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**CHEMICAL DURABILITY AND
DEGRADATION MECHANISMS OF HT9
BASED ALLOY WASTE FORMS WITH
VARIABLE ZR CONTENT**

Fuel Cycle Research & Development

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SUMMARY

Corrosion studies were undertaken on alloy waste forms that can result from advanced electrometallurgical processing techniques to better classify their durability and degradation mechanisms. The waste forms were based on the RAW3-(URe) composition, consisting primarily of HT9 steel and other elemental additions to simulate nuclear fuel reprocessing byproducts. The solution conditions of the corrosion studies were taken from an electrochemical testing protocol, and meant to simulate conditions in a repository. The alloys durability was examined in alkaline and acidic brines.

Surrogate alloy waste forms were made into electrodes and examined electrochemically in alkaline and acidic brines. A testing approach was developed in collaboration with Argonne National Laboratory, and consisted of 4 main steps, 1) measure the bare surface corrosion behavior in reference solutions at open circuit over a wide imposed potential range, 2) perform several differing potentiostatic holds to measure the time-evolution of anodic current, evolution of surface properties, and radionuclide concentrations in solution, 3) relate steady-state current to radionuclide (or surrogate) release rates through periodic sampling of test electrolyte, and 4) identify the corroding phase/phases using SEM/EDS. Step 2 could last up to 30 days with mostly fixed electrode potential and periodic potential interruptions for a variety of electrochemical analyses. This combined electrochemical and extended immersion testing is referred to as hybrid testing.

Three alloys were tested that were based on the RAW3-(URe). The three alloys differed in their ratio of U to Zr, with the U concentration kept mostly constant, but the Zr concentration varied. The alloys are referred to by their ratio of U:Zr, with the three ratios being 3U:1Zr, 1U:1Zr, and 1U:2Zr.

The hybrid testing and electrochemical characterization, solution sampling, and SEM/EDS indicated that the 1U:2Zr was the most stable in the test solutions. The 3U:1Zr alloy was the least stable.

U concentration levels in the solutions usually increased or stayed flat during test duration, but sometimes dipped. Re concentrations tended to increase throughout the test durations. Re concentrations in the solutions are of interest because Re is the surrogate for Tc. Tc is expected to result in the largest source of dose to the public from a repository. Re comes primarily from the steel phase in the waste form. SEM/EDS found evidence of the high-Z phases responsible for sequestration of the U, but not of localized corrosion of the steel phases, indicating general uniform corrosion was occurring on the steel phases.

Due to localized corrosion seen in the high-Z phases, it is recommended to examine alloys with increased Ni content because Ni should segregate preferentially to the high-Z phases, decreasing their electrochemical activity and reducing the localized corrosion attack of this phase.

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ACRONYMS

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| FCRD | DOE Fuel Cycle Research and Development Program |
| SEM | Scanning Electron Microscopy |
| EDS | Energy Dispersive X-ray Emission Spectroscopy |
| BSE | Backscattered Electron |
| SS | Stainless Steel (Unspecified) |
| UDS | Undissolved Solids |
| YSZ | Y ₂ O ₃ Stabilized ZrO ₂ |
| Zr | Zirconium |
| U | Uranium |
| DU | Depleted Uranium |
| AWF | Alloy Waste Form |
| TRUEX | Transuranic Extraction Process |
| NIST | National Institute of Standards |
| 316SS | Type 316 Stainless Steel |
| HT9 | A type of ferritic steel |
| RAW | Reference Alloy Waste form |
| EBR-II | Experimental Breeder Reactor II |
| EMT | Electrometallurgical Treatment |
| M | Molar |
| DI | Deionized |
| FY12 | Fiscal Year 2012 |
| °C | Degree Celsius |
| scfh | Standard cubic foot per hour |
| NMFP | noble metal fission product |
| x | magnification |
| FCCI | fuel cladding chemical interaction |
| 1U:2Zr | Alloy surrogate waste form made in FY12 with 1:2 U/Zr concentration ratio |
| 1U:1Zr | Alloy surrogate waste form made in FY12 with 1:1 U/Zr concentration ratio |
| 3U:1Zr | Alloy surrogate waste form made in FY12 with 3:1 U/Zr concentration ratio |

CHEMICAL DURABILITY AND DEGRADATION MECHANISMS OF HT9 BASED ALLOY WASTE FORMS WITH VARIABLE ZR CONTENT

1. INTRODUCTION

1.1 Background

Iron-based alloy waste forms are being investigated to immobilize metallic radioactive species expected from electrometallurgical treatment methods, such as the undissolved solids (UDS) remaining in the anode basket of the electrolyzer after electrometallurgical treatment (EMT). For conventional sodium cooled fast reactors with metallic fuel and operating in a closed nuclear fuel cycle, these species likely consist primarily of the alloy fuel cladding, and metals not dissolved in the electrolyzer, primarily Zr, undissolved U diffused into the Zr or cladding, and other trace metals. Similar species are expected from aqueous recycling schemes for used fuel reprocessing, and the iron-based waste form is adaptable to those waste streams. Prototype waste forms are being formulated and tested to evaluate the capacity to accommodate the radionuclides present in waste streams in durable host phases. Previous work has shown U and transuranics to be immobilized in Zr-Fe intermetallic phases, and formulation of the waste form for EBR-II wastes was established to accommodate these radionuclides in the waste streams. The multiple phases in the waste forms can be expected to have differing corrosion behaviors and to affect each other's corrosion rates due to electrical coupling. It is important to characterize the effects on corrosion of electrically coupled phases in the multiphase waste form to better predict waste form degradation and environmental source terms.

The primary objective of this study was to characterize the long term electrochemical response and evolution of metallic waste forms exposed to dilute alkaline and acidic brines. This long-term electrochemical monitoring of corrosion is termed hybrid testing in this report. Solution samples were taken from the hybrid test cell electrolyte to determine the release rates of specific elements and relate those release rates to the measured electrochemical responses. The reference RAW3-(URe) alloys were utilized for this work, with the Zr content of the alloys being varied from about 0.3-2 % (atomic). The RAW3-(URe) waste form is a surrogate for advanced electrometallurgical treatment waste.

1.2 Present Work

This report documents activities that included electrochemical, SEM/EDS characterization, and solution sampling to investigate the ability of using electrochemical methods to provide an accelerated degradation metric and correlate to the electrochemical responses the leach rates of specific elements from the simulated waste forms. Three alloys were characterized, based on the RAW3-(URe) formulations that varied in their Zr concentration. The RAW3-(URe) based alloys were formulated in FY12 (Olson 2012). RAW3 represents waste forms made with HT9 steel and a minimum amount of added Zr, with the Zr content varied from 0.3-2 % (atomic). The Zr in the alloy forms a phase responsible for sequestration of actinides. In this report the evaluated alloys are referred to using the ratio of Zr to U in the alloy, with U acting as the surrogate for other actinides and kept at a constant 1% (atomic).

This report meets milestone M3FT-15SR0307112 *Prepare samples and conduct electrochemical/immersion testing*, for work package # FT-15SR030711 Rev 1.

2. EXPERIMENTAL

2.1 Materials

The materials used for alloy fabrication were high purity. The material components and procedures for fabricating them can be found in earlier work (Olson 2012). The chemicals used to make solutions for electrochemical testing were high purity, and the water used for the solutions was demineralized.

2.2 Electrode Chemistry and Phase Distribution Data

Select relevant data on the electrodes used in this work was compiled and is listed. This data is of use for analyzing the electrochemical data.

Three alloys based on RAW3 formulation studies were also examined. Their formulation and averaged elemental make up can be found in Table 1 and Table 2 respectively. The alloys have a U:Zr ratio of 1:2, 1:1, and 3:1.

Table 1 HT9 bearing AWF test matrix measured mass additions.

| Name | Reagent Mass (g) | | | | | | | |
|--------|------------------|------|------|------|------|------|------|------|
| | HT9 | Zr | Mo | Ru | Pd | Re | Rh | U |
| 1U:2Zr | 8.08 | 0.33 | 0.19 | 0.18 | 0.19 | 0.32 | 0.18 | 0.46 |
| 1Zr:1U | 8.08 | 0.17 | 0.19 | 0.18 | 0.19 | 0.32 | 0.18 | 0.46 |
| 3U:1Zr | 8.08 | 0.06 | 0.19 | 0.18 | 0.19 | 0.32 | 0.18 | 0.45 |

Table 2 HT9 bearing AWF test matrix compositions, present in the alloys above 0.1% by mass or atom, including those of the HT9. Due to rounding, some elements may not show up in all measurement units

| Name | Mass Percent | | | | | | | | | | |
|--------|----------------|-----|------|-----|-----|-----|-----|-----|-----|-----|-----|
| | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1U:2Zr | 9.7 | 0.6 | 68.7 | 0.5 | 3.3 | 2.7 | 1.8 | 1.8 | 1.9 | 3.2 | 4.6 |
| 1U:1Zr | 9.9 | 0.6 | 69.9 | 0.5 | 1.7 | 2.8 | 1.8 | 1.8 | 1.9 | 3.3 | 4.7 |
| 3U:1Zr | 10.0 | 0.6 | 70.7 | 0.5 | 0.6 | 2.8 | 1.9 | 1.9 | 2.0 | 3.3 | 4.7 |
| Name | Atomic Percent | | | | | | | | | | |
| | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1U:2Zr | 11.5 | 0.6 | 76.2 | 0.5 | 2.3 | 1.8 | 1.1 | 1.1 | 1.1 | 1.1 | 1.2 |
| 1U:1Zr | 11.7 | 0.6 | 77.1 | 0.5 | 1.2 | 1.8 | 1.1 | 1.1 | 1.1 | 1.1 | 1.2 |
| 3U:1Zr | 11.8 | 0.7 | 77.7 | 0.5 | 0.4 | 1.8 | 1.1 | 1.1 | 1.1 | 1.1 | 1.2 |

Several regions are present in the alloys that will be referred to as phases in this report, although they are more properly thought of as commonly found composition regions. The main phases commonly observed are shown in Figure 1, and their corresponding elemental compositions described in Table 3, Table 4, Table 5, Table 6, and Table 7.

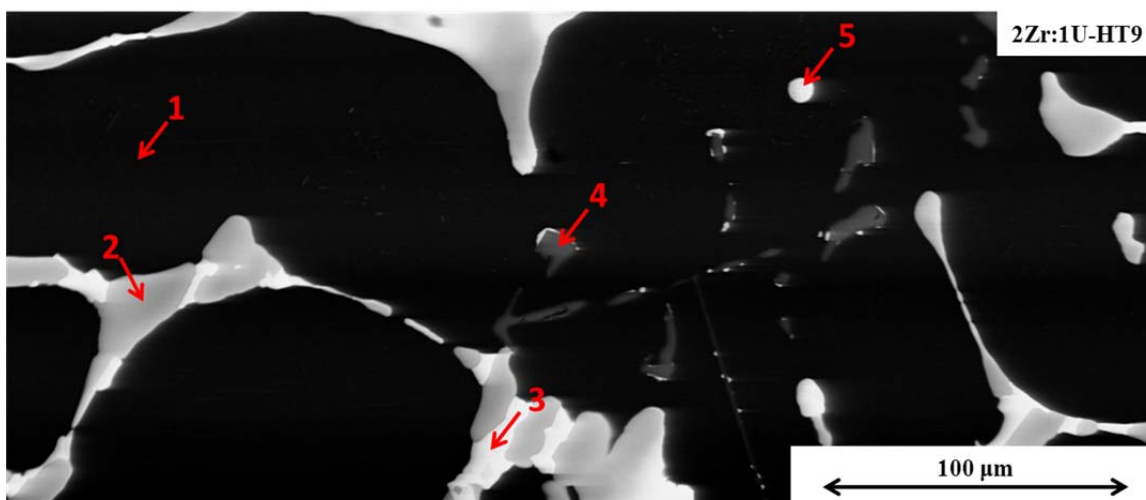


Figure 1. 2Zr:1U-HT9 alloy.

Table 3 EDS point scans of apparent similar composition ranges corresponding to composition region 1 in Figure 1, and corresponding compositions from the 1Zr:1U-HT9 alloy. This region is believed to correspond to ferrite.

| Steel Phase (atomic %) | | | | | | | | | | | |
|------------------------|--------------|-----|------|-----|-----|-----|-----|-----|-----|-----|-----|
| Alloy | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 2Zr:1U-HT9 | 12.8 | 2.2 | 80.4 | 0.4 | 0.0 | 1.8 | 0.9 | 0.4 | 0.3 | 1.0 | 0.0 |
| 1Zr:1U-HT9 | 12.2 | 2.3 | 80.4 | 0.4 | 0.0 | 1.8 | 1.2 | 0.5 | 0.3 | 0.9 | 0.0 |
| 1Zr:3U-HT9 | Not analyzed | | | | | | | | | | |

Table 4 EDS point scans of apparent similar composition ranges corresponding to composition region 2 in Figure 1, and corresponding compositions from the 1Zr:1U-HT9 bearing alloy. This phase was not observed in any EDS measurements of the 1Zr:3U-HT9 scoping alloy, although many EDS point measurements were taken in regions thought likely to contain this composition.

| (Zr, U)Fe ₂ Phase (atomic %) | | | | | | | | | | | |
|---|--------------|-----|------|-----|------|-----|-----|-----|-----|-----|-----|
| Alloy | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 2Zr:1U-HT9 | 2.4 | 0.9 | 57.3 | 1.7 | 20.9 | 1.1 | 1.1 | 4.9 | 3.3 | 0.0 | 6.4 |
| 1Zr:1U-HT9 | 2.2 | 1.0 | 55.3 | 1.7 | 19.9 | 1.4 | 1.1 | 7.2 | 2.9 | 0.0 | 7.5 |
| 1Zr:3U-HT9 | Not detected | | | | | | | | | | |

Table 5 EDS point scans of apparent similar composition ranges corresponding to composition region 3 in Figure 1, and corresponding compositions from the 1Zr:1U-HT9 and 1Zr:3U-HT9 alloys.

| (Zr, U)Pd ₂ Phase (atomic %) | | | | | | | | | | | |
|---|-----|-----|-----|-----|------|-----|-----|-----|------|-----|------|
| Alloy | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 2Zr:1U-HT9 | 0.6 | 0.0 | 5.6 | 1.3 | 12.5 | 0.8 | 0.0 | 0.0 | 62.1 | 0.0 | 17.1 |
| 1Zr:1U-HT9 | 0.0 | 0.0 | 4.9 | 1.8 | 12.5 | 0.9 | 0.0 | 0.0 | 61.1 | 0.0 | 18.8 |
| 1Zr:3U-HT9 | 1.0 | 0.0 | 3.8 | 1.1 | 0.4 | 0.0 | 0.0 | 0.0 | 66.8 | 0.0 | 26.8 |

Table 6 EDS point scan of apparent sphere composition region shown by point 5 in Figure 1, and corresponding sphere compositions from the 1Zr:1U-HT9 and 2Zr:1U-HT9 alloys.

| (Zr, U)(Fe, Pd, Rh) ₃ Phase (atomic %) | | | | | | | | | | | |
|---|-----|-----|------|-----|------|-----|-----|------|------|-----|------|
| Alloy | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 2Zr:1U-HT9 | 2.5 | 0.8 | 51.6 | 3.1 | 8.3 | 0.0 | 0.0 | 8.2 | 7.2 | 0.0 | 18.5 |
| 1Zr:1U-HT9 | 1.9 | 0.6 | 39.2 | 3.8 | 10.2 | 0.0 | 0.0 | 12.7 | 11.7 | 0.0 | 19.8 |
| 1Zr:3U-HT9 | 2.6 | 0.4 | 46.8 | 2.8 | 0.0 | 0.0 | 3.5 | 12.2 | 7.2 | 0.0 | 24.5 |

Table 7 EDS point scans of apparent similar composition ranges corresponding to composition region 4 in Figure 1, and corresponding compositions from the 1Zr:1U-HT9 and 1Zr:3U-HT9 bearing alloys.

| Zr Rich Phase (atomic %) | | | | | | | | | | | |
|--------------------------|-----|-----|-----|-----|------|-----|-----|-----|-----|-----|-----|
| Alloy | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 2Zr:1U-HT9 | 0.6 | 0.0 | 3.5 | 0.1 | 90.7 | 5.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 1Zr:1U-HT9 | 1.2 | 0.1 | 6.3 | 0.1 | 87.3 | 4.8 | 0.0 | 0.0 | 0.0 | 0.0 | 0.2 |
| 1Zr:3U-HT9 | 0.4 | 0.0 | 2.8 | 0.0 | 88.7 | 5.7 | 0.0 | 0.0 | 0.0 | 0.0 | 2.3 |

The data in Table 8 is of direct use for performing calculations with the electrochemical data; such as analyzing the LPR and Tafel data to determine the polarization resistance (R_p), anodic Tafel constant (β_A), cathodic Tafel constant (β_C), corrosion rates, and corrosion currents. There were two electrodes made from the 1U:2Zr alloy, with one being used for the alkaline brine tests, and the other for the acidic brine tests. This was done so that hybrid testing could continue while electrodes were in cue for analysis by SEM/EDS, or being repolished. The electrode surface areas were kept constant by attaching an insulated wire to the alloys, then embedding the alloys in Epofix nonconductive epoxy, prior to electrode surface preparation.

Table 8 Electrode density, equivalent weight, and Surface area of the alloys.

| Designation | Density (g/cm ³) | Equivalent Weight | Surface area (cm ²) |
|-------------|------------------------------|-------------------|---------------------------------|
| 1U:2Zr (a) | 9.0 | 27.0 | 1.026 |
| 1U:2Zr (b) | 9.0 | 27.0 | 0.292 |
| 1U:1Zr | 9.1 | 27.1 | 0.931 |
| 3U:1Zr | 9.1 | 27.1 | 0.801 |

Figure 2 and Figure 3 show the microstructure of the as fabricated pre-exposure multiphase alloys used in the hybrid tests.

