# Waste Package: Corrosion Testing and Model Development

Presentation to: Nuclear Waste Technical Review Board (NWTRB)

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## Introduction

- Long-term containment (10,000 years) requires materials with exceptional corrosion resistance
  - Very small penetration rates must be measured
  - Measurement error must be minimized to the extent possible
- Site Recommendation (SR) & License Application (LA) require credible predictive models based on sound understanding
- Such models have been developed for relevant degradation modes
  - General & Localized Corrosion
  - Stress Corrosion Cracking
  - Juvenile Failure
  - Phase Stability

#### Introduction (continued)

#### • Preliminary conclusions

- No significant localized corrosion expected
- Life not limited by general corrosion
- Phase stability appears to be acceptable
- Focus on SCC at final closure weld

#### **Abstracted Model for Waste Package Degradation**





## Determination of Crevice pH for Waste Package Materials



# Type 1: Alloy 22 in SSW at 120° C (DEA033)



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# Atomic Force Microscopy of Alloy 22 surface exposed to vapor phase SAW for 1 year at 90°C

5 um



of 0.05 to 1 micron per year.

Long Term Corrosion Test Facility at LLNL

## SCC Model Three Primary Contributions to Stress



## SCC Model - Critical Stress & Flaw Size

Criteria for stress corrosion cracking

 $K \geq K_{ISCC}$ 

• Stress intensity factor for ideal crack

 $K = bs (pa)^{1/2}$ 

Stress intensity factor for crack at base
 of elliptical flaw (or pit)

$$K = abs [p(a_{flaw} + da)]^{1/2}$$

$$K = b K_{t} s (d a)^{1/2}$$

$$K_{t} = 1 + \frac{2a}{c}$$



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## Measurement of Residual Weld Stress in a Prototypical Closure Weld of Waste Package



# Double-Pass Laser Peening (4340 Steel)



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# **Mechanisms for WP Juvenile Failure**

#### • Types of generic flaws applicable to waste packages

- Weld or base metal flaws
- Out-of-spec material in weld or base metal
- Improper heat treatment
- Surface contamination
- Handling damage
- Administrative error leading to unanticipated environment

#### Generic flaws not considered applicable to waste packages include

- Improper weld flux
- Poor weld joint design
- Missing welds
- Mislocated welds



## Review of Early Failures in Various Containers

#### • Types of containers include:

- Boilers and Pressure Vessels
- Nuclear Fuel Rods
- Radioactive Cesium Capsules
- Dry Storage Casks for SNF
- Food Storage Cans
- Manufacturing defect related failure rates in the range of 10<sup>-4</sup> to 10<sup>-6</sup> per container
- Review identified eleven generic types of flaws that can affect welded metallic containers

# Alloy 22 Weld Flaw Distributions

- Preliminary flaw distributions developed based on data from recent NRC sponsored modeling of nuclear piping welds
  - 20 mm and 25 mm thick, Stainless Steel, TIG welds
  - Includes reliability of UT, PT, and RT inspections as appropriate



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## Precipitated μ Phase Observed in Welded Alloy 22 Aged for 40,000 hr at 427°C







Work is now underway at LLNL to better understand formation of intermetallic phases that may form in Alloy 22 welds.

# Time Temperature Transformation (TTT) Diagram for Alloy C-22



## Inputs to Waste Package Degradation Process Model Report

- Environment on Drip Shield & Waste Package Surface (July 99)
- Juvenile Failures (July 99)
- Phase Stability & Aging (July 99)
- Mechanical Failure Due to Rockfall (August 99)
- General Corrosion of Waste Package (July 99)
- Localized Corrosion of Waste Package (July 99)
- General Corrosion of Drip Shield (August 99)
- Localized Corrosion of Drip Shield (August 99)
- Stress Corrosion Cracking of Waste Package (September 99)
- Stress Corrosion Cracking of Drip Shield (September 99)
- Hydrogen Induced Cracking of Drip Shield (September 99)
- Degradation of Stainless Steel Structural Material (September 99)
- Abstractions for WAPDEG (August 99 to October 99)

