of 304 stainless steel in 1 M NaCl at 15° C (59 ° F). The potential was stepped to 700 mV (SCE) for 10 minutes, stepped to 450 mV for 1 minute, and then backscanned until repassivation (Ernst and Newman, 2002). Pit depth at 600 mV at 15° C (59 ° F) for 440 s ~ 0.10 to 0.15 mm. D: diffusivity of dissolved cations, h: diffusion length, C_S : saturated concentration, C^* : critical concentration, i_S : saturated current density, i^* : critical current density

Localized Corrosion: Active Dissolution

Active dissolution of 304 stainless steel under salt fog or spray tests (DeForce, 2004; Enos and Bryan, 2013)



Potential versus chloride concentration corrosion map for 304 stainless steel In pH 1, showing regions of active, passive and pitting corrosion. No active peak at pH 2. (DeForce, 2010) (permission by The Pennsylvania State University: the voltage in the right should be V, not mV)

Pit-Induced SCC

Probability	0.001	0.05	0.25	0.75	0.95
K (MPa m ^{1/2})	0.43	1.57	2.59	4.57	6.94

Cumulative probability of stress intensification factor from GoldSim software (Ahn, 2013),
 $K(\text{MPa m}^{1/2}) = \pi^{1/2} \times \text{stress} \times (\text{crack size})^{1/2}$, 1 MPa m^{1/2} = 0.91 ksi in^{1/2}

- **Pitting: A Precursory Step for SCC (AMEC, S. Africa NPP, 2013)**
- **Data on SCC with Pitting in a Chloride-Bearing Environment with Sufficient Stress and Aqueous Conditions (EPRI, 2005).**
- **Cumulative Probability of Stress Intensification Factor Using Observed Pit Size and an Example Weld Stress (Shirai, et al., 2011).**
- **Possible Stress Intensifications Fall in the Range of Values in Measured Laboratory Tests (EPRI, 2005).**

Propagation of Single SCC Crack : Observation

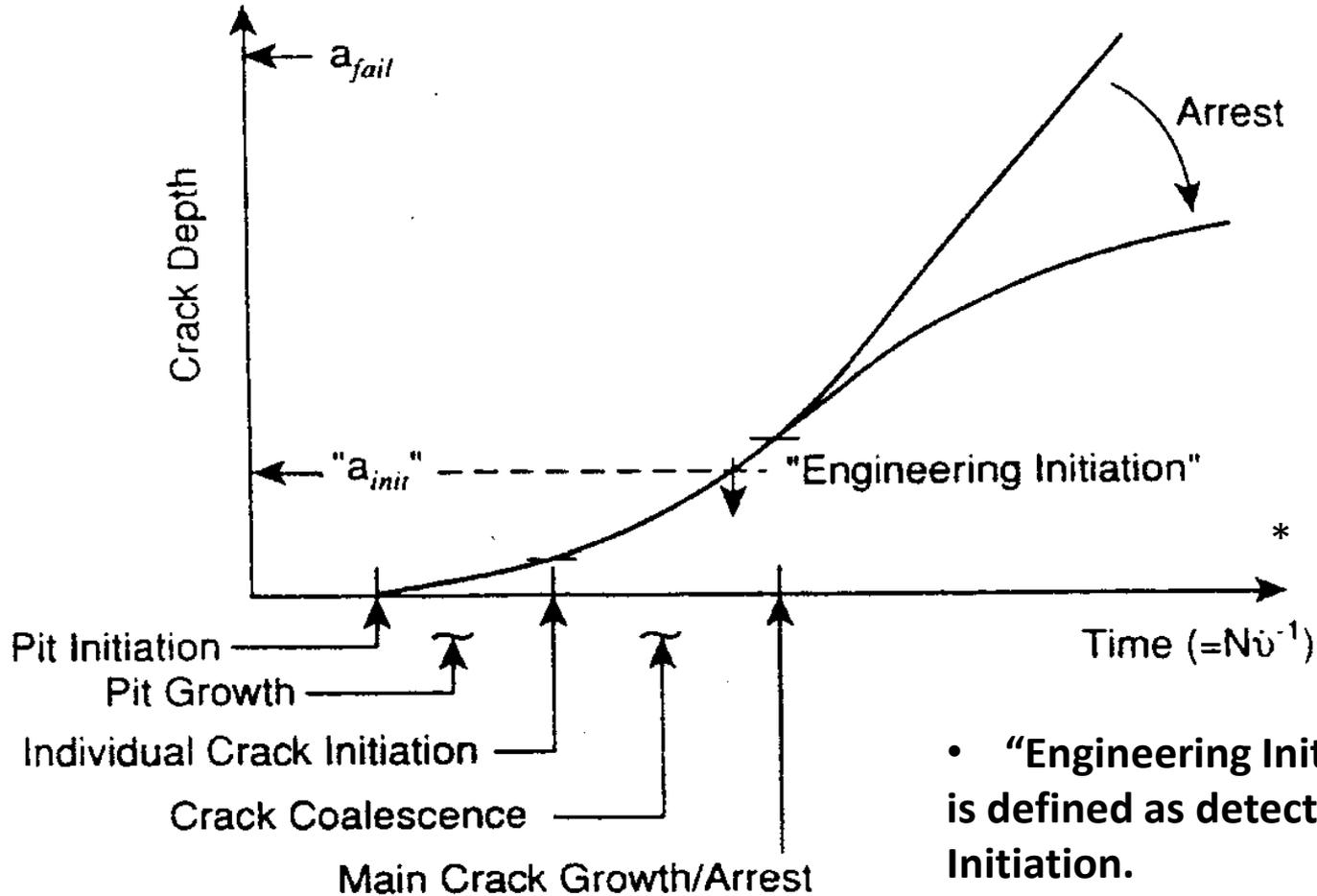
- **Stress along the thickness of the canister varies; will be redistributed during crack propagation, with plasticity change.**
- **Crack branching or tortuous crack path.**
- **Rapid decrease of residual stress away from the weld or seismic-impacted area.**
- **No significant stress from internal gas pressure; insignificant neutron irradiation.**
- **Environmental variations (e.g., seasonal temperature).**