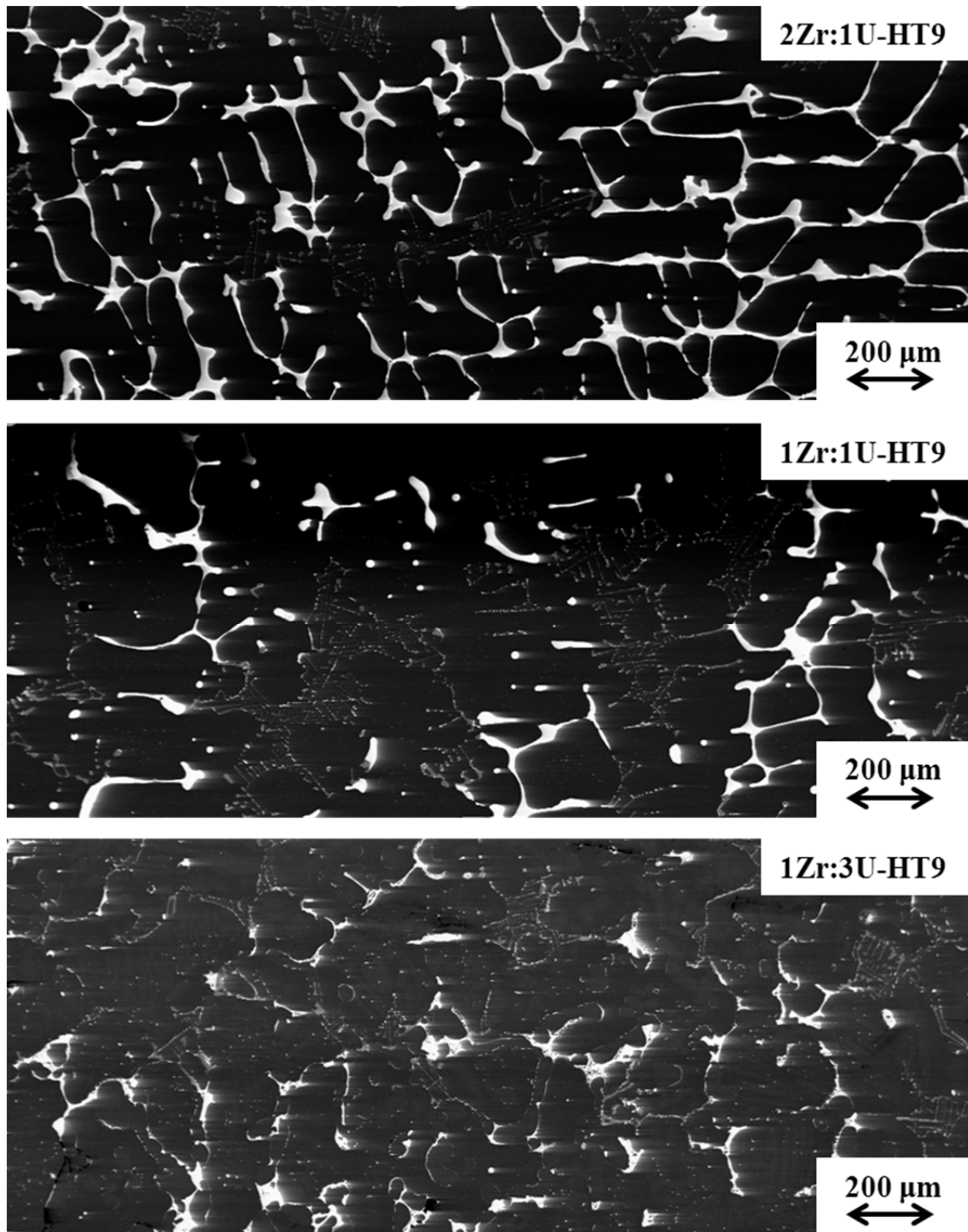


Figure 2 Low magnification SEM showing characteristic morphologies of the fabricated alloys for Zr:U studies.

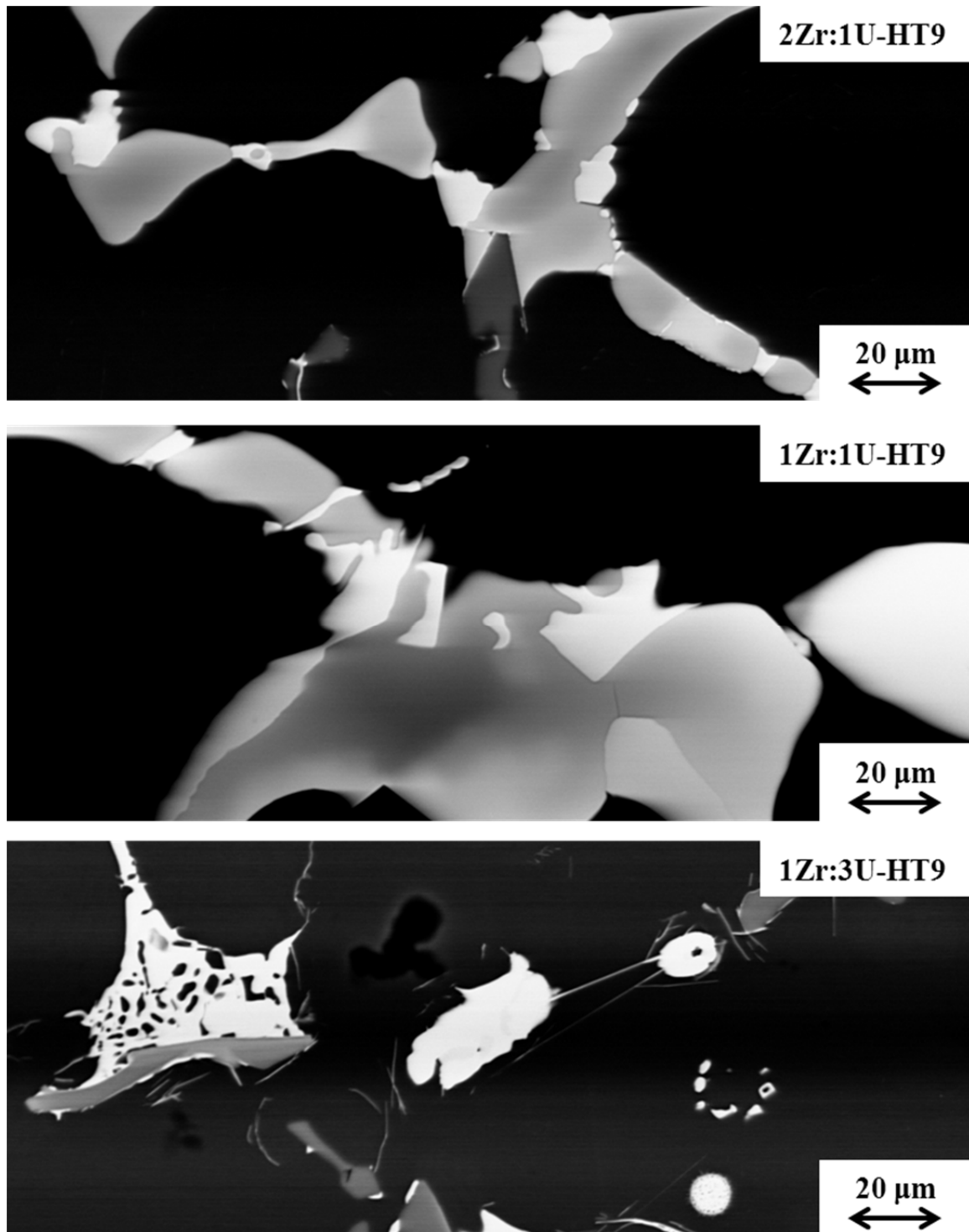
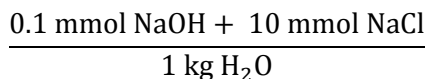


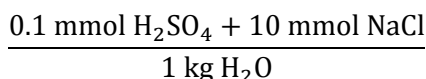
Figure 3 Higher magnification SEM showing characteristic morphologies of the fabricated alloys for Zr:U studies.

2.3 Electrochemistry Parameters

Specific test parameters can be found in APPENDIX 1: Electrochemical Measurement Data, with highlights summarized here. The electrochemical testing was undertaken in Greene cells that held 700 mL of solution. The solutions used were alkaline brine and acidic brine, respectively, from the Electrochemical Testing Protocol:



And



To make the solution, a concentrated 100x strength solution was made, then diluted with demineralized water. The demineralized water was aerated by being left exposed to air for 24 hours. The solution temperatures held constant at 23 °C.

The following electrochemical measurements schedule was mostly adhered to for the Zr matrix tests:

30 sec. OCP → LPR → 30 sec. OCP → Tafel → 30 sec. OCP → Pot. Dyn. → 30 sec. OCP

Electrode Repolished

Repeat below action 5 times

30 sec. OCP →

EIS at Hold Potential →

LPR →

Hold Potential for 24 hr →

Repeat below action 5 times

30 sec. OCP →

EIS at Hold Potential →

LPR →

Repeat below action 4 times

Hold Potential for 24 hr →

30 sec. OCP → LPR → 30 sec. OCP → Tafel

In practice, some of the tests were paused and eventually restarted, or terminated prior to reaching conclusion due to power interruptions or other events. The hybrid tests undertaken and conditions are summarized in Table 9.

Table 9 Summary of test conditions and length of hybrid tests undertaken.

| Alloy | Potential (mV) | Solution | Ended Program | Explanation |
|--------|----------------|----------------|---------------|--|
| 1U:2Zr | 50 | Alkaline brine | Yes | - |
| 1U:2Zr | 65 | Alkaline brine | Yes | - |
| 1U:2Zr | 300 | Acidic brine | No | power outage of 5 days at day ~22 then resumed |
| 3U:1Zr | 50 | Alkaline brine | Yes | - |
| 3U:1Zr | -200 | Alkaline brine | No | Power outage led to tests ending 1 week early |
| 1U:1Zr | 300 | Acidic brine | No | Short between WE and RE at day ~13 power outage of 5 days at day ~22 then resumed |

The following individual electrochemical measurement parameters were used:

Open Circuit Potential

Time per point (s): 10

Duration (s): from 30s when used just before measurements, to ~23 hours for the long-term measurements

Drift Rate (mV/min): 0

Potentiostatic hold

Potential (V): 50, 65, 300, or -200 mV vs SCE (RHE used in tests)

Time per point (s): 10

Duration (s): ~23 hours (sometimes repeated 3 times in a row)

Potentiostatic EIS

AC Properties

Start Freq (Hz): 100 000

End Freq (Hz): 0.001

Amplitude (mV RMS): 10

DC Properties

Step or Scan Potential (V): Step=0, vs OC for OCP, At Potentiostatic hold potential for those measurements

Scan Properties

Points per Decade: 10

Data Quality: 1

Measurements Delay (s): 0

Linear Polarization Resistance (LPR)

Initial Potential (V): -0.02 vs OC

Final Potential (V): 0.02 vs OC

Sweep rate (mV/s): 0.167

Tafel

Initial Potential (V): -0.25 vs OC

Final Potential (V): 0.25 vs OC

Sweep rate (mV/s): 0.167

Potentiodynamic

Initial Potential (V): -0.2 vs OC

Final Potential (V): 0.7 vs OC

Sweep rate (mV/s): 0.167

Current limit (mA): 10

2.4 Analytical Equipment

SEM and EDS Analysis

A LEO 440 SEM with an Inca Wave-7060 or Oxford X-ray microanalysis system for EDS was used for post exposure morphological examination. The SEM and EDS system was contained in a negative atmosphere glove box for handling radiological samples. The majority of the SEM images utilized a backscattered electron (BSE) detector. The accelerating voltage used for the BSE and EDS analyses was 30 kV.

Mass Measurements

The mass of the alloy waste form components, crucibles, and all other pertinent mass information, was measured on a Sartorius CP324S analytical balance.

Gamry Potentiostats

Electrochemical measurements were made with Gamry Reference 600 Potentiostats controlled with Gamry software. Electrochemical data was analyzed using Microsoft Excel and CorrView software.

3. RESULTS AND DISCUSSION

Three alloys based on the RAW3-(URe) formulation were characterized with electrochemical measurements during extended potentiostatic holds of up to 5 weeks while exposing the alloys to corrosive conditions. This characterization was termed hybrid testing. Simultaneously, solution measurements were periodically taken to correlate to the electrochemical data. Post exposure/monitoring select electrodes were characterized via SEM/EDS.

3.1 Potentiodynamic Measurements

Potentiodynamic measurements were taken on freshly polished electrodes in alkaline brines and acidic brines, shown respectively in Figure 4 and Figure 5. A hump appears near ~10 mV in the anodic half of the curve in the alkaline brines. In both solutions, the higher Zr content alloy waste

form has higher current densities in mildly oxidizing potentials as well as a more noble corrosion potential.

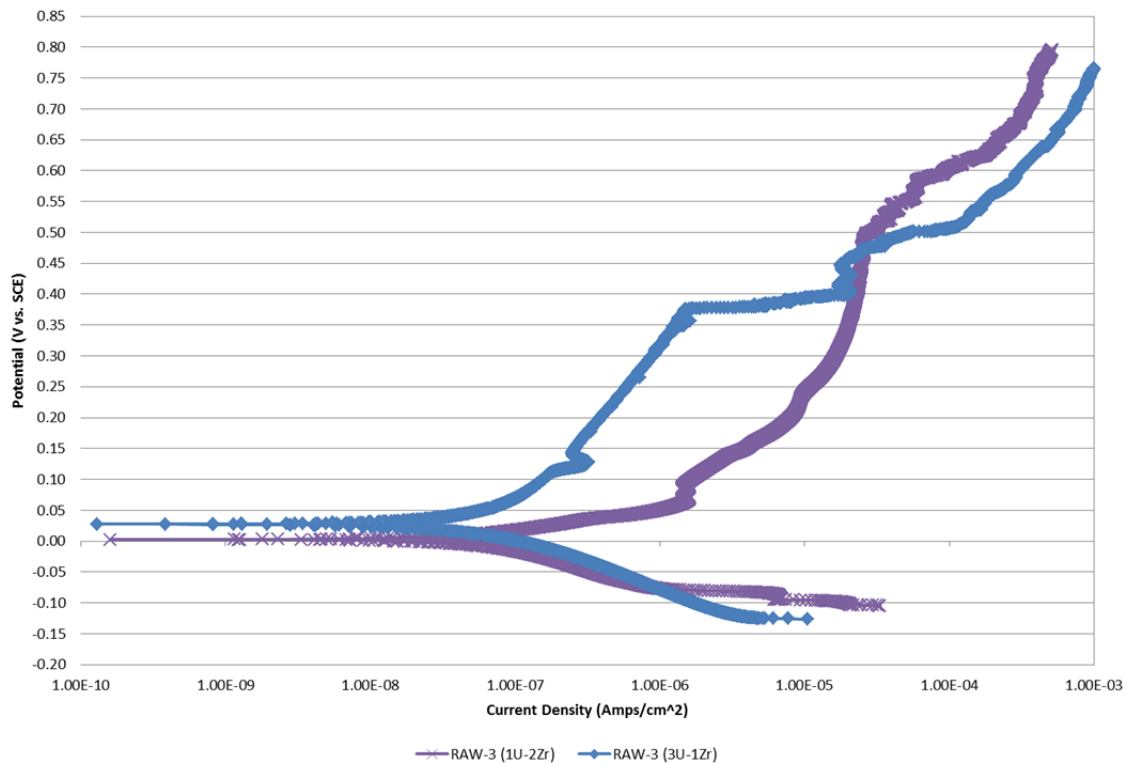


Figure 4 Comparison of Potentiodynamic scans of RAW-3(URe) alloys with fresh polished surfaces in alkaline brine.

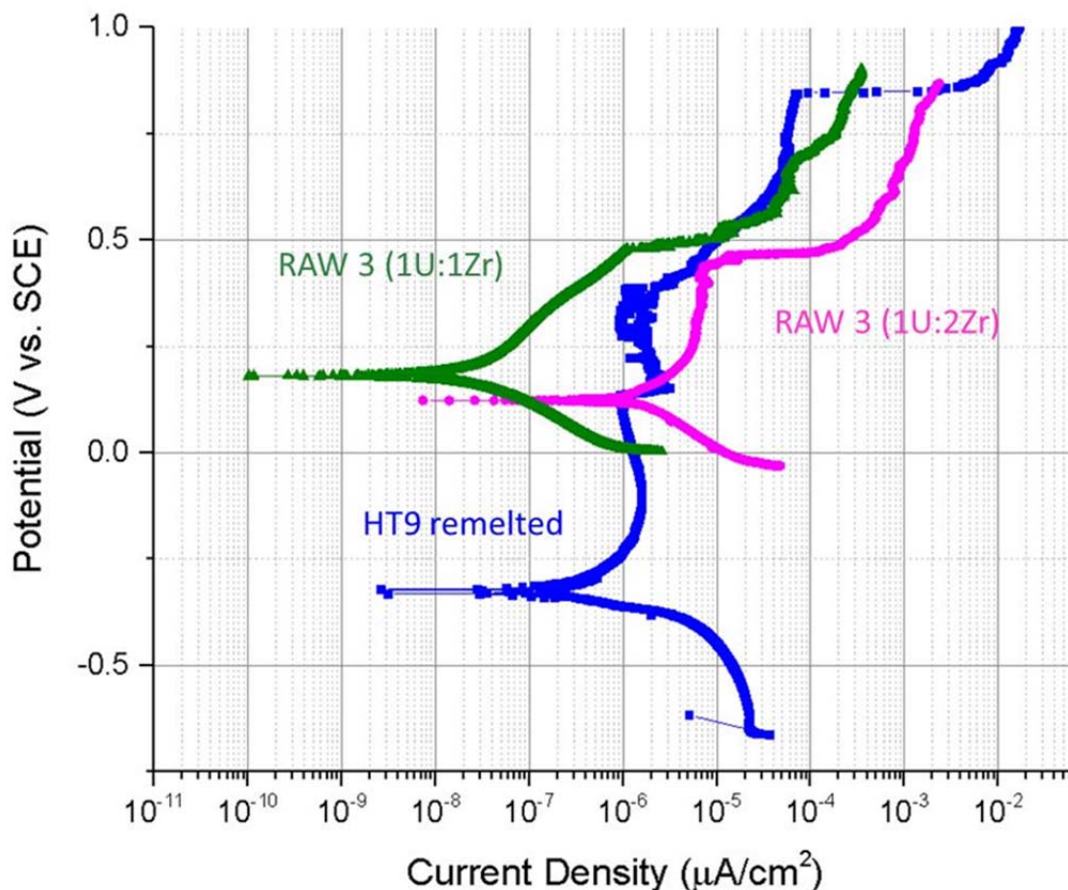


Figure 5 Comparison of Potentiodynamic scans of RAW-3(URe) alloys with fresh polished surfaces in acidic brine.

3.2 Potentiostatic Corrosion Current Densities

The primary measurement of the hybrid immersion/electrochemical test was the recording of c alloy waste form corrosion currents under constant electrode potential. This data was recorded for 6 hybrid tests, and the corrosion currents are summarized in Figure 6, Figure 7, Figure 8, Figure 9, Figure 10, and Figure 11. After the Potentiodynamic measurements described in section 3.1, other electrochemical measurements on freshly polished surfaces, and after subsequent hybrid tests, the electrode surfaces were repolished for subsequent hybrid tests.

The alkaline brine 50 mV potentiostatic hold potential for the 1U:2Zr alloy electrode investigates the region where the corrosion current appears to stabilize in a Potentiodynamic scan for a bare surface electrode, shown in Figure 4. The 3U:1Zr alloy electrode in alkaline brine was held at the same 50 mV potential of the 1U:2Zr alloy electrode for comparison. The alkaline brine 65 mV potentiostatic hold potential for the 1U:2Zr alloy electrode further investigates this region. The alkaline brine 65 mV potentiostatic hold of the 1U:2Zr alloy indicates a reducing surface condition, indicating that perhaps the diamond polishing of the alloy electrode surface may have been insufficient to refresh the electrode surface back to pre-immersion condition. The same

electrode was used for the alkaline brine 50 mV and the 65 mV tests, with the electrode surface repolished using 1 micron diamond past after the 50 mV test and before the 65 mV test; a new electrode was used for the acidic brine 300 mV test.

The alkaline brine -200 mV potentiostatic hold potential for the 3U:1Zr alloy electrode investigates the potential where this alloy electrode drifted to during open circuit hybrid monitoring reported in earlier work (Olson 2014). The 300 mV potential holds in the acidic brine for the 1U:2Zr and 1U:1Zr alloys was chosen to investigate electrode behavior in a similar passive region observed for the acidic brine potentiodynamic scans observed in Figure 5.