#### Updated design

- Stainless steel & titanium were not used in TSPA-VA design
- Tests on these materials have just started
- Limited availability of qualified data
- Increased gamma radiolysis

#### Fabrication processes

- Shrink fitting with outer barrier of Alloy 22
  - » Precipitation of undesirable phases during heating
  - » Development of excessive tensile stress during cooling

#### - Unannealed closure welds

- » Initiation of SCC at weld defects
- » Possible mitigation of residual weld stress with laser peening

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#### • Competing models for Stress Corrosion Cracking (SCC)

#### - Initiation based upon threshold stress intensity factor

- » Method employed by Yucca Mountain Project
- » Double Cantilever Beam Method (Ajit Roy)
- » Data have been obtained for Alloy 22 in NaCl solutions

#### - Finite propagation rate based upon film-rupture model

- » Method employed by General Electric Corporation
- » Reverse DC Method (Peter Andresen)
- » No data have been generated for Alloy 22

(continued)

### Microbial Influenced Corrosion (MIC)

#### Microbes may pose unique threats

- » Sulfate reducing bacteria could produce sulfide, a species known to promote SCC of Alloy 22
- » Iron oxidizing bacteria could convert Fe(II) to Fe(III), thereby pushing the corrosion potential closer to threshold for localized attack
- Quantitative models have not yet been developed

(continued)

#### • Effects of increased radiation field on corrosion processes

- Gamma radiolysis can produce hydrogen peroxide
- Hydrogen peroxide can shift the corrosion potential in anodic direction closer to thresholds for localized attack
- A strategy has been formulated for addressing any enhanced radiolysis effects in the EDA II design
  - Re-examination stainless steel corrosion data from gamma pit that was produced by Yucca Mountain Project in the mid 1980's
  - Discussion & collaboration with investigators at General Electric Corporation
  - In the absence of gamma radiation, investigate the effect of hydrogen peroxide concentration on the corrosion potential (and other electrochemical responses) of WP materials
  - Repeat gamma pit studies with Alloy 22 and Ti Gr 7, thereby augmenting early Project data for stainless steels

# Summary

- Long-term containment (10,000 years) requires materials with exceptional corrosion resistance
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#### • Preliminary conclusions

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# **Scientific & Technical Contributions**

- Definition of Interfacial Waste Package Environment
  - -Greg Gdowski & Francis Wang
- Long Term Corrosion Testing
  - —Dan McCright, John Estill, Ken King, Steve Gordon & Larry Logotetta
- Electrochemical Studies & Surface Physics
  - —Joe Farmer, John Estill, Ken King, Steve Gordon & Larry Logotetta
  - -Peter Bedrossian & David Fix
- Phase Stability
  - —Tammy Summers, Patrice Turchi & Larry Kaufman
- Stress Corrosion Cracking Studies
  - —Ajit Roy, John Estill, Maura Spragge, Dennis Fleming & Beverly Lum
- Microbial Influenced Corrosion

—JoAnn Horn, Denny Jones, Tiangan Lian,

Welding Processes, Residual Stress Analysis & Laser Peening

—Don Stevens, Lloyd Hackel, Fritz Harris (MIC) & Al Lingenfelter

Waste Package Modeling

-Joe Farmer, Stephen Lu, Bob Riddle & Jia-Song Huang

## **Additional Supporting Data**

#### Cyclic Polarization

- Pt baseline
- Type 1: Alloy 22 in SAW
- Type 2: Alloy 22 in SCW
- Type 3: 316L in SSW
- Type 4: Ti Gr 7 in SSW

#### Crevice pH & Current

- Stainless Steel 316L: 4M NaCl, 200 mV, 23 Centigrade pH
- Alloy 22 in 4M NaCl at 23 Centigrade pH
- Alloy 22 in 4M NaCl at 23 Centigrade Current
- Alloy 22 in SCW at 23 Centigrade pH
- Determination of Crevice pH for WP Materials

