EFFECT OF CLADDING ON RELEASE OF RADIONUCLIDES

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Appendix 7 Meeting: Container Life and Source Term
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SPENT NUCLEAR FUEL (SNF) CLADDING

Presentation Outline

- NRC Concerns in Container Life and Source Term (CLST) Issue Resolution Status Report (IRSR), Revision 1

- Presentation of Preliminary Results of Zircaloy-4 Cladding Material Corrosion Experiments (NRC/CNWRA)

EFFECT OF CLADDING ON RELEASE OF RADIONUCLIDES

- SNF Cladding Can Act as a Barrier to the Release of Radionuclides
  - ➤ DOE Considers Cladding as an Additional Metallic Barrier in TSPA-VA

- Cladding is Subject to Several Degradation Mechanisms That can Impair This Beneficial Action
  - ➤ Localized Corrosion, Stress Corrosion Cracking, Creep, Delayed Hydride Cracking, Hydrogen Embrittlement, Mechanical Failure from Rock Fall, Fuel and Cladding Oxidation

Potential Degradation Mechanisms of Zircaloy Cladding

- General Corrosion Rate of Zircaloy is Extremely Low Due to Protective ZrO_2 Passive Film

- Localized Corrosion (Pitting)
  - Neutral and Acidic Chloride Solutions
  - Above a Critical Potential, E_{rp} (Repassivation Potential)
  - E_{rp} can be Reached in the Presence of Oxidizing Species

- Stress Corrosion Cracking (SCC)
  - Same Environments and Potentials That Promote Pitting
  - Local Tensile Stresses Could Enhance Potential for SCC

DOE has Considered the Possibility of Localized Corrosion and SCC of Zircaloy in the Presence of Oxidizing Chloride Solutions Unimportant

Cladding Degradation Mechanisms (*Continued*)

- Diffusion-Controlled Cavity Growth (DCCG) Creep Failure Mechanism
  - Concern with Model Extrapolation to Lower Stresses and Temperatures
  - Cladding Temperature May Reach 500°C During Storage and Transportation
  - Corresponding Increase in Pressure

- Effect of Alternate EBS Design
  - Failure by Creep Rupture is Unlikely With No Backfill

- Delayed Hydride Cracking (DHC)
  - May not be Important Because the Operating Stress Intensity is Below the Crack Growth Threshold Stress Intensity, K_{IH}
  - May Need to be Reevaluated Considering Higher Stresses

Cladding Degradation Mechanisms (*Continued*)

- Hydride Embrittlement
  - ➤ Hydrogen Pick-up in the Repository Expected to be Small
  - ➤ Hydride Reorientation of Existing Circumferential Hydrides
    - Temperature
    - Stress

Hydrogen Embrittlement May Be an Important Failure Mechanism Depending on the Cladding Temperature and Resulting Hydride Reorientation

Cladding Degradation Mechanisms (*Continued*)

- Fuel and Cladding Oxidation
  - ➤ Slow Rate of Cladding Oxidation Below T = 350°C
  - ➤ Perforated Fuel Rods
    - Sluggish Fuel Oxidation Kinetics Below T = 250°C
    - Above 250°C Fuel Oxidation Volume Expansion May "Unzip" Cladding (UO_2 -> U_3O_8)
    - Below 250°C, Volume Expansion and Failure May Occur Due to Secondary Mineral Formation In Aqueous Environment

DOE is Addressing this Problem Through Experiments Currently Being Conducted at Argonne National Laboratory (ANL)

Mechanical Failure of Zircaloy Cladding - Rock Fall

(Kwai Chan at SwRI)

Objective: to develop a fracture mechanics-based model for treating the potential failure of degraded fuel cladding tubes caused by rock fall impacting on waste packages in the repository

Approach:

- Assume cladding rods to contain cracks or flaws of known size, geometry, and distribution
- Assume cladding properties to be degraded by hydride
- Treat impacting rocks in terms of a distributed load
- Perform an elastic bending analysis and apply elastic fracture mechanics to a single rod

Rock Fall - Mechanical Failure of Cladding (*Continued*)

Approach (*Continued*)

- Treat cladding failure based on a critical stress intensity factor (K_{IC}) fracture criterion
- Extend single-rod solution to multiple rods
- Determine the critical size and weight of rock fall for cladding failure as a function of impact parameters (impact angle, drop height, and impact velocity), crack parameters (size, shape, and population), and material parameters (UTS, K_{IC}, hydrogen content).