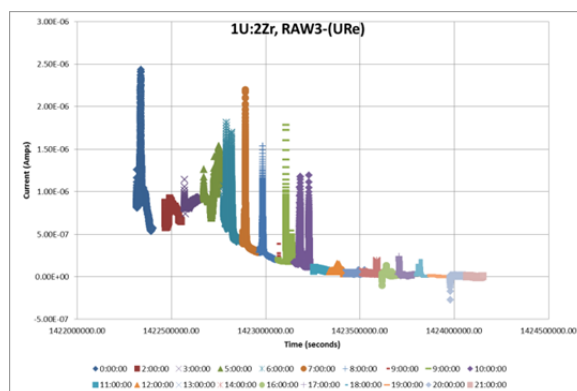


Figure 6 Current compilation with time for the 1U:2Zr electrode held at 50 mV in alkaline brine.

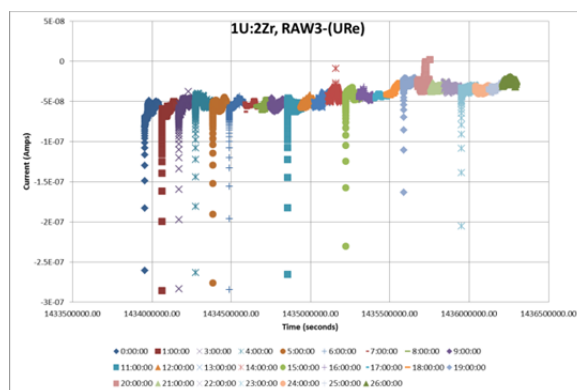


Figure 7 Current compilation with time for the 1U:2Zr electrode held at 65 mV in alkaline brine.

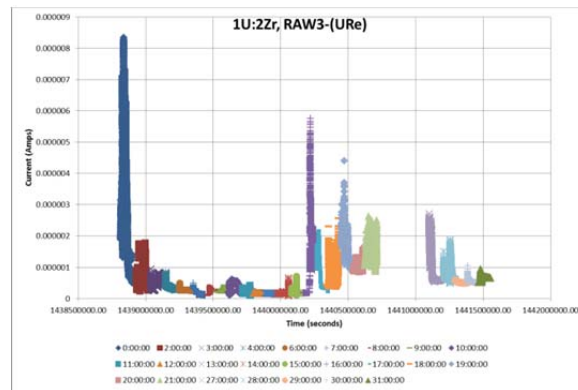


Figure 8 Current compilation with time for the 1U:2Zr electrode held at 300 mV in acidic brine.

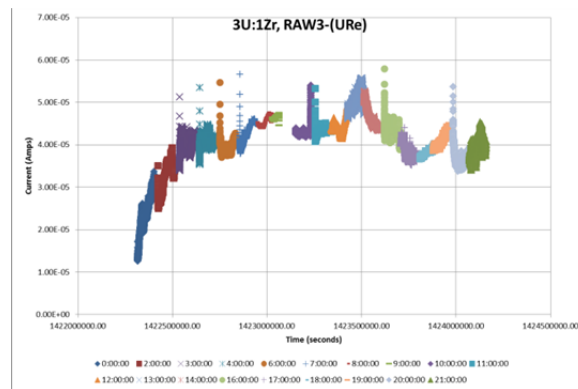


Figure 9 Current compilation with time for the 3U:1Zr electrode held at 50 mV in alkaline brine.

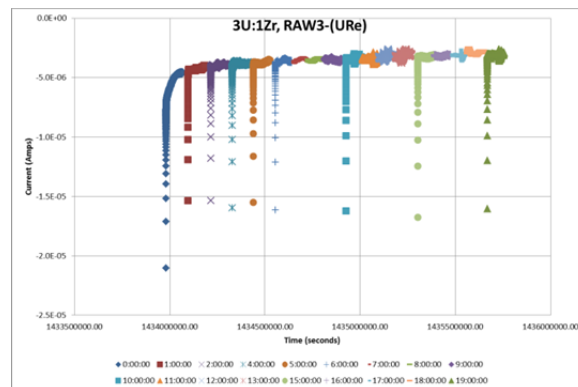


Figure 10 Current compilation with time for the 3U:1Zr electrode held at -200 mV in alkaline brine.

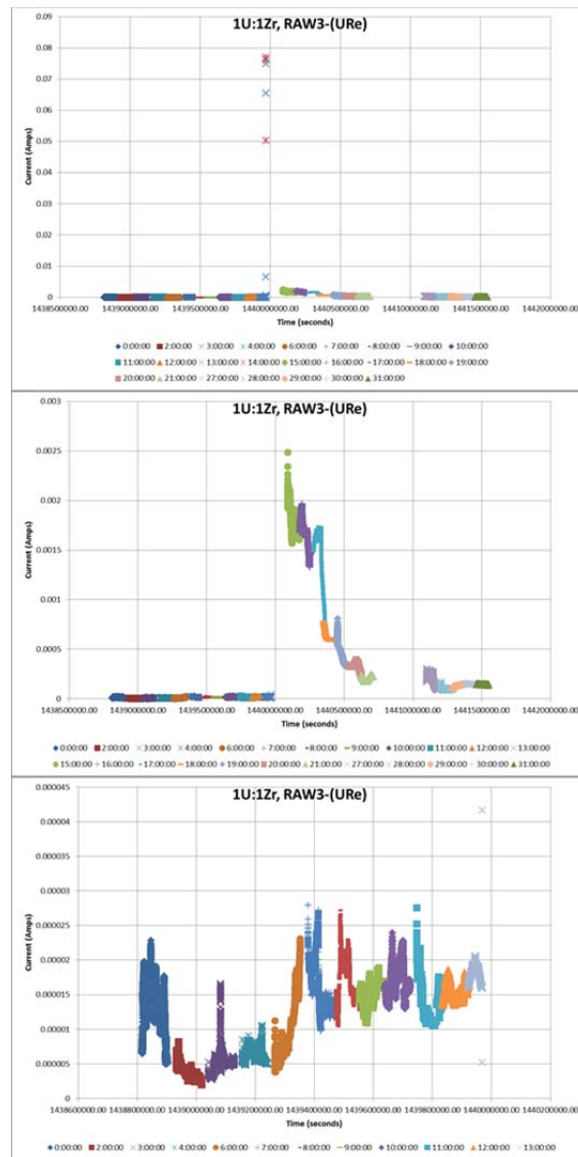


Figure 11 Current compilation with time for the 1U:1Zr electrode held at 300 mV in acidic brine.

3.3 Tafel: Initial and Final

Tafel measurements from before and after hybrid tests and analyzed results are summarized in Table 10 and related Tafel scans compared in Figure 12, Figure 13, Figure 14, and Figure 15. The Tafel constants and Stern-Geary coefficients calculated from the Tafel measurements were later used in analysis of the linear polarization resistance (LPR) measurements. The data fits used in the CorrView software to analyze the electrochemical measurement data are shown in APPENDIX 1: Electrochemical Measurement Data.

Table 10 Tafel measurements immediately after polishing the alloy electrodes and after the long term exposures in alkaline and acidic brines. Tafel analysis from data using CorrView Auto fit functionality. *Manual fit to data using some Tafel auto fit parameters. Note, for the 3U:1Zr electrode, the -200 mV Tafel was not taken due to a prolonged power outage that stopped testing prior to the final Tafel measurement.

| Alkaline brine | | | | | | | |
|----------------|--------------------------------|--------------------------------------|--|------------------------------|-----------------------|-----------------------|------------|
| alloy | surface condition | Anodic Tafel Constant β_A (mV) | Cathodic Tafel Constant β_C (mV) | I_0 (Amp/cm ²) | E_0 (Volts vs. SCE) | Corrosion Rate (mmPY) | S-G Coeff. |
| 1U:2Zr | fresh polish | 107 | 87 | 3.42E-08 | 0.0180 | 8.70E-10 | 20.9 |
| 1U:2Zr | 21 Days at OCP | 173 | 94 | 4.83E-08 | 0.0254 | 4.75E-04 | 26.5 |
| 1U:2Zr | 28 days at 50 mV | 171 | 113 | 8.78E-08 | 0.0107 | 8.63E-04 | 29.6 |
| 1U:2Zr | 27 days at 65 mV | 274 | 173 | 1.19E-07 | 0.0273 | 1.17E-03 | 46.1 |
| 1U:2Zr | 27 days at 80 mV | 350 | 95 | 4.60E-07 | 0.0591 | 1.04E-02 | 32.5 |
| 3U:1Zr | fresh polish | 231 | 124 | 1.21E-07 | 0.0123 | 0.0012 | 35.1 |
| 3U:1Zr | 27 days at OCP | 520 | 266 | 4.77E-06 | -0.2147 | 0.0465 | 76.5 |
| 3U:1Zr | 28 days at 50 mV | 209 | 162 | 5.93E-06 | -0.3036 | 0.0579 | 39.7 |
| Acidic brine | | | | | | | |
| 1U:2Zr | fresh polish | 125.0 | 21.1 | 5.67E-07 | 0.1287 | 0.0056 | 7.8 |
| 1U:2Zr | 32 days at 300 mV ^a | 977.4 | 323.1 | 2.55E-06 | 0.0770 | 0.0251 | 105.6 |
| 1U:1Zr | fresh polish | 234.7 | 123.4 | 3.85E-08 | 0.1595 | 0.0004 | 35.2 |
| 1U:1Zr | 32 days at 300 mV ^b | 412.6 | 239.0 | 7.01E-05 | -0.1629 | 0.6843 | 65.8 |

^a=majority of time

^b= short 1/2 way through, 300 mV majority time

In Figure 12, it is seen that the 50 mV and 65 mV Tafel of the 1U:2Zr alloy electrodes does not result in a very different result compared to the fresh surface condition, or the surface condition

from a hybrid test that was run at open circuit conditions. However, holding the electrode at 80 mV did result in a significantly different behavior.

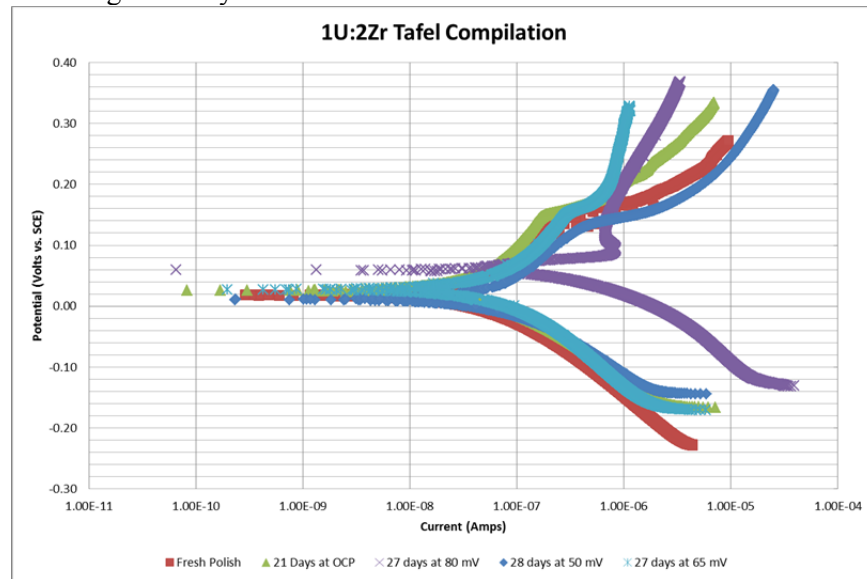


Figure 12 Tafel measurements immediately after polishing the 1U:2Zr electrode and after the long term exposures in alkaline brine.

Figure 13 indicate a corrosion potential at more reducing conditions when allowed to freely corrode or held at oxidizing potentials for long time periods.

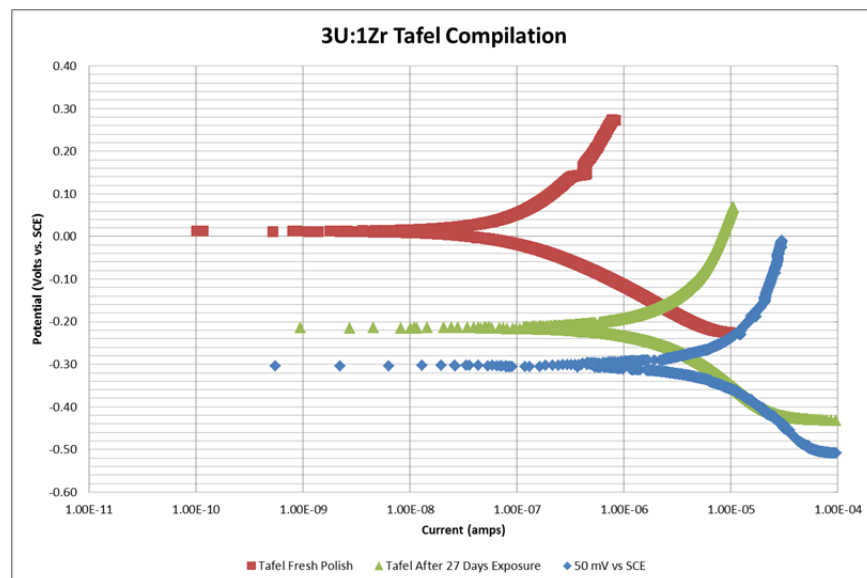


Figure 13 Tafel measurements immediately after polishing the 3U:1Zr electrode and after the long term exposures in alkaline brine. Note the -200 mV Tafel was not taken due to a prolonged power outage that stopped testing prior to the final Tafel measurement.

Figure 14 shows the Tafel measurement of the 1U:1Zr alloy electrode after a short that resulted in largescale corrosion of the surface, and therefore this measurement while being provided for completeness, should not be used for calculations.

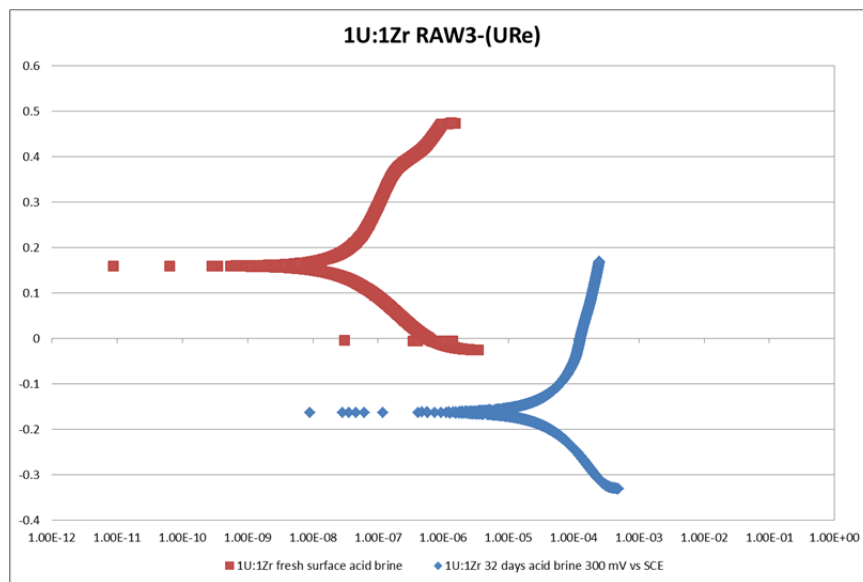


Figure 14 Tafel measurements immediately after polishing the 1U:1Zr electrode and after the long term exposure in acidic brine. Note the Tafel after the 300 mV potentiostatic test is of a heavily stripped surface due to a short between the WE and RE, that led the potentiostat to push large amounts of current into the WE to try to reestablish the 300 mV potential. Note also a sustained power outage at day ~22 may have led to a change in the passivation surface.

Figure 15 shows the corrosion potential of the 1U:2Zr alloy electrode became more reducing after extended exposure in acidic brine and in general appeared to become somewhat more passive.

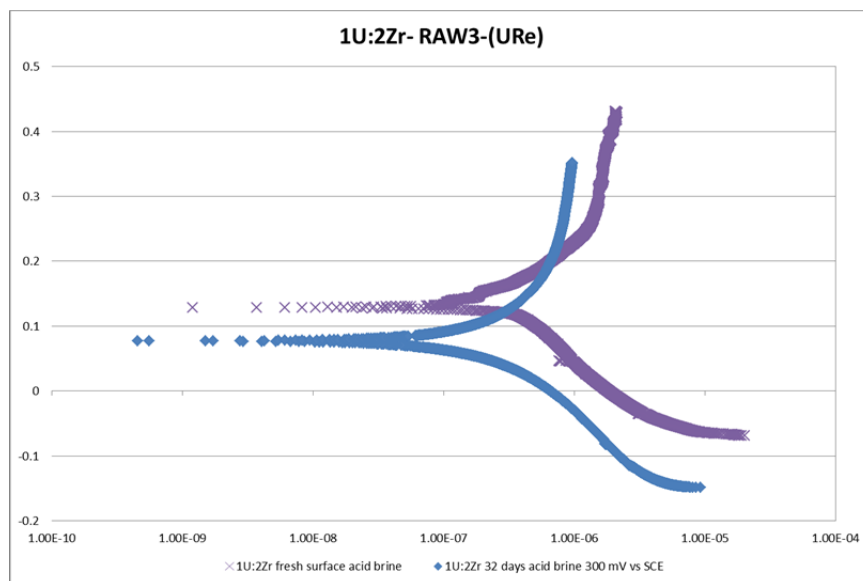


Figure 15 Tafel measurements immediately after polishing the 1U:2Zr electrode and after the long term exposure in acidic brine. Note also a sustained power outage at day ~22 may have led to a change in the passivation surface.

3.4 Polarization Resistance (LPR)

Analyzed linear polarization resistance data that was gathered during hybrid testing is compiled in Table 11, Table 12, Table 13, Table 14, Table 15, and Table 16. Figures showing the general trends, if any, for corresponding to each table for applicable data, are shown in Figure 16, Figure 17, Figure 18, Figure 19, Figure 20, and Figure 21.

As seen in Table 11 and Figure 16, for the alkaline brine 50mV 1U:2Zr alloy electrode, the polarization resistance increases the first week, before dropping back to near pre-test values.