Propagation of Single SCC Crack: Selective Conditions for SCC

The decreased probability of SCC propagation in a canister is furthered by:

- **Selective range of environmental and materials conditions for SCC**
- **More susceptibility in weld area (including heat affected zone [HAZ]) and deformed area under seismic conditions**
- **Limited number of pits or flaws**
- **Few through-wall cracks (Rudland, et al., 2009)**
- **Inspection, remediation, and mitigation (e.g., thermal annealing, applying compressive stress)**

Propagation of Single SCC Crack: Crack Behavior (Ford and Andresen, 2004)



- **"Engineering Initiation"** is defined as detectable Initiation.

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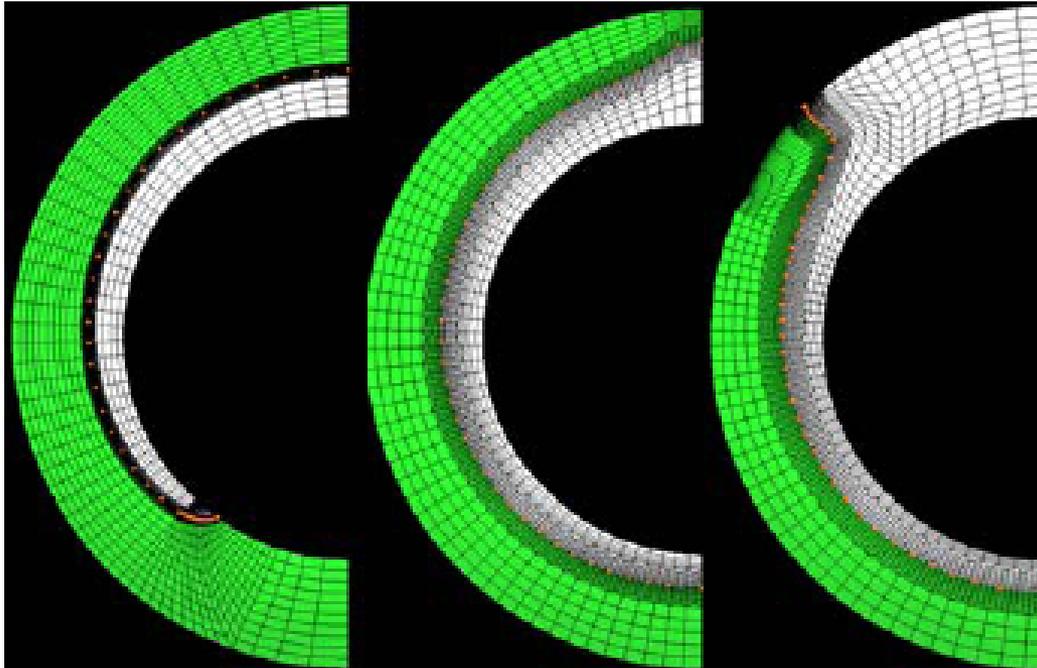
Propagation of Single SCC Crack: Crack Arrest