Rock Fall - Mechanical Failure of Cladding (*Continued*)

Current Status:

- Performed an elastic bending analysis to obtain cladding stress due to rock fall on a single rod
- Obtained K solutions for appropriate crack geometries
- Formulated a fracture mechanics approach for predicting critical load and rock size at fuel rod fracture
- Extended single-rod solution to multiple rods
- Developed a computer algorithm to apply the fracture model to treat failure of multiple rods at different levels in a fuel assembly
- Preliminary calculations of critical rock size are being generated and the results are being evaluated.

Mechanical Failure of Cladding - Rock Fall (*Continued*)

DOE Should:

- Develop a Rock Fall Model Based on Fracture Mechanics
- Incorporate Flaws or Defects in SNF Cladding
- Extend Model To Assess the Effect of Rock Fall From Drifts and Seismic Events

NRC's Main Concern With DOE's Approach To SNF Cladding Degradation is the Temperature Estimates For SNF Cladding

- Temperature has an Important Effect on Various Failure Processes:
  - ➢ Creep and
  - ➢ Hydride Reorientation and Embrittlement

- DOE Should
  - ➢ Complete an Evaluation of the Range of Cladding Temperatures Expected Under Repository Conditions
  - ➢ Determine Probability of Cladding Failure Based on Temperature and Associated Uncertainty
  - ➢ Determine Consequences of Cladding Failure in Terms of Radionuclide Release

Zircaloy-4 Cladding Material Corrosion Experiments (NRC/CNWRA)

Objectives:

- Determine the repassivation and corrosion potentials of Zr-4 SNF cladding under potential near-field environmental conditions of the proposed Yucca Mountain high-level waste (HLW) repository.
- Study the effects of pH, temperature and chloride concentration on the localized corrosion of Zr-4.
- Study the variations in the localized corrosion of Zr-4 due to the presence of oxidizing species, e.g., radiolysis and corrosion products
- Use the results of these experiments to bound current SNF cladding performance assessment modeling.

Zr-4 Cladding Corrosion Experiments (*Continued*)

Results:

- Effect of Chloride Concentration in Simulated J-13 Well Water
  - Follows Well Known Equation