Table 11 Analyzed values from the linear polarization resistance measurements taken during periodic interruptions to the potentiostatic holds. Analyzed using CorrView.

| 1U:2Zr, 50 mV, Alkaline Brine | | | | |
|-------------------------------|-----------------------------|--|--|-------------------------------|
| Elapsed Time (Days) | Corrosion Rate (mm/year) | R _p (Ω/cm ²) | I ₀ (Amp/cm ²) | E ₀ (V vs. SCE) |
| 0:00:00 | 0.0062 | 4.15E+04 | 6.29E-07 | -0.0004 |
| 2:00:00 | 0.0245 | 1.05E+04 | 2.50E-06 | -0.0162 |
| 3:00:00 | 0.0021 | 1.24E+05 | 2.11E-07 | -0.0709 |
| 5:00:00 | 0.0020 | 1.27E+05 | 2.05E-07 | -0.0887 |
| 6:00:00 | 0.0018 | 1.41E+05 | 1.85E-07 | -0.0846 |
| 7:00:00 | 0.0012 | 2.15E+05 | 1.21E-07 | -0.0516 |
| 11:00:00 | 0.0056 | 4.58E+04 | 5.69E-07 | 0.0234 |
| 16:00:00 | 0.0055 | 4.66E+04 | 5.59E-07 | 0.0508 |
| 20:00:00 | 0.0104 | 2.46E+04 | 1.06E-06 | 0.0756 |

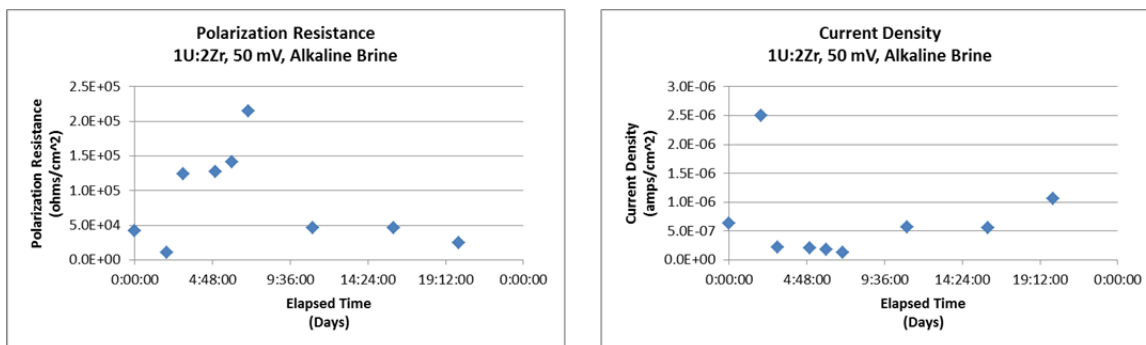


Figure 16 Linear polarization resistance and current density values calculated from measurements during periodic interruptions to the potentiostatic holds. Values taken from associated table.

As seen in Table 12 and Figure 17, for the alkaline brine 65mV 1U:2Zr alloy electrode, the polarization resistance remains flat during the entire hybrid test duration, indicating a passivating surface either developed very early, or did not develop at all.

Table 12 Analyzed values from the linear polarization resistance measurements taken during periodic interruptions to the potentiostatic holds. Analyzed using CorrView.

| 1U:2Zr, 65 mV, Alkaline Brine | | | | |
|-------------------------------|-----------------------------|--|--|-------------------------------|
| Elapsed Time (Days) | Corrosion Rate (mm/year) | R _p (Ω/cm ²) | I ₀ (Amp/cm ²) | E ₀ (V vs. SCE) |
| 0:00:00 | 0.0010 | 2.00E+05 | 1.04E-07 | 0.0783 |
| 1:00:00 | 0.0012 | 1.65E+05 | 1.27E-07 | 0.0763 |
| 3:00:00 | 0.0013 | 1.62E+05 | 1.29E-07 | 0.0743 |
| 4:00:00 | 0.0013 | 1.62E+05 | 1.29E-07 | 0.0719 |
| 5:00:00 | 0.0013 | 1.59E+05 | 1.31E-07 | 0.0728 |
| 6:00:00 | 0.0013 | 1.62E+05 | 1.29E-07 | 0.0736 |
| 11:00:00 | 0.0016 | 1.63E+05 | 1.60E-07 | 0.0718 |
| 15:00:00 | 0.0015 | 1.72E+05 | 1.52E-07 | 0.0694 |
| 19:00:00 | 0.0015 | 1.76E+05 | 1.48E-07 | 0.0640 |
| 23:00:00 | 0.0015 | 1.72E+05 | 1.52E-07 | 0.0677 |
| 27:00:00 | 0.0014 | 1.80E+05 | 1.45E-07 | 0.0650 |

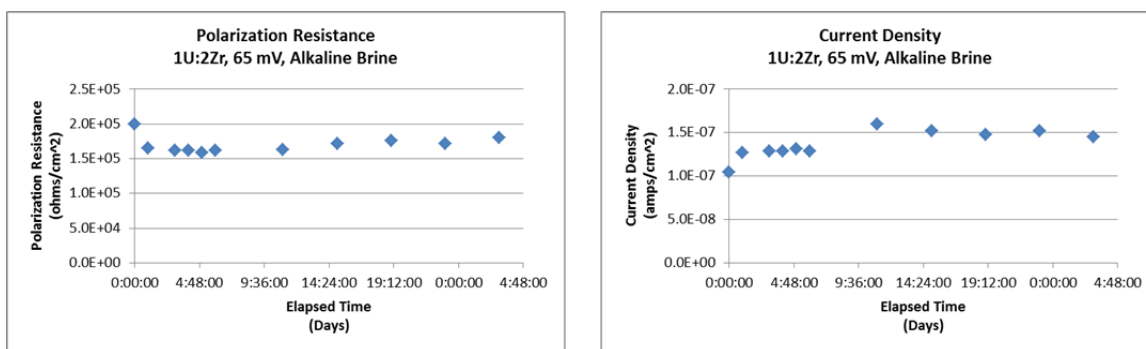


Figure 17 Linear polarization resistance and current density values calculated from measurements during periodic interruptions to the potentiostatic holds. Values taken from associated table.

As seen in Table 13 and Figure 18, for the alkaline brine 50 mV 3U:1Zr alloy electrode, the polarization resistance decreases within the first several days before remaining flat for the rest of the hybrid test duration, indicating a surface that becomes more active at first, then stays at that active state.

Table 13 Analyzed values from the linear polarization resistance measurements taken during periodic interruptions to the potentiostatic holds. Analyzed using CorrView.

| 3U:1Zr, 50 mV, Alkaline Brine | | | | |
|-------------------------------|-----------------------------|--|--|-------------------------------|
| Elapsed Time (Days) | Corrosion Rate (mm/year) | R _p (Ω/cm ²) | I ₀ (Amp/cm ²) | E ₀ (V vs. SCE) |
| 0:00:00 | 0.0357 | 7.13E+03 | 3.66E-06 | -0.162 |
| 2:00:00 | 0.0649 | 3.92E+03 | 6.66E-06 | -0.212 |
| 3:00:00 | 0.0806 | 3.16E+03 | 8.26E-06 | -0.247 |
| 4:00:00 | 0.0801 | 3.18E+03 | 8.21E-06 | -0.251 |
| 6:00:00 | 0.0842 | 3.02E+03 | 8.63E-06 | -0.248 |
| 7:00:00 | 0.0921 | 2.76E+03 | 9.44E-06 | -0.246 |
| 11:00:00 | 0.0959 | 2.65E+03 | 9.83E-06 | -0.255 |
| 16:00:00 | 0.0983 | 2.59E+03 | 1.01E-05 | -0.257 |
| 20:00:00 | 0.0871 | 2.92E+03 | 8.93E-06 | -0.256 |

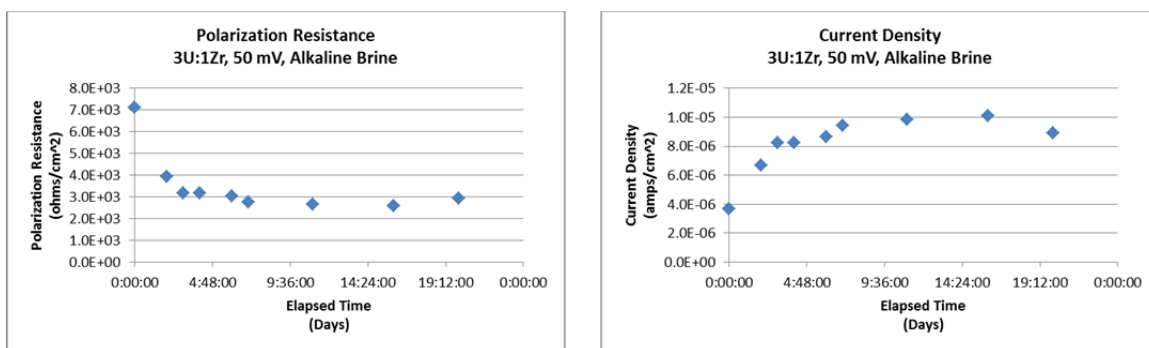


Figure 18 Linear polarization resistance and current density values calculated from measurements during periodic interruptions to the potentiostatic holds. Values taken from associated table.

As seen in Table 14 and Figure 19, for the alkaline brine -200 mV 3U:1Zr alloy electrode, the polarization resistance remains flat initially, before increasing gradually during the remaining hybrid test duration, indicating a gradually passivating surface.

Table 14 Analyzed values from the linear polarization resistance measurements taken during periodic interruptions to the potentiostatic holds. Analyzed using CorrView.

| 3U:1Zr, -200 mV, Alkaline Brine | | | | |
|---------------------------------|-----------------------------|-----------------------------------|--------------------------------|----------------------|
| Elapsed Time (Days) | Corrosion Rate (mm/year) | R_p (Ω/cm^2) | I_0 (Amp/ cm^2) | E_0 (V vs. SCE) |
| 0:00:00 | 0.0800 | 4.28E+03 | 8.20E-06 | -0.1551 |
| 2:00:00 | 0.0883 | 3.88E+03 | 9.05E-06 | -0.1734 |
| 3:00:00 | 0.0843 | 4.06E+03 | 8.64E-06 | -0.1735 |
| 5:00:00 | 0.0783 | 4.37E+03 | 8.02E-06 | -0.1714 |
| 6:00:00 | 0.0710 | 4.82E+03 | 7.28E-06 | -0.1716 |
| 7:00:00 | 0.0654 | 5.24E+03 | 6.70E-06 | -0.1670 |
| 11:00:00 | 0.0570 | 6.01E+03 | 5.84E-06 | -0.1652 |
| 16:00:00 | 0.0515 | 6.65E+03 | 5.28E-06 | -0.1598 |
| 20:00:00 | 0.0408 | 8.38E+03 | 4.19E-06 | -0.1583 |

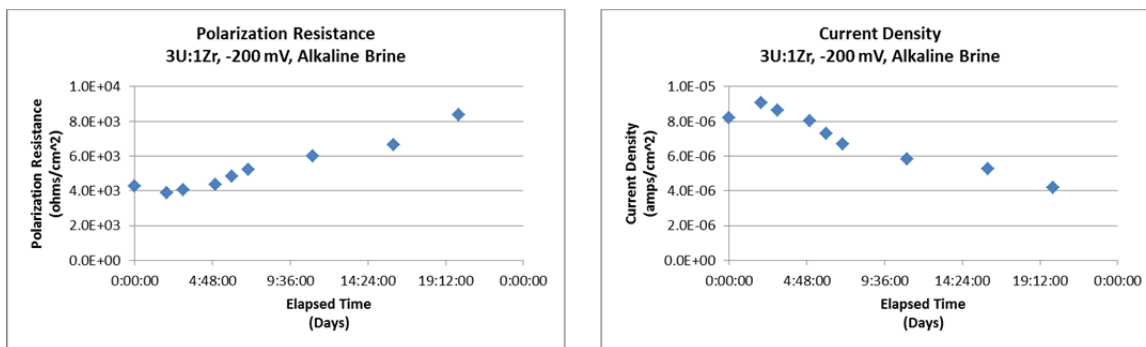


Figure 19 Linear polarization resistance and current density values calculated from measurements during periodic interruptions to the potentiostatic holds. Values taken from associated table.

As seen in Table 15 and Figure 20, for the acidic brine 300mV 1U:1Zr alloy electrode, the polarization resistance increases at first, before decreasing for the remainder of the hybrid test duration, indicating a passivating surface may have developed but did not remain stable.

Table 15 Analyzed values from the linear polarization resistance measurements taken during periodic interruptions to the potentiostatic holds. Analyzed using CorrView.

| 1U:1Zr, 300 mV, Acidic Brine | | | | |
|------------------------------|-----------------------------|--|--|-------------------------------|
| Elapsed Time (Days) | Corrosion Rate (mm/year) | R _p (Ω/cm ²) | I ₀ (Amp/cm ²) | E ₀ (V vs. SCE) |
| 0:00:00 | 0.0005 | 7.65E+05 | 5.03E-08 | 0.2126 |
| 0:00:00 | 0.0228 | 1.65E+04 | 2.34E-06 | 0.0623 |
| 2:00:00 | 0.0130 | 2.88E+04 | 1.34E-06 | 0.0461 |
| 3:00:00 | 0.0132 | 2.84E+04 | 1.35E-06 | 0.0482 |
| 4:00:00 | 0.0175 | 2.14E+04 | 1.80E-06 | 0.0100 |
| 6:00:00 | 0.0364 | 1.03E+04 | 3.73E-06 | -0.0264 |
| 7:00:00 | 0.0381 | 9.86E+03 | 3.90E-06 | -0.0663 |
| 11:00:00 | 0.0584 | 6.44E+03 | 5.98E-06 | -0.0957 |
| ^a 15:00:00 | 1.2050 | 3.12E+02 | 1.24E-04 | -1.0581 |
| ^a 19:00:00 | 1.1575 | 3.25E+02 | 1.19E-04 | -0.2874 |
| ^{a,b} 28:00:00 | 0.9396 | 4.00E+02 | 9.63E-05 | 0.0394 |
| ^{a,b} 32:00:00 | 0.9550 | 3.93E+02 | 9.79E-05 | -0.0545 |

^a pH change led to potential change due to pH dependency of RHE. Yellow and blue is corrected using neutral pH from previous alkaline brine tests. Not sure if pH change is completed until test on 28, but likely was at least close by 19 if not 15

^b loss of power led to change in surface potential, and therefore potential change in surface morphology after day 19, power went off on 8/27

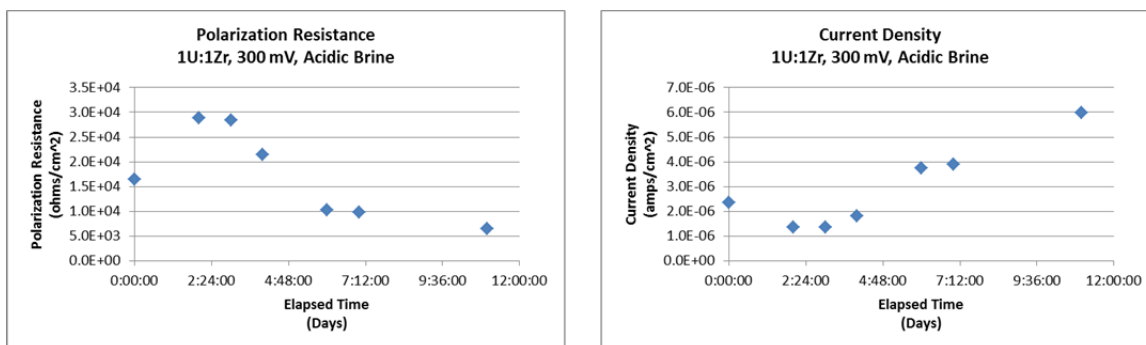


Figure 20 Linear polarization resistance and current density values calculated from measurements during periodic interruptions to the potentiostatic holds. Values taken from associated table.

As seen in Table 16 and Figure 21, for the acidic brine 300mV 1U:2Zr alloy electrode, the polarization resistance gradually increases during the duration of the hybrid test duration, indicating increased passivation of the surface.

Table 16 Analyzed values from the linear polarization resistance measurements taken during periodic interruptions to the potentiostatic holds. Analyzed using CorrView.

| 1U:2Zr, 300 mV, Acidic Brine | | | | |
|------------------------------|-----------------------------|--|--|-------------------------------|
| Elapsed Time (Days) | Corrosion Rate (mm/year) | R _p (Ω/cm ²) | I ₀ (Amp/cm ²) | E ₀ (V vs. SCE) |
| 0:00:00 | 0.0116 | 4.63E+04 | 1.18E-06 | 0.1750 |
| 0:00:00 | 0.0149 | 3.61E+04 | 1.52E-06 | 0.0616 |
| 2:00:00 | 0.0097 | 5.57E+04 | 9.82E-07 | 0.1356 |
| 3:00:00 | 0.0133 | 4.05E+04 | 1.35E-06 | 0.0954 |
| 4:00:00 | 0.0089 | 6.02E+04 | 9.09E-07 | 0.1273 |
| 6:00:00 | 0.0090 | 5.97E+04 | 9.16E-07 | 0.1409 |
| 7:00:00 | 0.0094 | 5.70E+04 | 9.60E-07 | 0.1444 |
| 11:00:00 | 0.0067 | 7.99E+04 | 6.85E-07 | 0.1660 |
| 15:00:00 | 0.0071 | 7.63E+04 | 7.17E-07 | 0.1692 |
| 19:00:00 | 0.0376 | 1.43E+04 | 3.83E-06 | -0.0147 |
| ^a 28:00:00 | 0.0240 | 2.24E+04 | 2.45E-06 | 0.0798 |
| ^a 32:00:00 | 0.0165 | 3.27E+04 | 1.67E-06 | 0.1038 |

^a loss of power led to change in surface potential, and therefore potential change in surface morphology after day 19, power went off on 8/27

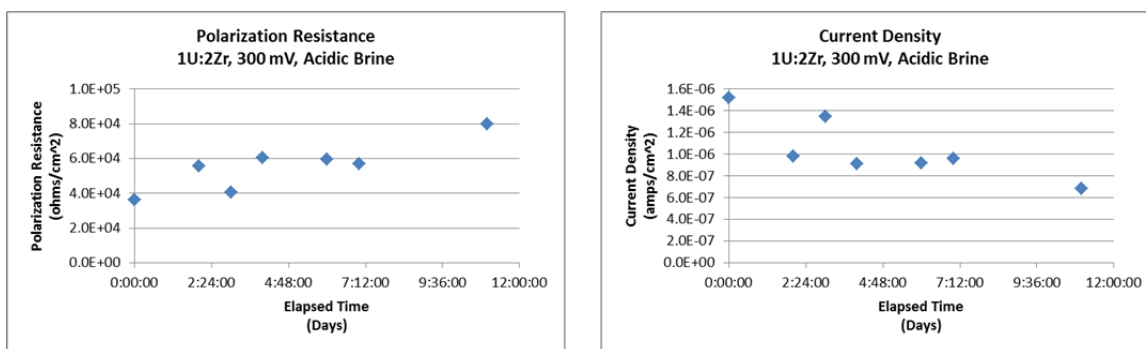


Figure 21 Linear polarization resistance and current density values calculated from measurements during periodic interruptions to the potentiostatic holds. Values taken from associated table.

3.5 Electrochemical Impedance Spectroscopy

The EIS data is used to probe the nature of the degradation of the alloy electrode surface and formation of any corrosion product films. The EIS data collectively indicates the 1U:2Zr alloy

forms a more protective oxidation layer for the tested conditions, in comparison to the 1:1Zr and 3U:1Zr alloys. The EIS data is presented as Bode gain plots, Bode phase plots, and Nyquist plots.

3.5.1 Bode Plots

In the Bode gain plots, Figure 22, the high frequency region of the curve is heavily influenced by solution resistance and the lower frequency range by the interaction between the electrode and the solution, with the lowest values giving understanding similar to the polarization resistance. The 1U:2Zr alloys appears more stable than the other alloys in both solutions based on the Bode gain plots, but less so in the acidic brine compared to the alkaline brine. Large changes in the electrode/solution interaction are observed for the 1) 1U:2Zr-50 mV-alkaline brine, 2) 3U:1Zr-50 mV-alkaline brine, and 3) 1U:1Zr-300 mV-acidic brine hybrid tests. In the case of the 1U:1Zr-300 mV-acidic brine hybrid test, the observed change is most likely due to neutralization of the solution due to the short that occurred between the alloy electrode and the reference electrode, that resulted in a large amount of metal to be rapidly dissolved into solution.