**Type 304 stainless steel piping contaminated with chlorides. Crack depth is about 1.2 mm (50 mils) (Stein, et al., 1986).
*(permission from ASM International)***

Propagation of Single SCC Crack: Through Wall

- **Simulation of natural crack fronts
(Rudland, et al., 2008)**



- **Result in leaking of reactor component**

Multiple SCC Cracks: Possible Maximum Opening Area of Multiple Cracks – Sandia National Laboratories (SNL) Model Assumptions

- **The SNL seismic model* conservatively assumes all possible surface cracks to penetrate through the wall thickness.**
- **The environment for SCC is present:
stress-based model**

* SNL, “Stress Corrosion Cracking of waste Package Outer Barrier and Drip Shield Materials,” ANL-EBS-MD-000005 REV 04 ERD2, 2007.

Multiple SCC Cracks: Possible Maximum Opening Area of Multiple Cracks - SNL Model Assumptions (continued)

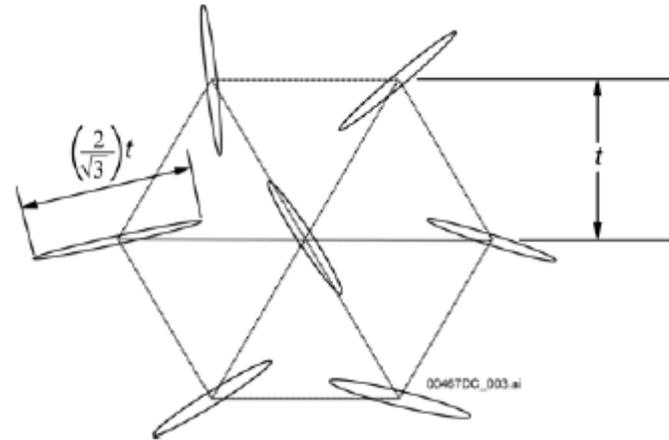
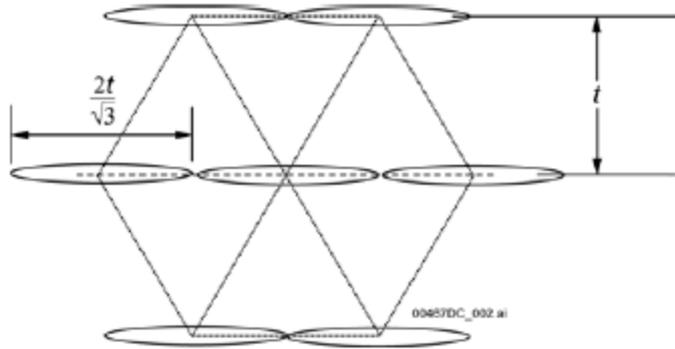
- **The distance between two neighboring through-wall cracks would need to be greater than the wall thickness for the stress (and resultant stress intensity) to be sufficient to drive a flaw through-wall. This conclusion is based on stress field interactions between closely spaced parallel cracks. The stress profile of a crack network is shown in the next slide.**
- **The aspect ratio of a crack (ratio of length to depth) has distribution for various possible crack geometries in a probabilistic system approach, and each value was sampled in the assessment of confinement or limited radionuclide release.**

Propagation of Single SCC Crack: Observation (continued)

- **Through-wall growth of neighboring cracks has not been observed (SNL, 2007).**