    $$E_{rp} = E^{\circ}_{rp} - B \log [Cl^-]$$

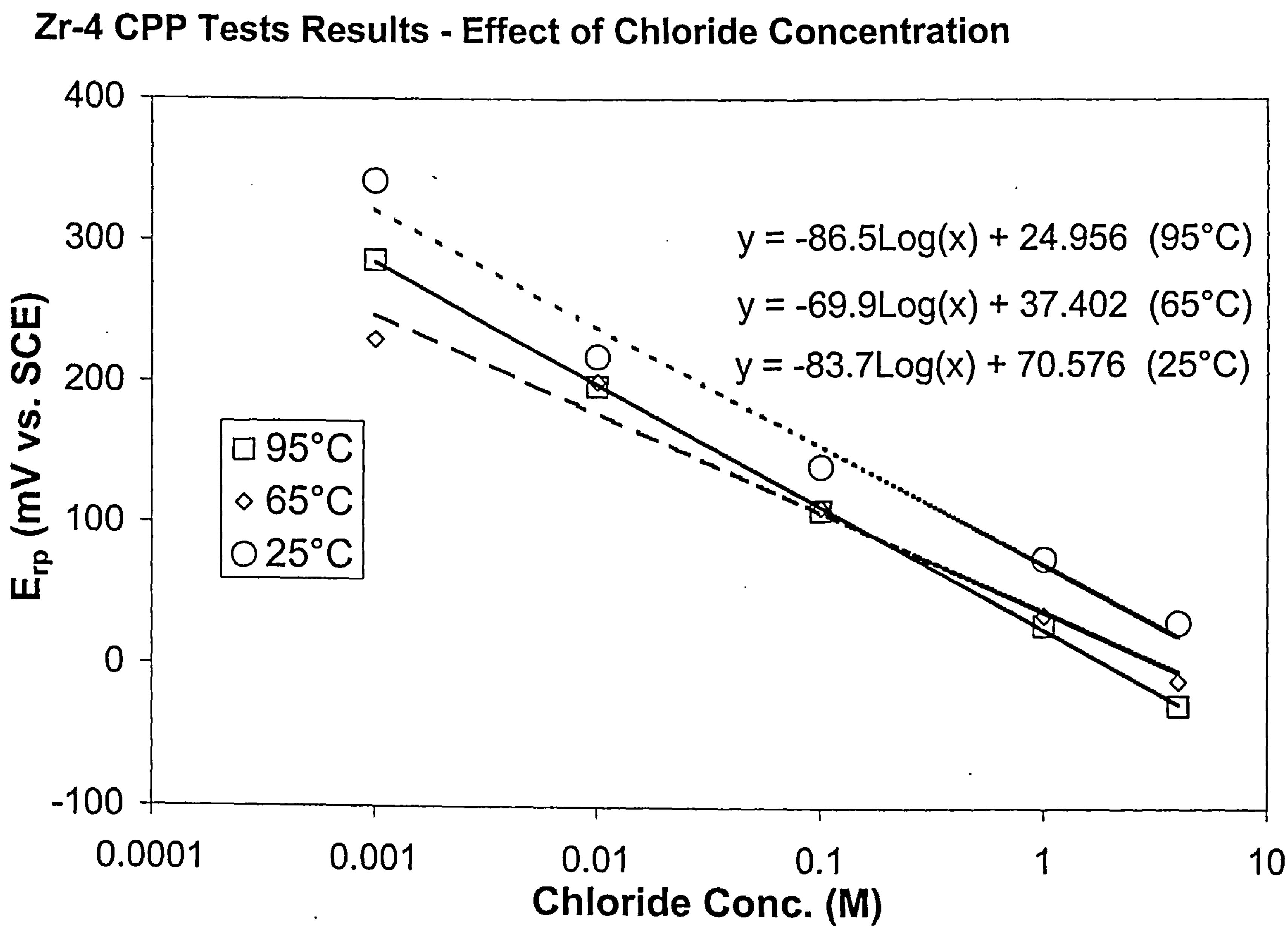
  - Pitting Attack, but No Crevice Attack
  - Experiments in Non-J-13 Chloride Solutions Indicated Minor Inhibitor Effect of Anions in J-13 Water