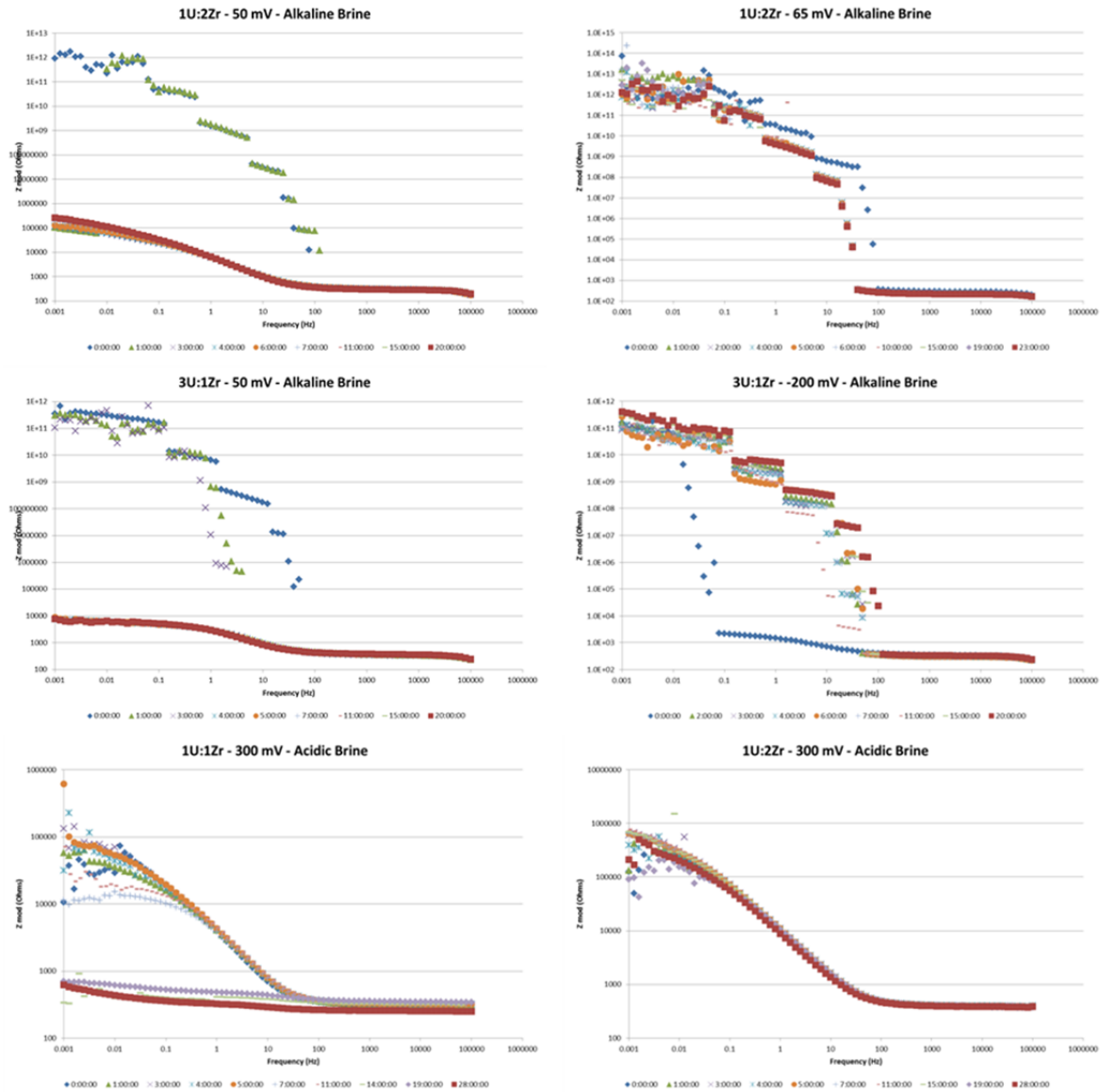


Figure 22 Bode gain plots for the 3 alloy electrodes in acidic brines and alkaline brines at a variety of imposed potentials.

The Bode phase plots, Figure 23, indicate that there are fast and slow events occurring between the electrode surface and the solution. The slow events decrease in phase angle within the first ~2 days for the 50 mV exposures in the alkaline brine for both the 1U:2Zr and 3U:1Zr alloys. There does not appear to be stabilization in slow events in the 65mV, 1U:2Zr alkaline brine hybrid test, nor the 3U:1Zr, -200 mV, alkaline brine tests.

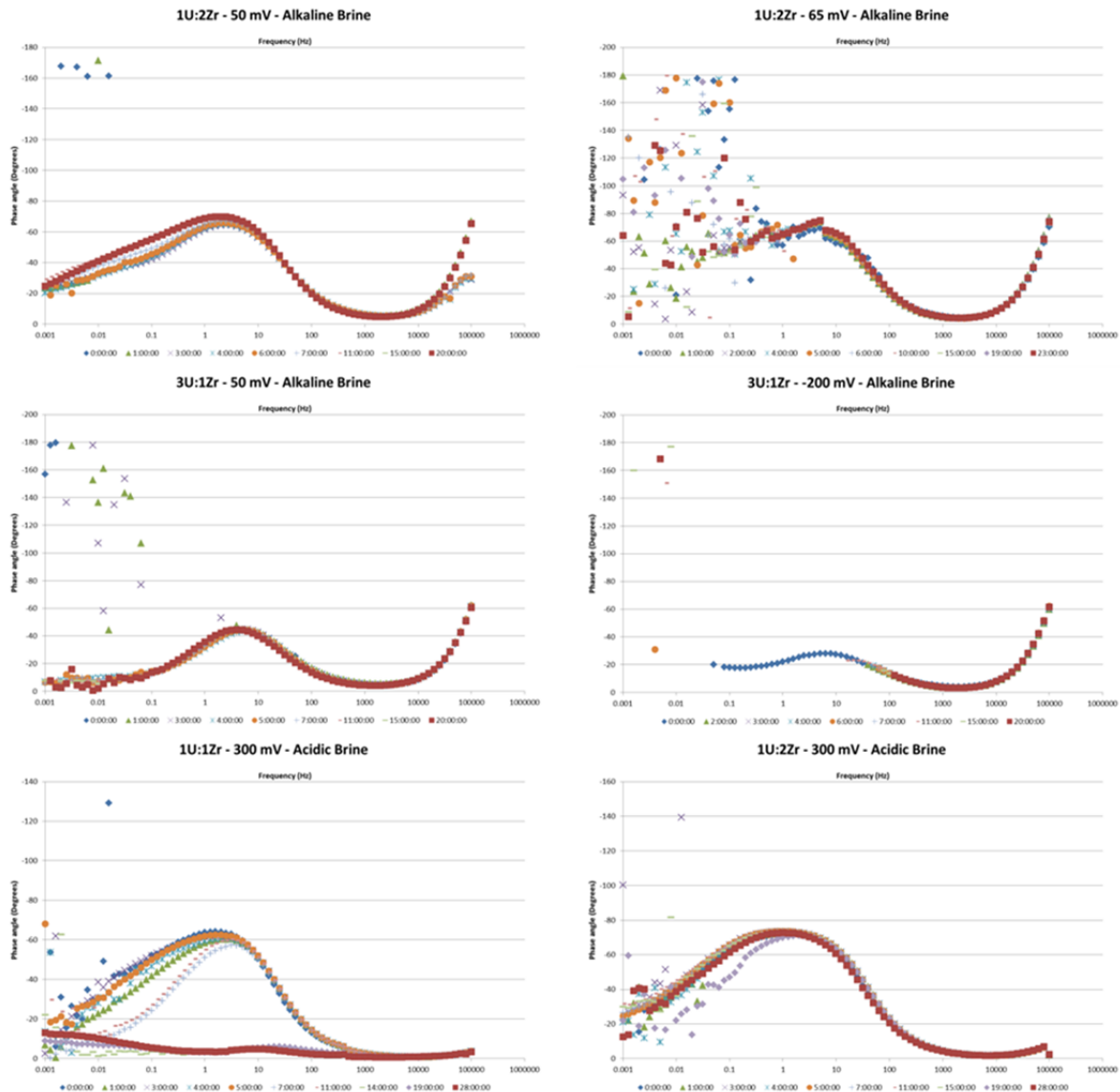


Figure 23 Bode phase plots of the 3 alloy electrodes in acidic brines and alkaline brines at a variety of imposed potentials.

3.5.2 Nyquist Plots

The Nyquist plots indicate an oxidation layer was likely forming and stable during the test duration for the 50 mV, 1U:2Zr and 3U:1Zr tests in alkaline brine, although it appeared to be more protective for the 1U:2Zr alloy electrode. The 1U:2Zr alloy surface passivation was tenuous in the acidic brines, with passivation decreasing after day 15.

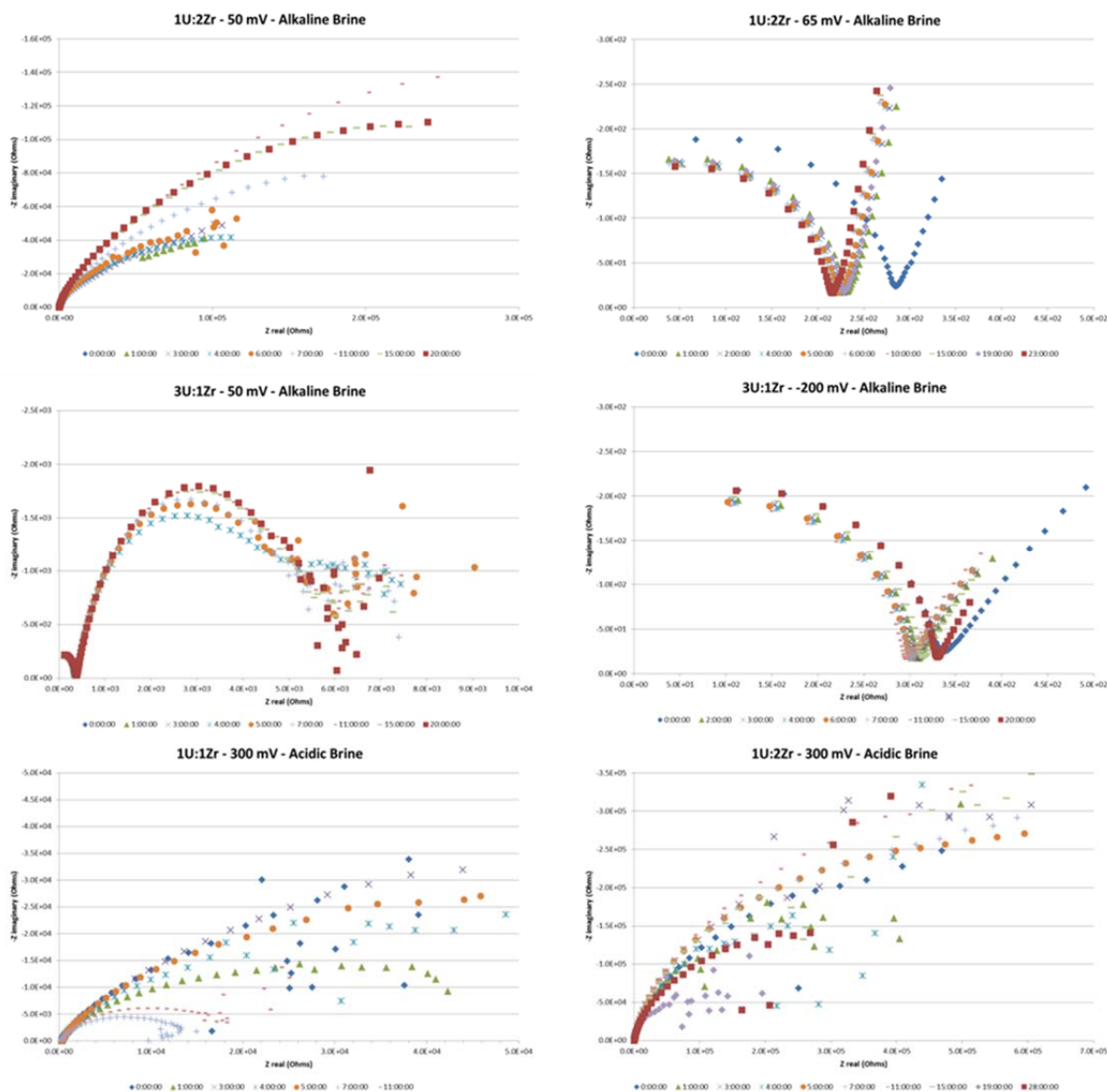


Figure 24 Nyquist plots of the 3 alloy electrodes in acidic brines and alkaline brines at a variety of imposed potentials.

3.6 Post Exposure SEM/EDS Analysis

The two versions of the 1U:2Zr electrode, 1U:1Zr, and 3U:1Zr electrodes were evaluated by SEM/EDS post hybrid testing. Caution is prudent when evaluating this EDS data because the region immediately under the surface of the EDS measurement point may have a drastically different composition not observable via BSE.

3.6.1 1U:2Zr 50 mV Alkaline Brine Post Exposure SEM/EDS Analysis

EDS large area analysis of a relatively oxide free area reported oxygen levels near 38% atomic, Figure 25 and Table 17. This regions chemical composition, when calculated excluding oxygen from the EDS analysis subroutine, gave a similar composition to the as fabricated alloy EDS analysis, Table 17.

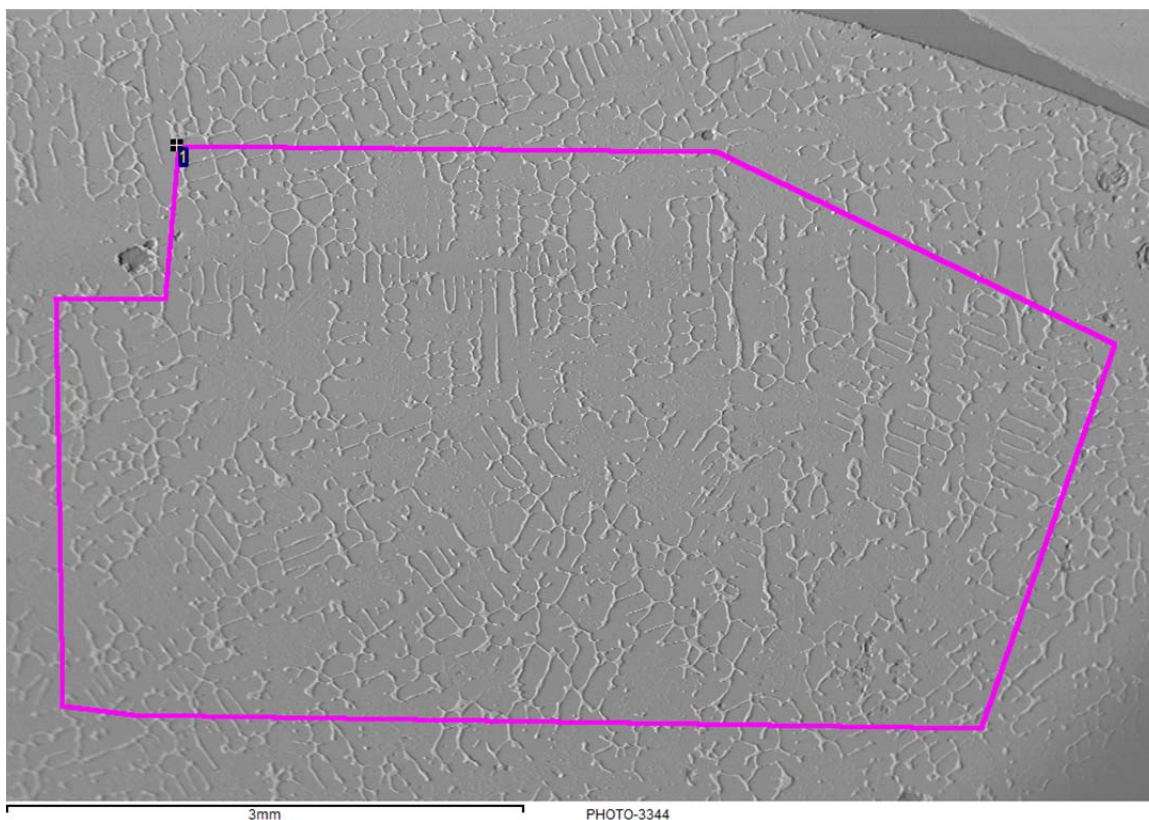


Figure 25 SEM image showing the region from which a large area EDS analysis for the 1U:2Zr alloy in Table 17 was made.

Table 17 Large area EDS scan from the 1U:2Zr electrode showing mean oxygen concentration on the electrode surface. Area of scan corresponds to region within enclosed region in Figure 25.

| With Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|--|---|----|----|----|----|----|----|----|----|----|----|---|
| Area | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |

| | | | | | | | | | | | | |
|---|------|------|-----|------|-----|-----|-----|-----|-----|-----|-----|-----|
| 1 | 38.3 | 9.1 | 0.0 | 48.6 | 0.6 | 1.2 | 0.8 | 0.4 | 0.3 | 0.4 | 0.0 | 0.3 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 14.7 | 0.0 | 77.7 | 1.0 | 2.1 | 1.4 | 0.6 | 0.6 | 0.6 | 0.8 | 0.6 |
| As fabricated EDS data from (Olson 2011) | | | | | | | | | | | | |
| - | - | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| - | - | 11.5 | 0.6 | 76.2 | 0.5 | 2.3 | 1.8 | 1.1 | 1.1 | 1.1 | 1.1 | 1.2 |

To ease analysis, the individual phase EDS data from earlier work (Olson 2011), shown in Figure 1, and Table 3 through Table 7 in section 2.2 is summarized in a single table for the 1U:2Zr alloy and presented in Table 18.

Table 18 1U:2Zr Phase EDS data obtained from reference (Olson 2011).

| 1U:2Zr | | | | | | | | | | | | |
|----------------------------------|------|-----|------|-----|------|-----|-----|-----|------|-----|------|--|
| Phase | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U | |
| Steel | 12.8 | 2.2 | 80.4 | 0.4 | 0.0 | 1.8 | 0.9 | 0.4 | 0.3 | 1.0 | 0.0 | |
| (Zr, U)Fe ₂ | 2.4 | 0.9 | 57.3 | 1.7 | 20.9 | 1.1 | 1.1 | 4.9 | 3.3 | 0.0 | 6.4 | |
| (Zr, U)Pd ₂ | 0.6 | 0.0 | 5.6 | 1.3 | 12.5 | 0.8 | 0.0 | 0.0 | 62.1 | 0.0 | 17.1 | |
| (Zr, U)(Fe, Pd, Rh) ₃ | 2.5 | 0.8 | 51.6 | 3.1 | 8.3 | 0.0 | 0.0 | 8.2 | 7.2 | 0.0 | 18.5 | |
| Zr Rich | 0.6 | 0.0 | 3.5 | 0.1 | 90.7 | 5.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | |

Little degradation of the 1U:2Zr alloy electrode was observed from the 50 mV alkaline brine exposure. Figure 26 and Table 19 show the degradation that was observed. It appeared that the phases responsible for the U sequestration were forming fissures in them, this was predominantly seen in the (Zr, U)Fe₂ phase.