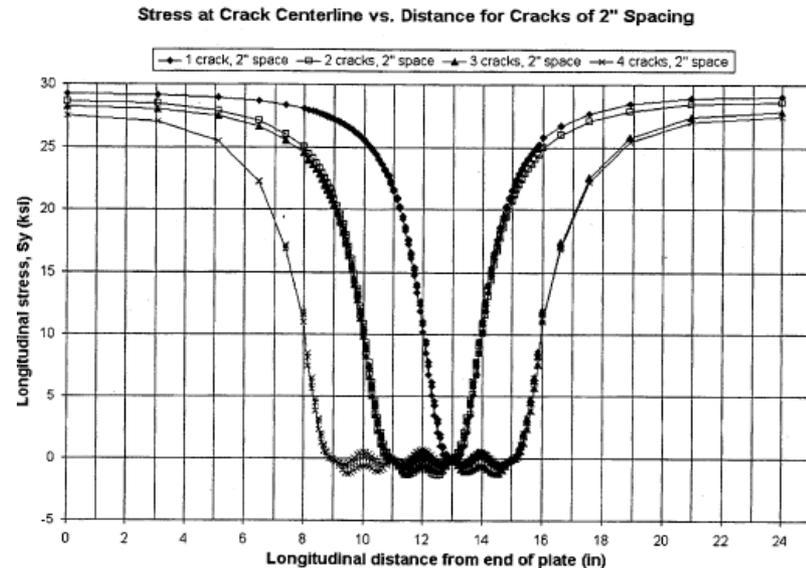
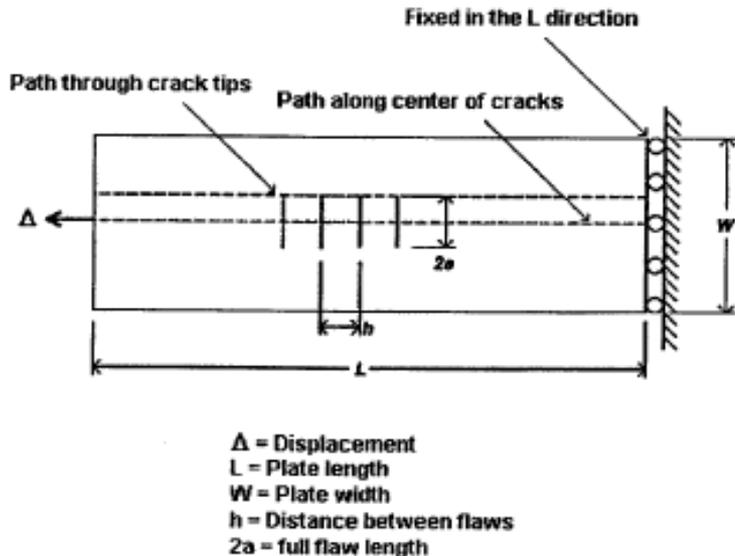
“Depending on the stress distribution, SCC may initiate and propagate through-wall. If several cracks were to initiate in the same area, coalesce, propagate through-wall while remaining straight (i.e., perpendicular to the surface), and maintain smooth crack faces, a sizable section of material could fall out. The occurrence of all of these events in conjunction is improbable. Only tight and relatively separate through-wall cracks are expected.”
(note: events such as coalescence of multiple cracks are important for the assessment of reactor structural integrity)

SNL Model: Crack Network from SCC



- **Maximum number of cracks**
- **“t” is radial thickness; greater than pit or flaw size**

SNL Model: Stress Analysis with Crack Network



- Dimensions of the plate and setup for the analysis (left)
- Longitudinal stress distribution along center with 2 inch spacing between cracks (right) (Structural Integrity Associates, 2002), 1 in = 2.54 cm; 1 ksi = 6.9 MPa

SNL Model: Mathematical Description

- **At a crack length, $a(t)$, the crack width, $w(t)$, will be**

$$w(t) = C \sigma a(t)/E$$

σ : applied stress (MPa)

E: Young's modulus (MPa)

C: geometric constant

t: time

under the conditions of plane stress and infinite size (conservative assumption) (SNL, 2007)

SNL Model: Mathematical Description (continued)