# **Additional Supporting Data**

#### Long Term Corrosion Testing Facility Data

- Dissolved Oxygen in LTCTF
- General Corrosion of Alloy 22: Weight Loss Samples
- General Corrosion of Ti Gr 16: Crevice Corrosion Samples
- Analysis of Error in Corrosion Rate Measurements
- AFM of Alloy 22 Samples from LTCTF
- AFM of Patterned Samples

#### Weld & Stress Corrosion Cracking

- Residual Stress in Prototypical Welds of Alloy 22
- SSRT of Alloy 22
- SSRT of Ti Gr 12 (Ti Gr 7 analog)

# **Additional Supporting Data**

(continued)

### Phase Stability

- Complete Coverage of Alloy 22 GBs at High Temperature
- 88,000 Years Required
- Juvenile Failure Model

# Baseline: Pt in SCW at 90° C (PT001)



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# Type 1: Alloy 22 in SAW at 90° C (DEA002)



Current (A)

# Type 2: Alloy in SCW at 90° C (DEA016)



Current (A)

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# Type 3: 316L in SSW at 100° C (PEA016)



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# Type 4: Ti Gr 7 in SSW at 120° C (NEA031s)



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# Stainless Steel 316L: 4 M NaCl, 200mV and 23° C



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# Alloy C-22 in 4 M NaCl at 23° C





# Alloy C-22 in 4 M NaCl at 23° C

#### Alloy C-22 in 4M NaCl at 23 Centigrade



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# Alloy C-22 in SCW at 23° C



# Determination of Crevice pH for Waste Package Materials



## Comparison of Dissolved Oxygen Measurements in LTCTF to Data for Synthetic Geothermal Brine



# General Corrosion of Alloy 22 Weight Loss Samples from LTCTF



# General Corrosion of Ti Gr 16 Crevice Samples from LTCTF



## Analysis of Error in Measurement of General Corrosion: Total Derivative

 $y = f(x_1, x_2, x_3, x_4 \cdots x_n)$ 

$$dy = \frac{\partial y}{\partial x_1} dx_1 + \frac{\partial y}{\partial x_2} dx_2 + \frac{\partial y}{\partial x_3} dx_3 + \frac{\partial y}{\partial x_4} dx_4 + \dots + \frac{\partial y}{\partial x_n} dx_n$$

$$dy = \sum_{j=1}^{n} \frac{\partial y}{\partial x_{j}} dx_{j}$$
$$\Delta y = \left| \frac{\partial y}{\partial x_{1}} \Delta x_{1} \right| + \left| \frac{\partial y}{\partial x_{2}} \Delta x_{2} \right| + \left| \frac{\partial y}{\partial x_{3}} \Delta x_{3} \right| + \left| \frac{\partial y}{\partial x_{4}} \Delta x_{4} \right| + \dots + \left| \frac{\partial y}{\partial x_{n}} \Delta x_{n} \right|$$
$$\Delta y = \sum_{j=1}^{n} \left| \frac{\partial y}{\partial x_{j}} \Delta x_{j} \right|$$

## Analysis of Error in Measurement of General Corrosion Rate: Application to Weight Loss Formula

$$\frac{dp}{dy} = \frac{w}{\mathbf{r} \times t} \frac{1}{[2(a \times b) + 2(b \times c) + 2(a \times c)]} \qquad y = \frac{dp}{dt}$$

$$dy = \frac{\partial y}{\partial w} dw + \frac{\partial y}{\partial \mathbf{r}} d\mathbf{r} + \frac{\partial y}{\partial t} dt + \frac{\partial y}{\partial a} da + \frac{\partial y}{\partial b} db + \frac{\partial y}{\partial c} dc$$

$$\Delta y = \left| \frac{\partial y}{\partial w} \Delta w \right| + \left| \frac{\partial y}{\partial \mathbf{r}} \Delta \mathbf{r} \right| + \left| \frac{\partial y}{\partial t} \Delta t \right| + \left| \frac{\partial y}{\partial a} \Delta a \right| + \left| \frac{\partial y}{\partial b} \Delta b \right| + \left| \frac{\partial y}{\partial c} \Delta c \right|$$