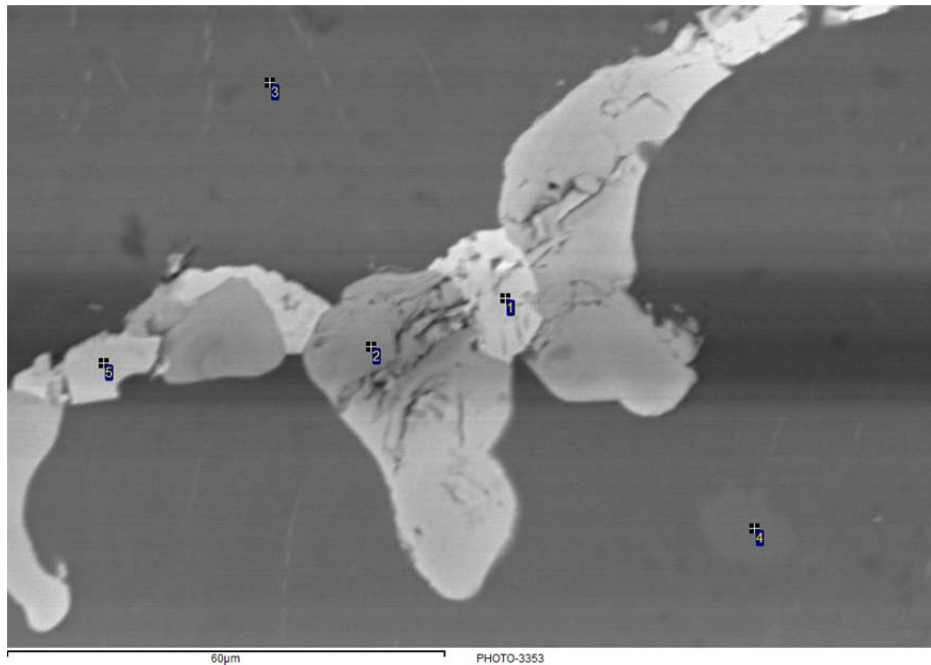


Figure 26 SEM showing regions where EDS point analyses were taken that are summarized in Table 19.

Table 19 EDS analyses of points shown in Figure 26.

| With Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|---|------|------|-----|------|-----|------|-----|-----|------|------|-----|------|
| Area | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | 81.4 | 0.0 | 0.0 | 2.6 | 0.0 | 1.5 | 0.0 | 0.0 | 2.1 | 10.2 | 0.0 | 2.2 |
| 2 | 58.7 | 1.4 | 0.0 | 28.9 | 0.0 | 6.2 | 0.0 | 0.0 | 1.9 | 1.5 | 0.0 | 1.5 |
| 3 | 28.0 | 9.7 | 0.0 | 62.3 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 4 | 31.7 | 9.3 | 0.0 | 58.3 | 0.7 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 5 | 78.0 | 0.0 | 0.0 | 4.1 | 0.0 | 2.5 | 0.0 | 0.0 | 2.2 | 11.0 | 0.0 | 2.1 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 0.0 | 0.0 | 13.5 | 0.0 | 8.1 | 0.0 | 0.0 | 11.3 | 54.8 | 0.0 | 12.2 |
| 2 | - | 3.3 | 0.0 | 68.5 | 0.0 | 15.7 | 0.0 | 0.0 | 4.9 | 3.9 | 0.0 | 3.8 |
| 3 | - | 13.5 | 0.0 | 85.5 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 1.0 | 0.0 |
| 4 | - | 13.6 | 0.0 | 84.3 | 1.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 1.0 | 0.0 |
| 5 | - | 0.0 | 0.0 | 18.2 | 0.0 | 11.5 | 0.0 | 0.0 | 10.1 | 50.4 | 0.0 | 9.8 |

Figure 27 and Table 20 show the (Zr, U)Fe₂ phase degradation over a larger area.

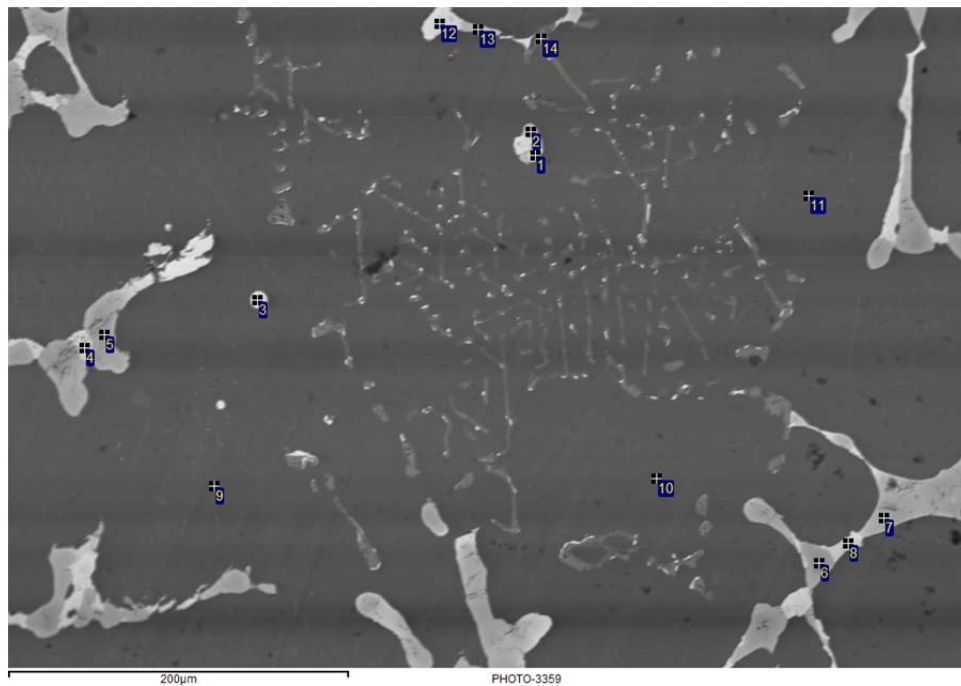


Figure 27 SEM showing regions where EDS point analyses were taken that are summarized in Table 20

Table 20 EDS analyses of points shown in Figure 27.

| With Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|---|------|------|-----|------|-----|------|-----|-----|------|------|-----|------|
| Area | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | 69.0 | 0.4 | 0.0 | 4.0 | 0.3 | 1.9 | 0.0 | 0.7 | 4.2 | 15.3 | 0.0 | 4.1 |
| 2 | 48.4 | 1.4 | 0.0 | 30.7 | 1.2 | 7.7 | 0.0 | 1.3 | 3.2 | 1.0 | 0.0 | 5.3 |
| 3 | 56.0 | 0.9 | 0.0 | 22.7 | 1.4 | 1.8 | 0.0 | 1.2 | 4.1 | 2.9 | 0.0 | 9.0 |
| 4 | 70.2 | 0.0 | 0.0 | 3.0 | 0.4 | 3.1 | 0.3 | 0.4 | 2.9 | 16.3 | 0.0 | 3.4 |
| 5 | 46.5 | 1.7 | 0.0 | 32.8 | 1.1 | 9.5 | 0.0 | 0.9 | 2.5 | 1.4 | 0.0 | 3.5 |
| 6 | 49.7 | 1.5 | 0.0 | 32.4 | 1.2 | 8.5 | 0.0 | 0.8 | 2.3 | 1.5 | 0.0 | 2.2 |
| 7 | 46.1 | 1.5 | 0.0 | 34.3 | 1.2 | 8.1 | 0.0 | 0.9 | 2.6 | 1.0 | 0.0 | 4.3 |
| 8 | 69.2 | 0.4 | 0.0 | 4.5 | 0.4 | 2.7 | 0.0 | 0.5 | 3.2 | 15.9 | 0.0 | 3.1 |
| 9 | 24.1 | 11.5 | 0.0 | 62.8 | 0.6 | 0.0 | 0.9 | 0.2 | 0.0 | 0.0 | 0.0 | 0.0 |
| 10 | 20.6 | 11.8 | 0.0 | 66.1 | 0.6 | 0.0 | 0.9 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 11 | 23.3 | 11.4 | 0.0 | 63.6 | 0.6 | 0.0 | 0.8 | 0.3 | 0.0 | 0.0 | 0.0 | 0.0 |
| 12 | 67.6 | 0.0 | 0.0 | 4.2 | 0.0 | 1.1 | 0.0 | 0.5 | 4.6 | 16.9 | 0.0 | 5.1 |
| 13 | 45.3 | 1.8 | 0.0 | 35.7 | 1.6 | 3.7 | 0.0 | 1.4 | 2.8 | 0.9 | 0.0 | 6.8 |
| 14 | 49.9 | 0.0 | 0.0 | 3.1 | 0.3 | 45.0 | 0.0 | 0.0 | 0.0 | 0.0 | 1.7 | 0.0 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 1.3 | 0.0 | 12.7 | 1.0 | 6.2 | 0.0 | 2.3 | 13.7 | 49.5 | 0.0 | 13.3 |
| 2 | - | 2.6 | 0.0 | 58.3 | 2.2 | 15.2 | 0.0 | 2.6 | 6.3 | 2.1 | 0.3 | 10.5 |
| 3 | - | 2.0 | 0.0 | 50.9 | 3.0 | 4.3 | 0.0 | 2.8 | 9.4 | 6.7 | 0.0 | 20.8 |

| | | | | | | | | | | | | |
|----|---|------|-----|------|-----|------|-----|-----|------|------|-----|------|
| 4 | - | 0.0 | 0.0 | 9.8 | 1.4 | 10.6 | 0.9 | 1.3 | 9.8 | 54.8 | 0.0 | 11.5 |
| 5 | - | 3.2 | 0.0 | 60.1 | 2.1 | 18.2 | 0.0 | 1.7 | 4.9 | 2.8 | 0.3 | 6.8 |
| 6 | - | 3.0 | 0.0 | 63.0 | 2.3 | 17.3 | 0.0 | 1.6 | 4.9 | 3.0 | 0.4 | 4.6 |
| 7 | - | 2.8 | 0.0 | 62.3 | 2.3 | 15.4 | 0.0 | 1.8 | 5.0 | 2.0 | 0.5 | 8.1 |
| 8 | - | 1.5 | 0.0 | 14.2 | 1.3 | 9.0 | 0.0 | 1.7 | 10.3 | 51.9 | 0.0 | 10.2 |
| 9 | - | 15.1 | 0.0 | 82.0 | 0.7 | 0.0 | 1.2 | 0.3 | 0.0 | 0.0 | 0.7 | 0.0 |
| 10 | - | 14.9 | 0.0 | 82.3 | 0.7 | 0.0 | 1.1 | 0.0 | 0.0 | 0.0 | 0.9 | 0.0 |
| 11 | - | 14.8 | 0.0 | 82.2 | 0.8 | 0.0 | 1.0 | 0.5 | 0.0 | 0.0 | 0.7 | 0.0 |
| 12 | - | 0.0 | 0.0 | 12.7 | 0.0 | 3.5 | 0.0 | 1.5 | 14.3 | 52.1 | 0.0 | 15.8 |
| 13 | - | 3.2 | 0.0 | 64.3 | 3.0 | 6.9 | 0.0 | 2.6 | 5.2 | 1.6 | 0.4 | 12.7 |
| 14 | - | 0.0 | 0.0 | 7.2 | 0.7 | 92.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |

3.6.2 1U:2Zr 65 mV Alkaline Brine Post Exposure SEM/EDS Analysis

EDS large area analysis of a relatively oxide free area reported oxygen levels near 27% atomic, Figure 28 and Table 21. This regions chemical composition, when calculated excluding oxygen from the EDS analysis subroutine, gave a similar composition to the as fabricated alloy EDS analysis although Rh and Pd were not detected, Table 21.

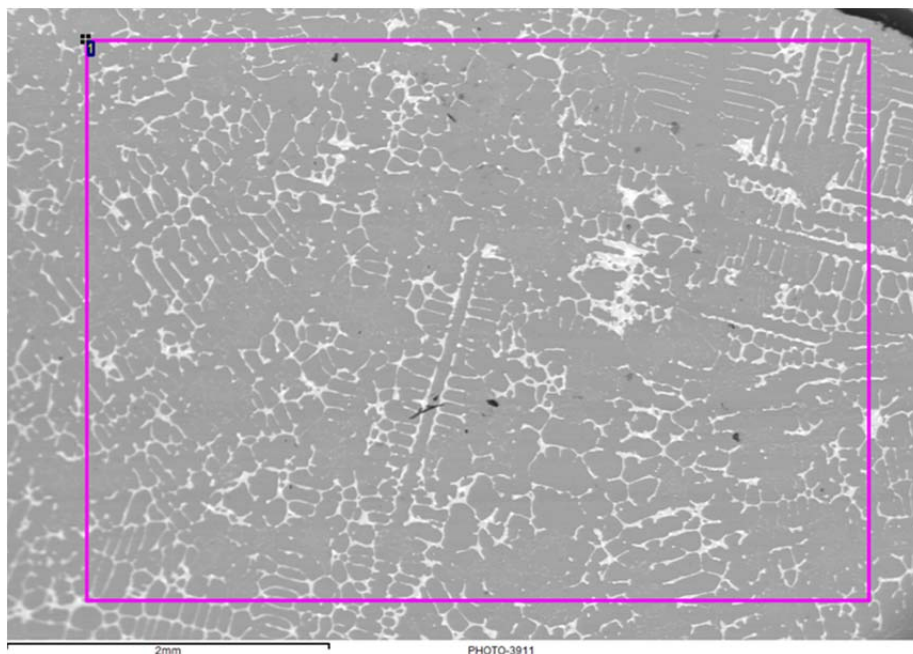


Figure 28 SEM image showing the region from which a large area EDS analysis for the 1U:2Zr alloy in Table 21 was made.

Table 21 Large area EDS scan from the 1U:2Zr electrode showing mean oxygen concentration on the electrode surface. Area of scan corresponds to region within enclosed region in Figure 28.

| |
|--|
| With Oxygen in Scan - All results in atomic% |
|--|

| Area | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
|---|------|------|-----|------|-----|-----|-----|-----|-----|-----|-----|-----|
| 1 | 27.0 | 8.0 | 2.2 | 57.3 | 0.5 | 2.1 | 1.7 | 0.4 | 0.0 | 0.0 | 0.0 | 0.9 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 11.0 | 2.9 | 77.1 | 0.7 | 3.0 | 2.5 | 0.5 | 0.0 | 0.0 | 1.1 | 1.2 |
| As fabricated EDS data from (Olson 2011) | | | | | | | | | | | | |
| - | - | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| - | - | 11.5 | 0.6 | 76.2 | 0.5 | 2.3 | 1.8 | 1.1 | 1.1 | 1.1 | 1.1 | 1.2 |

Refer to Table 18 for specific phase compositional analysis on the uncorroded base alloy.

Little degradation of the 1U:2Zr alloy electrode was observed from the 50 mV alkaline brine exposure. Figure 29 shows the degradation that was observed. It appeared that the phases responsible for the U sequestration were forming pits, this was predominantly seen in the (Zr,U)Fe₂ phase. The pitting was not widely observed in the alloy, and was more readily spotted in secondary electron imaging mode, which is more topographically sensitive.

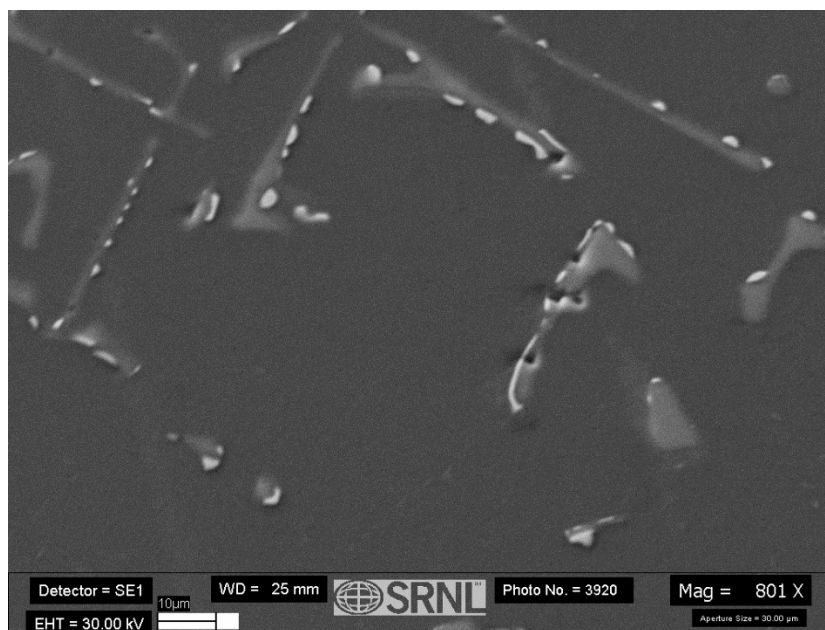


Figure 29 SEM showing regions with apparent pitting.

Figure 30 and Table 22 show the pitting in the (Zr, U)Fe₂ phase was not readily evident over a large area. It is observed that in general, the uranium bearing phases have higher oxygen concentrations associated with them, which is also the case for EDS analyses in Table 19, Table 20, and Table 26, but not the thick oxide films observed in Figure 32.

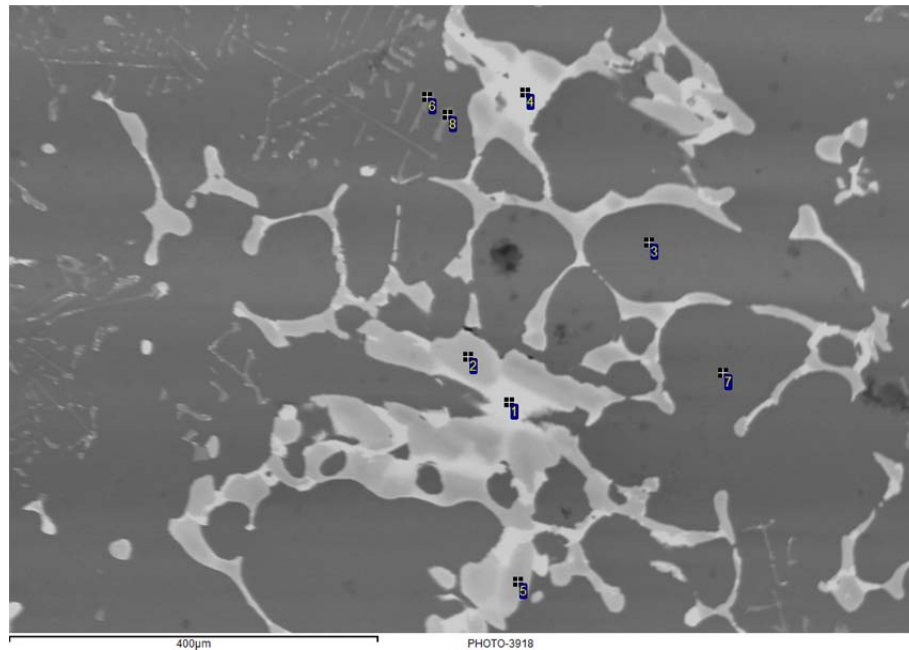


Figure 30 SEM showing regions where EDS point analyses were taken that are summarized in Table 22.