- **Each crack area is product of crack length and crack width. The number of cracks are proportional to sample area divided by $a(t)^2$. The maximum opening area of multiple cracks are the product of each crack area and the number of cracks (per unit deformed or weld/HAZ area).**

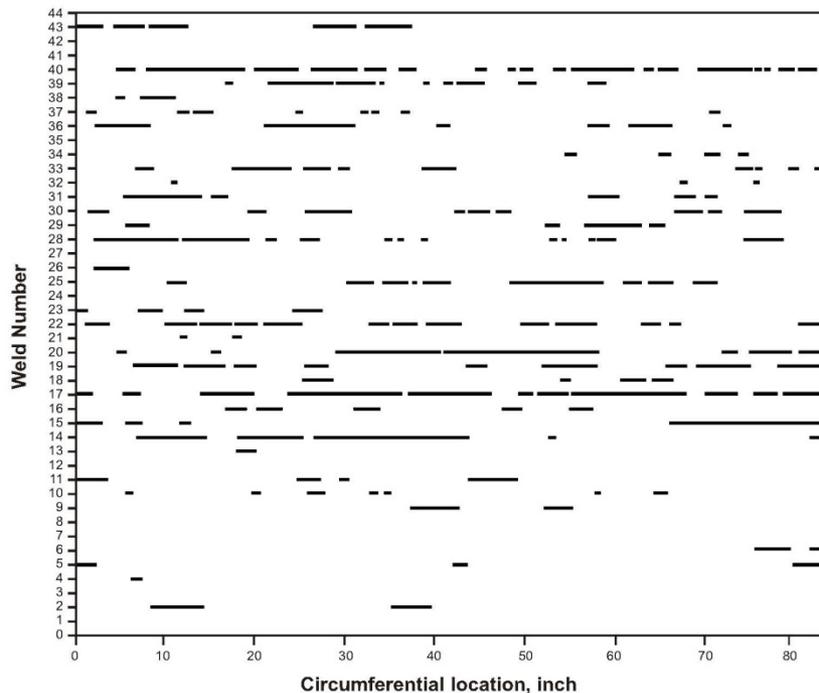
$$\delta \sim C \sigma/E$$

δ : crack areal density (m^2/m^2)

Application of SNL Model (Ahn, 2013)

- **Results of the numerical analysis of crack spacing were not sensitive to Young's modulus and Poisson's ratio (Structural Integrity Associates, 2002). This implies that the SNL model potentially can be applied to various metals.**
- **Example light-water reactor (LWR) case: the number and size of surface cracks were reported from welds at the Nine Mile Point Unit 1 main recirculation lines (Xu, et al., 2006).**

SNL Model Benchmark: Weld Cracks at Nine Mile Point Unit



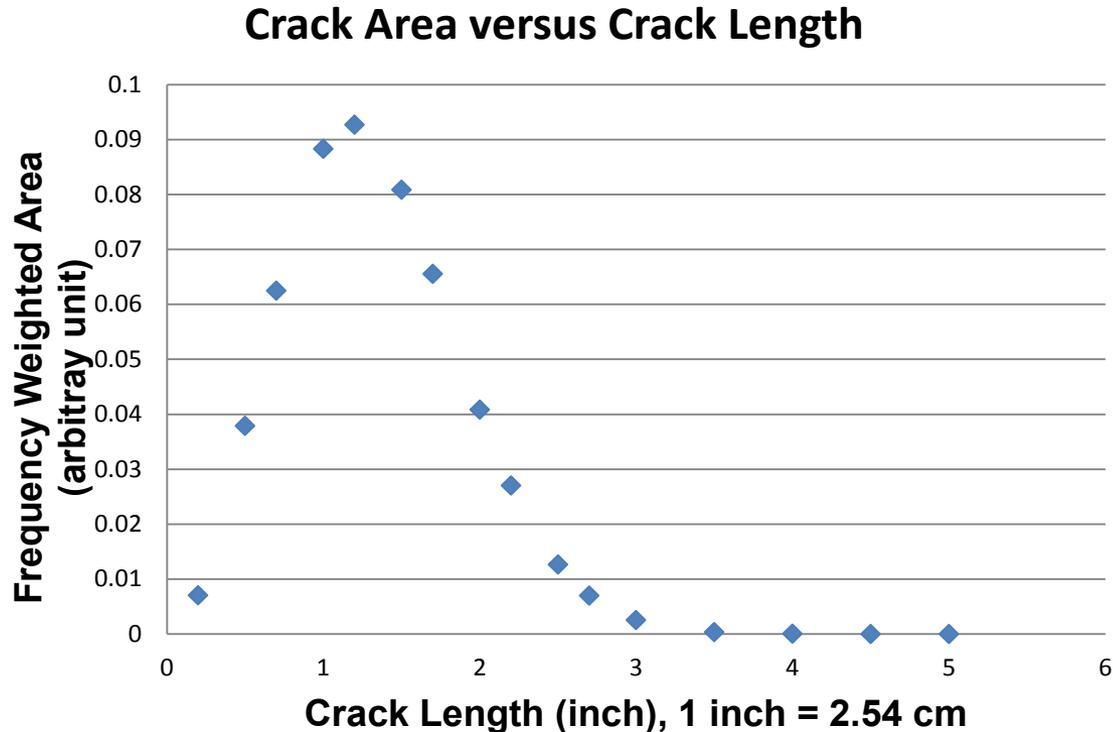
Nine Mile Point cracking data:

**Welder number vs.
circumferential location, inch
(2.54 cm) (Xu, et al., 2006,
*Permission by ASME,
PVP2006-ICPVT11-93966*)**

**The probability of through-wall
cracking is low.**

(Rudland, et al., 2009)

SNL Model Benchmark: Weld Cracks at Nine Mile Point Unit (continued)



- **Area is based on the width formula in slide 25.**
- **Plot is made by a curve-fitting of the data in slide 28.**

Application of SNL Model (continued)

- **The total number of surface cracks in welds and inter-crack distances are comparable to the SNL model.**
- **The dominant area of surface cracks appears to fall in a range of crack sizes, as suggested by the SNL model.**
- **Only a small fraction of surface cracks were through the wall thickness, with a low probability (Rudland, et al., 2009).**
- **The aspect ratio of a crack is considered to increase with large surface cracks from 10 to 150 (Xu, et al., 2006).**

Application of SNL Model (continued)

- **The SNL model appears to be conservative with the stress-based model. The model also appears to be consistent with the above LWR example observation with respect to most probable dominant crack size and potential aspect ratio present.**
- **This approach potentially provides an estimate for the possible maximum opening area of cracks for various metals. An example is shown in the next slide.**

Application of SNL Model (continued)

- **For stainless steel, the mean value of maximum crack opening area per unit weld/HAZ or deformed area is approximately 1.2×10^{-3} (fraction, cm^2/cm^2) for 170-310 MPa (24.6-44.9 ksi) of applied stress, $(193-207) \times 10^3$ MPa ($[28.0-30.1] \times 10^3$ ksi) of Young's modulus (Gwo, et al., 2011).**
- **The weld/HAZ area fraction is about $10^{-2} - 10^{-1}$ (Ahn, et al., 2012).**
- **The fraction of the opening area is small.**

Summary

- **The canister serves to confine radionuclides. If a canister degrades, limited radionuclides may be released through the opening of pits, crevice, or SCC cracks.**
- **The pit area can be limited by latent repassivation and cathode capacity.**
- **Examples were presented for pit repassivation inside or outside crevice for nickel-based alloy, and in a pencil electrode for stainless steel.**

Summary (continued)

- **Active dissolution of stainless steel occurred under crevice corrosion in 5% NaCl fog at ambient temperature. Long-term effects continue to be studied.**
- **Pits in stainless steel can serve as a precursory step for SCC in a chloride-bearing environment.**
- **The propagation rate of a single SCC may decrease during propagation. Also, susceptible conditions of environments and materials are selective. Uncertainties may be addressed by inspection and mitigation strategies.**

Summary (continued)

- **Possible maximum opening area of multiple cracks is estimated based on the SNL model for the seismic case in disposal. Example data in LWR welds suggest that the SNL model appears to be conservative and can be used to assess confinement and limited radionuclide release.**
- **The SNL model can be potentially used for various metals. For stainless steel, the fraction of possible maximum crack opening area appears to be limited with stress-based models.**