$$\frac{\partial y}{\partial w} = \frac{1}{\mathbf{r} \times t} \frac{1}{[2(a \times b) + 2(b \times c) + 2(a \times c)]} \qquad \frac{\partial y}{\partial a} = \frac{w}{\mathbf{r} \times t} \frac{[2b + 2c]}{[2(a \times b) + 2(b \times c) + 2(a \times c)]^2}$$

$$\frac{\partial y}{\partial \mathbf{r}} = \frac{w}{\mathbf{r}^2 \times t} \frac{1}{[2(a \times b) + 2(b \times c) + 2(a \times c)]} \qquad \frac{\partial y}{\partial b} = \frac{w}{\mathbf{r} \times t} \frac{[2a + 2c]}{[2(a \times b) + 2(b \times c) + 2(a \times c)]^2}$$

$$\frac{\partial y}{\partial t} = \frac{w}{\mathbf{r} \times t^2} \frac{1}{[2(a \times b) + 2(b \times c) + 2(a \times c)]} \qquad \frac{\partial y}{\partial c} = \frac{w}{\mathbf{r} \times t} \frac{[2a + 2c]}{[2(a \times b) + 2(b \times c) + 2(a \times c)]^2}$$

$$\frac{\partial y}{\partial c} = \frac{w}{\mathbf{r} \times t^2} \frac{[2a + 2b]}{[2(a \times b) + 2(b \times c) + 2(a \times c)]} \qquad \frac{\partial y}{\partial c} = \frac{w}{\mathbf{r} \times t} \frac{[2a + 2b]}{[2(a \times b) + 2(b \times c) + 2(a \times c)]^2}$$

$$\frac{\partial y}{\partial c} = \frac{w}{\mathbf{r} \times t} \frac{[2a + 2b]}{[2(a \times b) + 2(b \times c) + 2(a \times c)]} \qquad \frac{\partial y}{\partial c} = \frac{w}{\mathbf{r} \times t} \frac{[2a + 2b]}{[2(a \times b) + 2(b \times c) + 2(a \times c)]^2}$$

# Atomic Force Microscopy of Alloy 22 surface exposed to liquid phase SAW for 1 year at 90°C



Long Term Corrosion Test Facility at LLNL

# Reducing Uncertainty with Atomic Force Microscopy of Patterned Coupons

 AFM offers sub-nanometer vertical resolution for oxide thicknesses, pit depths, general corrosion and swelling, while a photoresist protects base metal. This is a novel approach.



## **Slow Strain Rate Testing of Alloy 22**



### **Slow Strain Rate Testing of Ti Gr 12**







#### **Mill Annealed**

#### **Aged Material**

# Time at Which Grain Boundaries Become Covered with Carbides, P and/or µ Phase



# Definition of WP Juvenile Failure

- Failure of a waste package, due to manufacturing or handling induced defects, at a time earlier than would be predicted by mechanistic degradation models for an "ideal" package
- Failure rates for all components exhibit a "bathtub" curve behavior over time





## Flaws in Shell and Lid Welds

- Over 20 years of research into the density and size distribution of weld flaws and the mechanisms which produce them
  - Formation mechanisms include: Lack of fusion, porosity, slag inclusions, centerline cracking (generally hydrogen induced), heat-affected zone cracking
  - NRC has sponsored weld flaw density and size distribution research by Rolls Royce and PNL for a variety of materials, weld thickness, weld methods, and post-weld inspections
  - Research provides input to probabilistic fracture mechanics analyses and risk informed in-service inspection proposals

## Flaws in Shell and Lid Welds

(continued)

- RR-PRODIGAL code developed to model flaw occurrence in welds
  - Surface breaking weld flaws expected to be most important contributor to WP performance
  - Represent pre-existing pits or crevices, and potential SCC sites
  - Unlike pressure vessels, no growth of embedded flaws expected due to lack of cyclic stresses

## **Juvenile Failure Model**

- Work currently in progress to develop initial probability and consequence estimates for remaining flaw types
- Future work includes:
  - Modeling of WP welds using RR-PRODIGAL code
  - Refined estimates of flaw occurrence probability
  - Refined estimates of flaw effects on WP performance