Table 22 EDS analyses of points shown in Figure 30.

| With Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|---|------|------|-----|------|-----|------|-----|-----|-----|-----|-----|------|
| Area | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | 40.9 | 0.0 | 1.0 | 26.3 | 1.8 | 2.4 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 27.6 |
| 2 | 31.5 | 1.2 | 1.1 | 35.9 | 1.0 | 15.5 | 1.3 | 0.0 | 1.2 | 0.9 | 0.0 | 10.4 |
| 3 | 32.5 | 7.6 | 1.9 | 55.1 | 0.6 | 0.0 | 1.3 | 0.7 | 0.0 | 0.0 | 0.4 | 0.0 |
| 4 | 44.1 | 0.0 | 0.6 | 23.6 | 1.3 | 2.2 | 0.0 | 1.3 | 2.7 | 0.0 | 0.0 | 24.1 |
| 5 | 31.4 | 1.3 | 1.0 | 35.8 | 1.6 | 16.6 | 1.4 | 0.0 | 1.5 | 0.0 | 0.0 | 9.3 |
| 6 | 34.6 | 0.3 | 0.0 | 1.5 | 0.0 | 59.5 | 4.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 7 | 21.0 | 8.9 | 2.3 | 64.2 | 0.5 | 0.0 | 1.6 | 0.6 | 0.0 | 0.0 | 1.1 | 0.0 |
| 8 | 32.4 | 0.0 | 0.0 | 1.9 | 0.3 | 60.9 | 4.5 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 0.0 | 1.7 | 44.5 | 3.0 | 4.2 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 46.7 |
| 2 | - | 1.7 | 1.6 | 51.4 | 1.4 | 22.9 | 1.9 | 0.0 | 1.8 | 1.4 | 0.7 | 15.3 |
| 3 | - | 11.2 | 2.7 | 80.9 | 0.8 | 0.0 | 2.1 | 1.0 | 0.0 | 0.0 | 1.2 | 0.0 |
| 4 | - | 0.0 | 1.1 | 42.0 | 2.3 | 4.1 | 0.0 | 2.3 | 4.9 | 0.0 | 0.0 | 43.3 |
| 5 | - | 1.9 | 1.5 | 51.2 | 2.3 | 24.6 | 2.1 | 0.0 | 2.2 | 0.0 | 0.7 | 13.7 |
| 6 | - | 0.4 | 0.0 | 2.3 | 0.0 | 90.6 | 6.6 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 7 | - | 11.2 | 2.8 | 81.1 | 0.7 | 0.0 | 2.0 | 0.8 | 0.0 | 0.0 | 1.4 | 0.0 |

| | | | | | | | | | | | | |
|---|---|-----|-----|-----|-----|------|-----|-----|-----|-----|-----|-----|
| 8 | - | 0.0 | 0.0 | 2.8 | 0.5 | 89.8 | 6.9 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
|---|---|-----|-----|-----|-----|------|-----|-----|-----|-----|-----|-----|

3.6.3 3U:1Zr 50 mV Alkaline Brine Post Exposure SEM/EDS Analysis

Figure 31 and corresponding Table 23 show regions of large area EDS scans and the EDS data, respectively. Even when a relatively thick oxide layer is present, the observed EDS spectrum did not vary much from the less oxidized surfaces. Compared to the as fabricated EDS, the oxidized areas appeared enriched in Fe, Ni, and Zr at the expense of Cr and the other components. It appears that the high-Z phases are getting partially covered in oxides in the boxed region on the right of Figure 31, compared to the boxed region on the left. The oxides covering primarily the high-Z phases compared to the base SS phase may indicate microgalvanic electrolytic reactions occurring on the electrode surface.

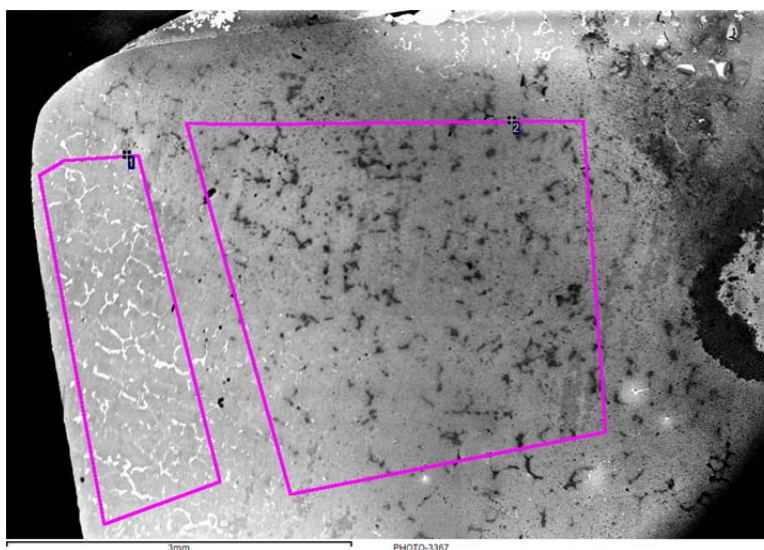


Figure 31 SEM images showing the areas from which large area EDS scans were made, corresponding to Table 23.

Table 23 Several large area EDS scans from the 3U:1Zr electrode showing mean oxygen concentrations on the surface. Area numbers correspond to numbers in Figure 31.

| With Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|---|------|------|-----|------|-----|-----|-----|-----|-----|-----|-----|-----|
| Area | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | 47.8 | 7.5 | 0.0 | 41.8 | 0.5 | 0.4 | 0.7 | 0.4 | 0.3 | 0.3 | 0.3 | 0.3 |
| 2 | 53.9 | 5.9 | 0.0 | 38.5 | 0.4 | 0.2 | 0.5 | 0.3 | 0.1 | 0.1 | 0.0 | 0.1 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 14.2 | 0.0 | 79.7 | 0.9 | 0.8 | 1.4 | 0.7 | 0.6 | 0.6 | 0.6 | 0.5 |
| 2 | - | 12.7 | 0.0 | 82.7 | 1.0 | 0.5 | 1.1 | 0.7 | 0.3 | 0.3 | 0.6 | 0.3 |
| As fabricated EDS data from (Olson 2012) | | | | | | | | | | | | |
| - | - | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |

| | | | | | | | | | | | | |
|---|---|------|-----|------|-----|-----|-----|-----|-----|-----|-----|-----|
| - | - | 11.8 | 0.7 | 77.7 | 0.5 | 0.4 | 1.8 | 1.1 | 1.1 | 1.1 | 1.1 | 1.2 |
|---|---|------|-----|------|-----|-----|-----|-----|-----|-----|-----|-----|

The as fabricated phase data from an earlier report (Olson 2012), is shown in Table 24 for quick reference and comparison to the EDS point scans in this section.

Table 24 Phase composition data from EDS point analysis for the 3U:1Zr alloy electrode.

| 3U:1Zr | | | | | | | | | | | |
|----------------------------------|--------------|-----|------|-----|------|-----|-----|------|------|-----|------|
| Phase | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| Steel | Not analyzed | | | | | | | | | | |
| (Zr, U)Fe ₂ | Not detected | | | | | | | | | | |
| (Zr, U)Pd ₂ | 1.0 | 0.0 | 3.8 | 1.1 | 0.4 | 0.0 | 0.0 | 0.0 | 66.8 | 0.0 | 26.8 |
| (Zr, U)(Fe, Pd, Rh) ₃ | 2.6 | 0.4 | 46.8 | 2.8 | 0.0 | 0.0 | 3.5 | 12.2 | 7.2 | 0.0 | 24.5 |
| Zr Rich | 0.4 | 0.0 | 2.8 | 0.0 | 88.7 | 5.7 | 0.0 | 0.0 | 0.0 | 0.0 | 2.3 |

Figure 32 and Table 25 show EDS area analyses on a thick oxide near a region where some of the oxide scale has cracked and revealed the relatively clean surface. Based on the EDS point scans, the scale appears to be primarily iron-chrome oxide and/or possibly iron-chrome hydroxides.

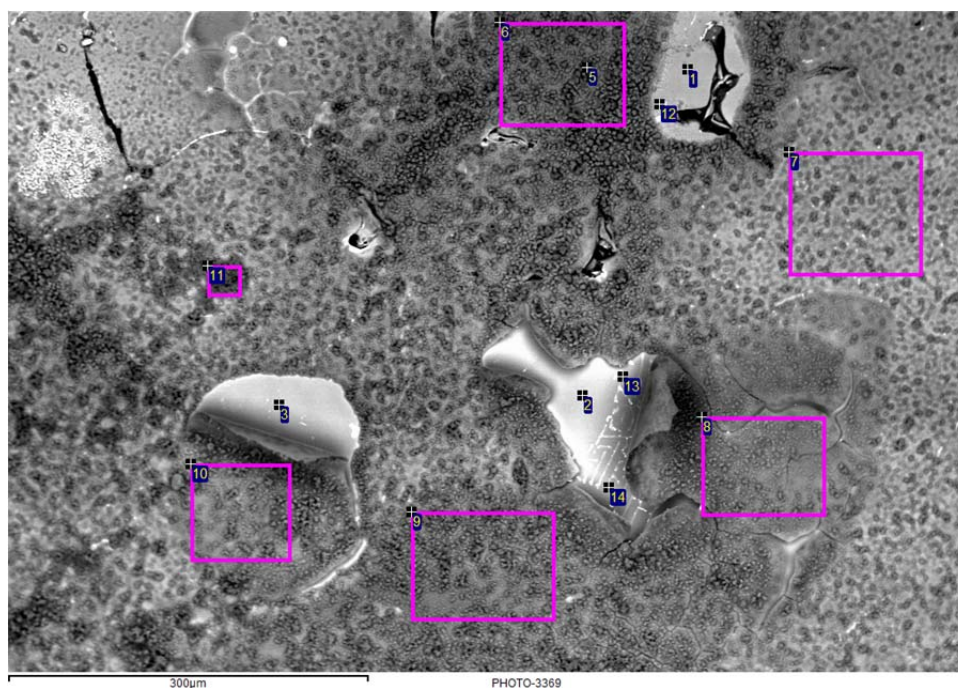


Figure 32 SEM images with point's numbers marking points where EDS point analysis was performed.

Table 25 EDS point analysis of the points in Figure 32.

| With Oxygen in Scan - All results in atomic% |
|--|
|--|

| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
|---|------|------|-----|------|-----|------|-----|-----|-----|------|-----|-----|
| 1 | 35.1 | 9.8 | 0.0 | 52.3 | 0.9 | 0.0 | 0.7 | 0.0 | 0.4 | 0.9 | 0.0 | 0.0 |
| 2 | 15.3 | 11.5 | 0.0 | 71.7 | 0.9 | 0.0 | 0.6 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 3 | 31.5 | 10.8 | 0.0 | 56.5 | 0.5 | 0.0 | 0.7 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 4 | 0.0 | 24.7 | 0.0 | 75.3 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 5 | 34.6 | 20.1 | 0.0 | 43.2 | 0.4 | 0.0 | 1.7 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 6 | 40.9 | 12.5 | 0.0 | 44.6 | 0.5 | 0.0 | 1.2 | 0.0 | 0.0 | 0.3 | 0.0 | 0.0 |
| 7 | 36.3 | 10.1 | 0.0 | 52.3 | 0.6 | 0.0 | 0.7 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 8 | 37.4 | 8.8 | 0.0 | 52.8 | 0.5 | 0.0 | 0.6 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 9 | 49.9 | 7.9 | 0.0 | 40.8 | 0.3 | 0.4 | 0.5 | 0.3 | 0.0 | 0.0 | 0.0 | 0.0 |
| 10 | 48.3 | 8.7 | 0.0 | 41.5 | 0.4 | 0.0 | 0.3 | 0.2 | 0.0 | 0.3 | 0.0 | 0.2 |
| 11 | 49.2 | 11.8 | 0.0 | 37.7 | 0.4 | 0.0 | 0.4 | 0.3 | 0.0 | 0.0 | 0.0 | 0.1 |
| 12 | 21.3 | 10.4 | 0.0 | 66.7 | 0.8 | 0.0 | 0.8 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 13 | 47.7 | 3.6 | 0.0 | 18.6 | 0.0 | 25.4 | 0.0 | 0.0 | 0.7 | 1.6 | 1.2 | 1.3 |
| 14 | 45.5 | 3.8 | 0.0 | 16.8 | 0.0 | 20.3 | 0.0 | 0.6 | 2.9 | 6.4 | 0.7 | 3.1 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 15.1 | 0.0 | 79.8 | 1.3 | 0.0 | 1.1 | 0.0 | 0.7 | 1.4 | 0.7 | 0.0 |
| 2 | - | 13.7 | 0.0 | 83.8 | 1.0 | 0.0 | 0.8 | 0.0 | 0.0 | 0.0 | 0.7 | 0.0 |
| 3 | - | 15.8 | 0.0 | 81.5 | 0.7 | 0.0 | 1.1 | 0.0 | 0.0 | 0.0 | 0.9 | 0.0 |
| 4 | - | 24.7 | 0.0 | 75.3 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 5 | - | 30.5 | 0.0 | 65.9 | 0.6 | 0.0 | 2.8 | 0.0 | 0.0 | 0.0 | 0.4 | 0.0 |
| 6 | - | 21.0 | 0.0 | 74.9 | 0.9 | 0.0 | 2.1 | 0.0 | 0.0 | 0.6 | 0.7 | 0.0 |
| 7 | - | 15.7 | 0.0 | 81.6 | 0.9 | 0.0 | 1.1 | 0.0 | 0.0 | 0.0 | 0.6 | 0.0 |
| 8 | - | 14.0 | 0.0 | 83.6 | 0.8 | 0.0 | 1.0 | 0.0 | 0.0 | 0.0 | 0.7 | 0.0 |
| 9 | - | 15.6 | 0.0 | 80.7 | 0.7 | 0.8 | 1.0 | 0.6 | 0.0 | 0.0 | 0.6 | 0.0 |
| 10 | - | 16.7 | 0.0 | 79.5 | 0.7 | 0.0 | 0.7 | 0.5 | 0.0 | 0.7 | 0.9 | 0.4 |
| 11 | - | 22.9 | 0.0 | 73.9 | 0.8 | 0.0 | 0.9 | 0.6 | 0.0 | 0.0 | 0.6 | 0.3 |
| 12 | - | 13.3 | 0.0 | 84.0 | 1.1 | 0.0 | 1.0 | 0.0 | 0.0 | 0.0 | 0.7 | 0.0 |
| 13 | - | 7.3 | 0.0 | 37.0 | 0.0 | 48.4 | 0.0 | 0.0 | 1.5 | 3.3 | 0.0 | 2.6 |
| 14 | - | 7.1 | 0.0 | 31.3 | 0.0 | 37.0 | 0.0 | 1.2 | 5.5 | 12.2 | 0.0 | 5.9 |

Figure 33 shows a region on the 3U:1Zr RAW-3 alloy where the oxide layer appeared to have cracked and partially peeled off. It can be observed that the relatively lighter gray area, analyzed by EDS and designated by numbers 1 and 5, appears likely to have been selectively corroded. Table 26 presents the corresponding EDS point scans.

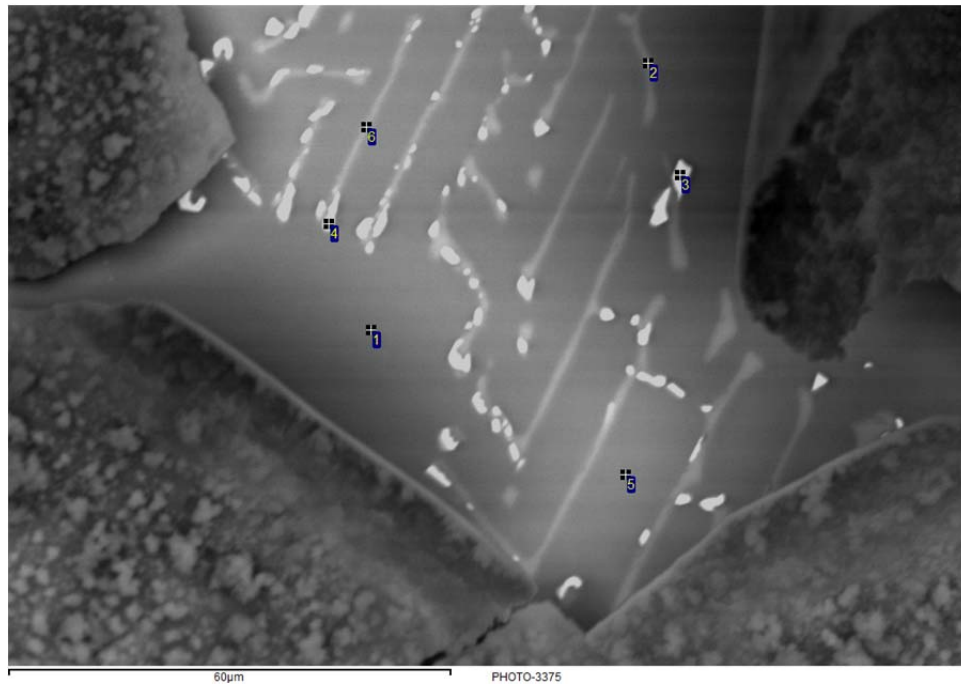


Figure 33 Magnification of area which was under an oxidized region.

Table 26 EDS point analysis of the points in Figure 33.

| With Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|---|------|------|-----|------|-----|------|-----|-----|-----|------|-----|-----|
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | 35.8 | 9.3 | 0.0 | 52.3 | 0.8 | 0.0 | 0.9 | 0.3 | 0.3 | 0.0 | 0.3 | 0.0 |
| 2 | 52.8 | 5.0 | 0.0 | 22.1 | 0.2 | 18.5 | 1.0 | 0.0 | 0.0 | 0.0 | 0.3 | 0.3 |
| 3 | 63.3 | 3.9 | 0.0 | 17.2 | 0.3 | 0.0 | 0.4 | 0.5 | 2.9 | 8.4 | 0.0 | 3.1 |
| 4 | 58.4 | 4.9 | 0.1 | 26.3 | 0.5 | 0.5 | 0.4 | 0.2 | 1.8 | 4.9 | 0.2 | 1.8 |
| 5 | 36.0 | 10.0 | 0.0 | 52.2 | 0.7 | 0.0 | 1.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 6 | 43.9 | 7.8 | 0.0 | 38.2 | 0.3 | 8.3 | 1.1 | 0.0 | 0.0 | 0.0 | 0.4 | 0.0 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 14.4 | 0.0 | 81.4 | 1.2 | 0.0 | 1.4 | 0.5 | 0.5 | 0.0 | 0.5 | 0.0 |
| 2 | - | 10.5 | 0.0 | 46.3 | 0.4 | 39.5 | 2.2 | 0.0 | 0.0 | 0.0 | 0.3 | 0.7 |
| 3 | - | 10.6 | 0.0 | 45.9 | 0.7 | 0.0 | 1.2 | 1.5 | 7.9 | 23.1 | 0.4 | 8.7 |
| 4 | - | 11.8 | 0.3 | 62.5 | 1.1 | 1.3 | 0.9 | 0.6 | 4.5 | 12.1 | 0.5 | 4.4 |
| 5 | - | 15.6 | 0.0 | 80.8 | 1.0 | 0.0 | 1.9 | 0.0 | 0.0 | 0.0 | 0.8 | 0.0 |
| 6 | - | 13.7 | 0.0 | 67.5 | 0.6 | 15.4 | 2.1 | 0.0 | 0.0 | 0.0 | 0.6 | 0.0 |

3.6.4 3U:1Zr -200 mV Alkaline Brine Post Exposure SEM/EDS Analysis

Figure 34 and corresponding Table 27 show regions of large area EDS scans and the EDS data, respectively. The observed EDS spectrum did not vary much from the pre-exposed surfaces. This alloy did have large oxide deposits.