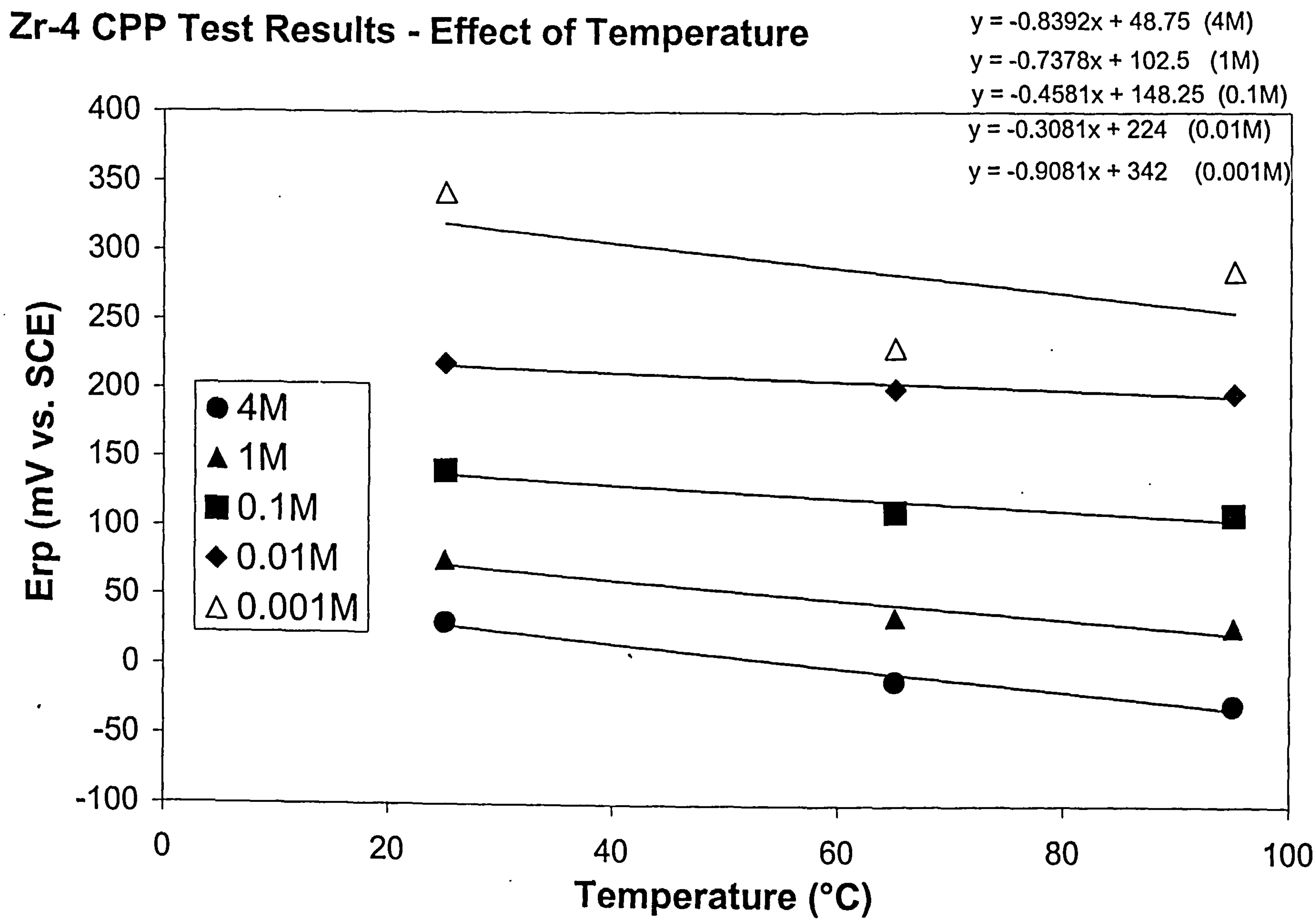
- Effect of pH
  - No Effect at High pH (pH = ~10 to 11)
  - Minor Effect at Low pH (pH = ~2)
  - Additional Data Points Needed

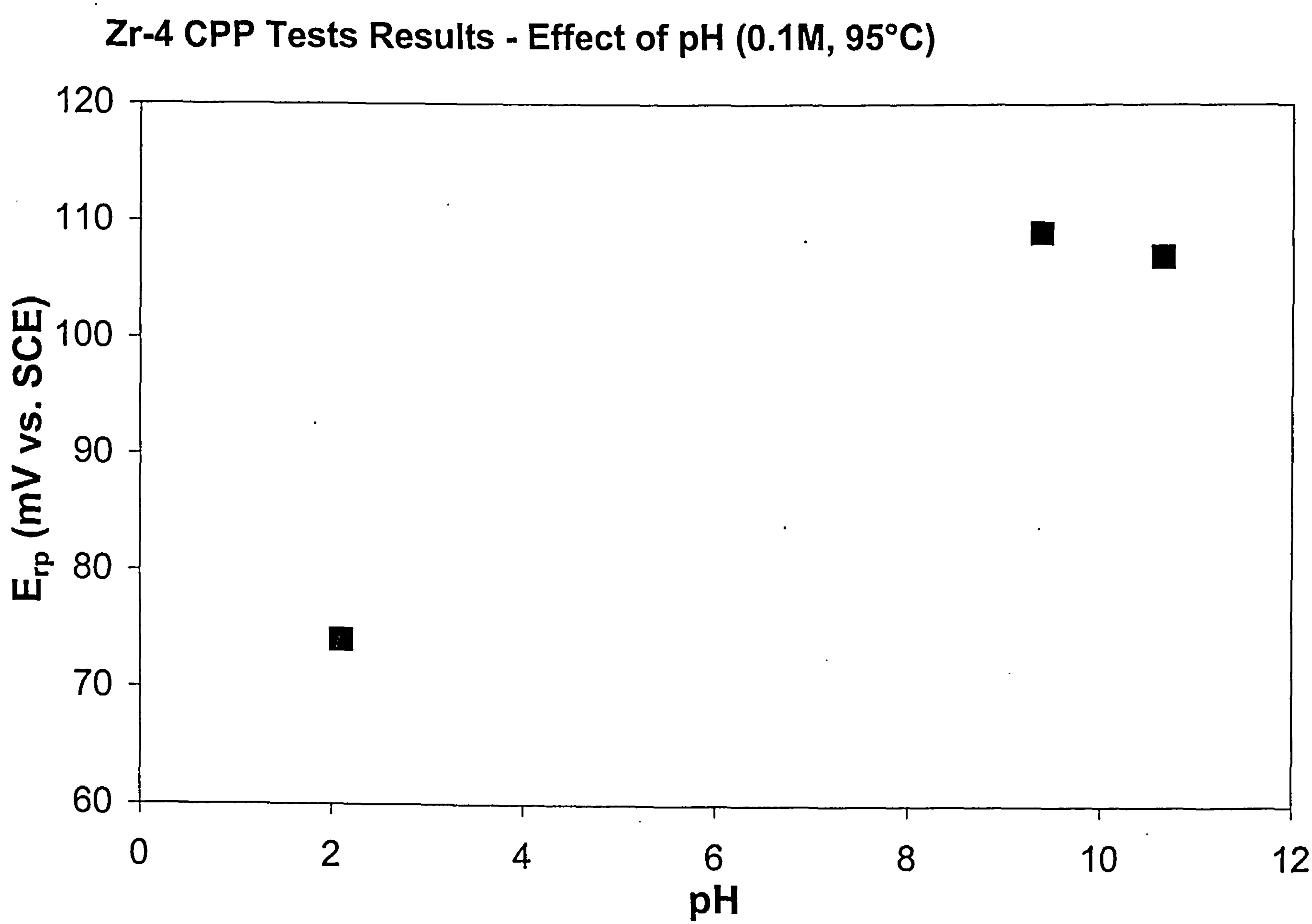
- Minor Effect of Temperature in the Range of 25 to 95°C

Zr-4 Cladding Corrosion Experiments (*Continued*)

(Graphs)


Zr-4 CPP Tests Results - Effect of Chloride Concentration
400
300
200
100
0
-100
E_rp (mV vs. SCE)
y = -86.5Log(x) + 24.956 (95°C)
y = -69.9Log(x) + 37.402 (65°C)
y = -83.7Log(x) + 70.576 (25°C)
95°C
65°C
25°C
0.0001
0.001
0.01
0.1
1
10
Chloride Conc. (M)


Zr-4 CPP Test Results - Effect of Temperature
y = -0.8392x + 48.75 (4M)
y = -0.7378x + 102.5 (1M)
y = -0.4581x + 148.25 (0.1M)
y = -0.3081x + 224 (0.01M)
y = -0.9081x + 342 (0.001M)
Erp (mV vs. SCE)
400
350
300
250
200
150
100
50
0
-50
-100
0
20
40
60
80
100
Temperature (°C)
4M
1M
0.1M
0.01M
0.001M



Zr-4 CPP Tests Results - Effect of pH (0.1M, 95°C)
120
110
100
90
80
70
60
0
2
4
6
8
10
12
pH
Erp (mV vs. SCE)

Zr-4 Cladding Corrosion Experiments (*Continued*)

Results (*Continued*):

- Open Circuit Potential (E_{corr}) Under Air-Saturated Conditions + Peroxide + Fe^{3+} Exceeded E_{rp}

This Indicates Localized Corrosion of Zr-4 Cladding May Occur Under The Range of Possible Environments within the Waste Package.

- Potentiostatic Hold Tests ~ 25 mV above E_{rp} Resulted in Extensive Attack

- Pre-oxidized Zr-4 by Heat Treating in Air at 200°C Before Testing in Chloride Solutions Resulted in Narrow and Deeper Pitting Attack.

SUMMARY

- SNF Cladding Can Act as a Barrier to the Release of Radionuclides
- Potential Degradation Processes of Zircaloy Cladding
  - Localized Corrosion, Creep, SCC, Delayed Hydride Cracking, Hydrogen Embrittlement, Mechanical Failure from Rock Fall, Fuel and Cladding Oxidation
- DOE Should Complete an Evaluation of the Range of Cladding Temperatures Expected Under Repository Conditions.
- DOE Should Develop a Rock Fall Model Based on Fracture Mechanics
- Localized Corrosion of Zr-4 Cladding Can Occur Under The Range of Oxidizing Chloride Environments Possible within the Waste Package.