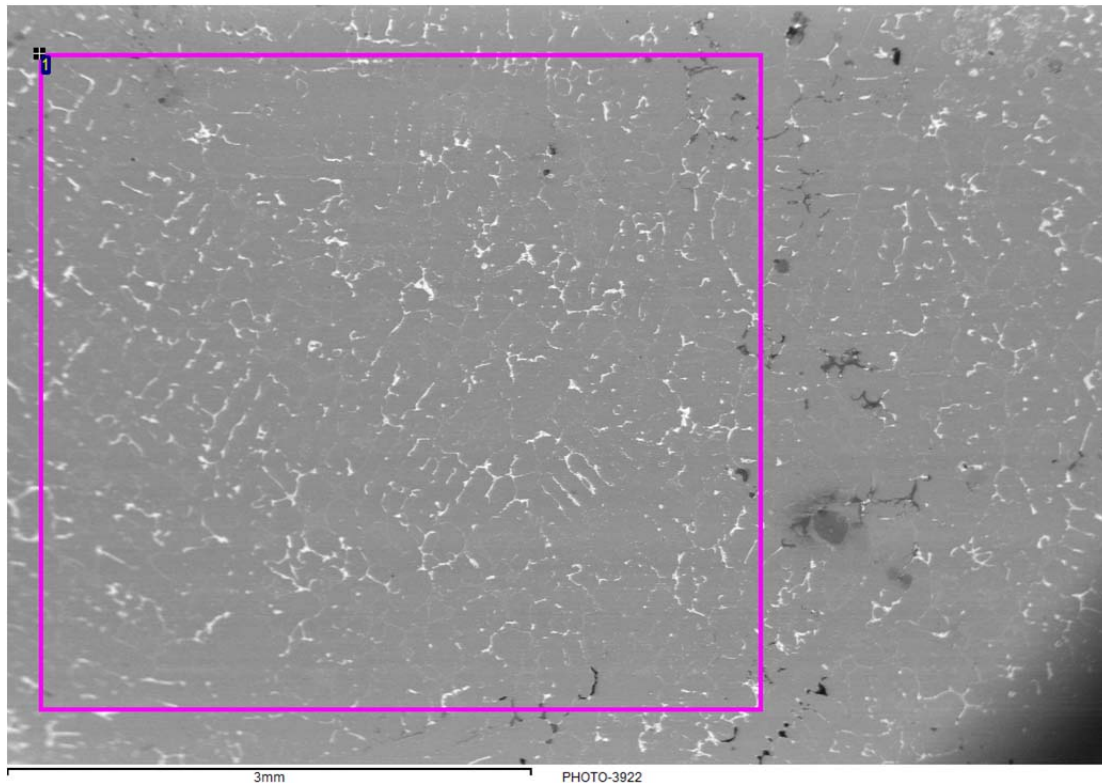


Figure 34 SEM images showing the areas from which large area EDS scans were made, corresponding to Table 27.

Table 27 Several large area EDS scans from the 3U:1Zr electrode showing mean oxygen concentrations on the surface. Area numbers correspond to numbers in Figure 34.

| With Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|---|------|------|-----|------|-----|-----|-----|-----|-----|-----|-----|-----|
| Area | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | 31.2 | 7.4 | 1.8 | 54.7 | 0.6 | 0.8 | 1.7 | 0.4 | 0.0 | 0.3 | 0.6 | 0.5 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 10.7 | 2.7 | 79.3 | 0.8 | 1.3 | 2.6 | 0.7 | 0.0 | 0.5 | 0.9 | 0.7 |
| As fabricated EDS data from (Olson 2012) | | | | | | | | | | | | |
| - | - | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| - | - | 11.8 | 0.7 | 77.7 | 0.5 | 0.4 | 1.8 | 1.1 | 1.1 | 1.1 | 1.1 | 1.2 |

Refer to Table 24 for specific phase compositional analysis on the uncorroded base alloy.

1mm

PHOTO-3932

Table 28 EDS point analysis of the points in Figure 35.

| With Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|---|------|------|-----|------|-----|-----|-----|-----|-----|------|-----|------|
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | 29.2 | 7.4 | 2.2 | 58.9 | 0.4 | 0.0 | 0.9 | 0.0 | 0.0 | 0.0 | 1.1 | 0.0 |
| 2 | 28.8 | 7.3 | 1.9 | 59.2 | 0.9 | 0.0 | 1.1 | 0.0 | 0.0 | 0.0 | 0.9 | 0.0 |
| 3 | 37.8 | 7.3 | 1.5 | 50.2 | 0.5 | 0.0 | 1.3 | 0.8 | 0.0 | 0.0 | 0.8 | 0.0 |
| 4 | 70.5 | 12.5 | 1.3 | 11.4 | 0.7 | 0.0 | 0.0 | 1.3 | 0.0 | 0.0 | 1.0 | 1.1 |
| 5 | 30.1 | 7.1 | 1.9 | 58.3 | 0.4 | 0.0 | 1.2 | 0.0 | 0.0 | 0.0 | 1.1 | 0.0 |
| 6 | 78.8 | 12.4 | 1.1 | 4.2 | 0.9 | 0.0 | 0.0 | 0.9 | 0.0 | 0.0 | 0.8 | 1.0 |
| 7 | 65.3 | 1.4 | 1.0 | 27.6 | 1.0 | 0.0 | 0.0 | 2.6 | 0.0 | 0.6 | 0.5 | 0.0 |
| 8 | 68.4 | 0.0 | 0.0 | 2.2 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 10.2 | 0.0 | 19.2 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |

| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
|-------|---|------|-----|------|-----|-----|-----|-----|-----|------|-----|------|
| 1 | - | 10.4 | 3.1 | 83.5 | 0.5 | 0.0 | 1.4 | 0.0 | 0.0 | 0.0 | 1.1 | 0.0 |
| 2 | - | 10.2 | 2.7 | 82.9 | 1.2 | 0.0 | 1.6 | 0.0 | 0.0 | 0.0 | 1.5 | 0.0 |
| 3 | - | 11.6 | 2.3 | 80.2 | 0.8 | 0.0 | 2.2 | 1.3 | 0.0 | 0.0 | 1.6 | 0.0 |
| 4 | - | 41.3 | 4.3 | 37.5 | 2.4 | 0.0 | 0.0 | 4.8 | 0.0 | 0.0 | 5.6 | 4.1 |
| 5 | - | 10.0 | 2.7 | 83.9 | 0.7 | 0.0 | 1.8 | 0.0 | 0.0 | 0.0 | 0.9 | 0.0 |
| 6 | - | 56.7 | 4.8 | 19.6 | 4.1 | 0.0 | 0.0 | 4.7 | 0.0 | 0.0 | 5.0 | 5.1 |
| 7 | - | 4.0 | 2.7 | 76.1 | 2.8 | 0.0 | 0.0 | 8.2 | 0.0 | 2.1 | 4.1 | 0.0 |
| 8 | - | 0.0 | 0.0 | 7.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 31.1 | 0.0 | 61.8 |

Figure 36 shows the same region shown in Figure 35 , but displays the backscatter image next to the secondary electron image. The backscatter image better shows the elemental differences, with higher Z elements appearing brighter, whereas the secondary electron image better shows the topographic information such as the pitting associated with localized corrosion at the phases interfaces.

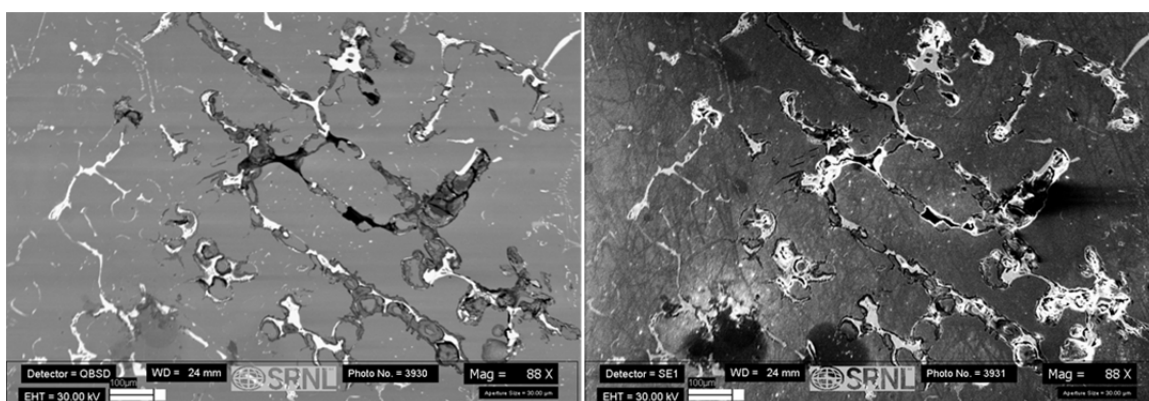


Figure 36 Back Scatter electron image (left) and secondary electron image (right) showing compositional and topographical effects of localized corrosion.

3.6.5 1U:1Zr 300 mV Acidic Brine Potentiostatic Hold Hybrid Test SEM/EDS

Figure 37 shows the depth of localized attack on the edge of the 1U:1Zr alloy electrode after the short between the RE and WE probably reached at least 1 mm. The short between the two electrodes led to relatively large amounts of current flowing into the alloy electrode that led to removal of significant alloy in the electrode and destruction of the alloy surface.

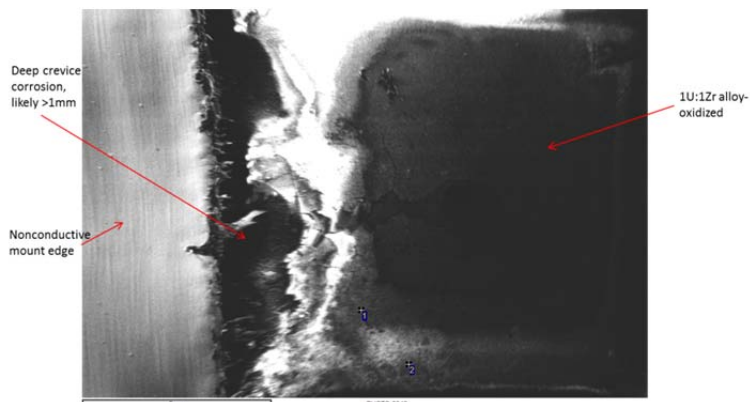


Figure 37 SEM image showing the depth of localized corrosion on the edge of the 1U:1Zr alloy reached at least 1 mm, likely due to a short that developed between the RE and the WE.

Figure 38 and Figure 39 show the compositional and topographic nature of the corrosion and oxides present on the electrode. It appears that either under the -200 mV imposed potential, or more likely the imposed current during the WE-RE short, that the steel phase was preferentially degraded, leaving a web of the other phases, primarily the (Zr, U)Pd₂ and the (Zr, U)Fe₂ phases.

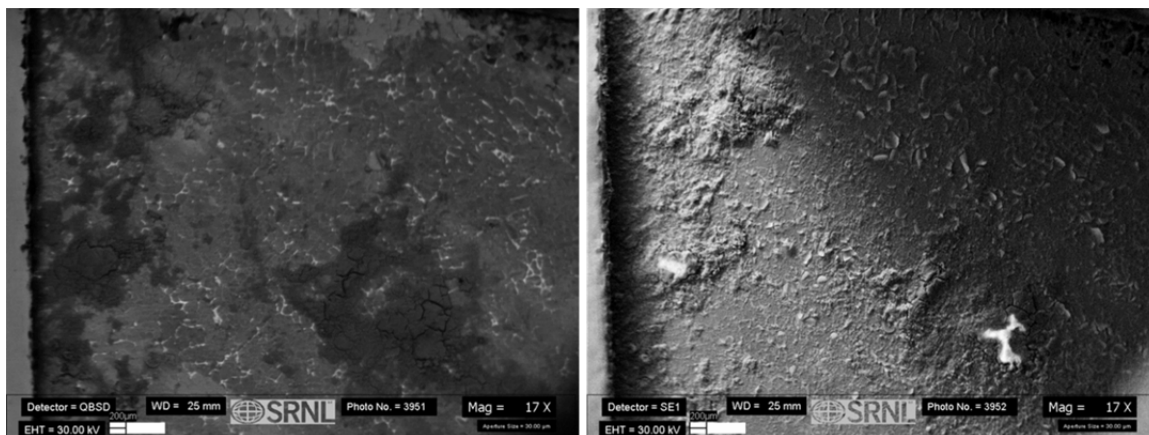


Figure 38 Back Scatter electron image (left) and secondary electron image (right) showing compositional and topographical effects of localized corrosion.

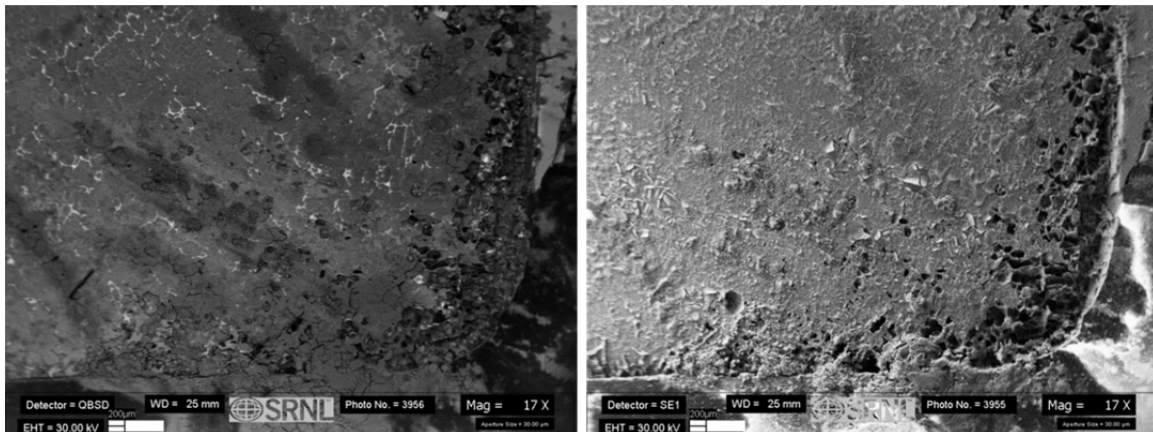


Figure 39 Back Scatter electron image (left) and secondary electron image (right) showing compositional and topographical effects of localized corrosion.

Figure 40 and Table 29 show EDS large area analysis, two smaller region analyses of oxides, and several point analyses of the 1U:2Zr electrode after the 65 mV potentiostatic hold hybrid test. Ni, Zr, Rh, and Pd are not detected, and Mo appears to be enriched in the largest area scan (analysis region 8).

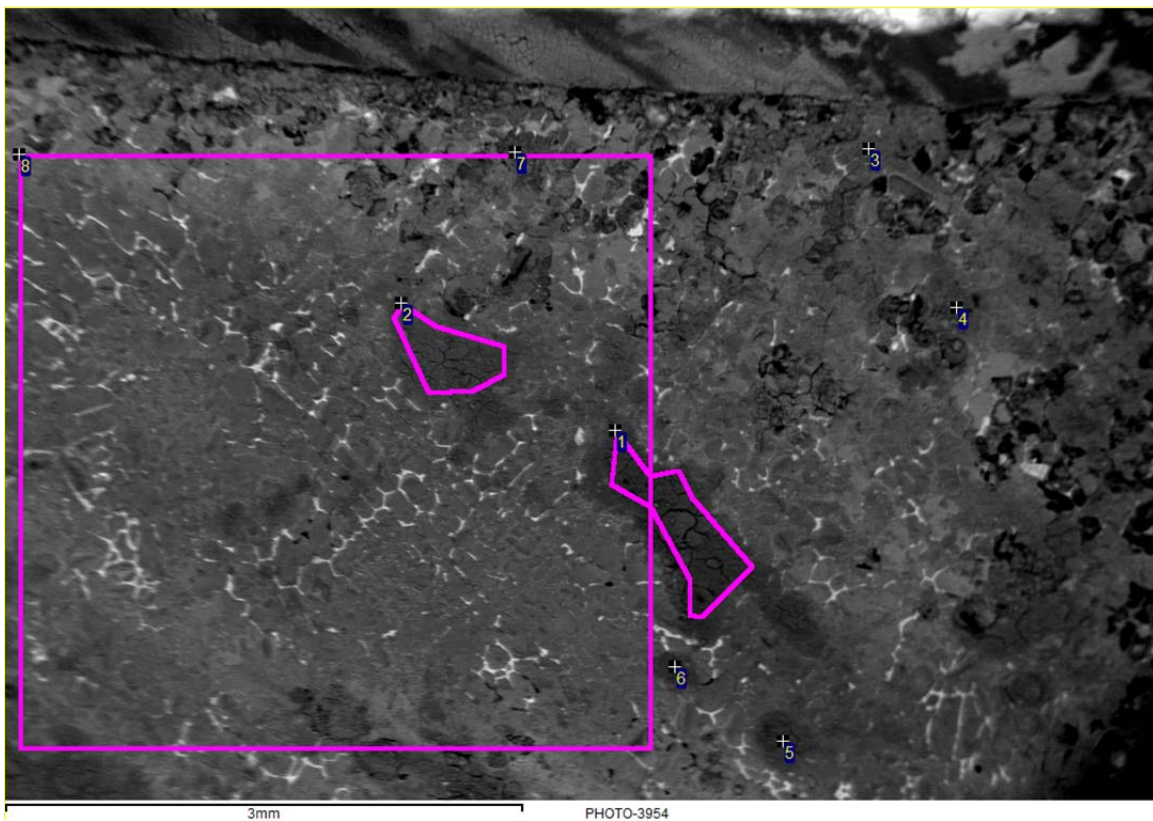


Figure 40 Large area EDS scans of several locations on the 1U:1Zr alloy after the 65 mV potentiostatic hold hybrid test.

Table 29 Several large area EDS scans from the 1U:1Zr electrode showing mean oxygen concentrations on the surface. Area numbers correspond to numbers in Figure 40.

| With Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|---|------|------|-----|------|-----|-----|------|-----|-----|-----|-----|-----|
| Area | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | 77.3 | 3.7 | 0.7 | 15.4 | 0.0 | 0.0 | 2.3 | 0.6 | 0.0 | 0.0 | 0.0 | 0.0 |
| 2 | 75.6 | 4.2 | 0.8 | 16.4 | 0.0 | 0.0 | 3.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 3 | 80.7 | 3.0 | 0.6 | 13.7 | 0.0 | 0.0 | 1.5 | 0.5 | 0.0 | 0.0 | 0.0 | 0.0 |
| 4 | 76.5 | 3.2 | 0.9 | 17.5 | 0.0 | 0.0 | 1.9 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 5 | 73.8 | 4.5 | 1.0 | 17.4 | 0.0 | 0.0 | 2.7 | 0.6 | 0.0 | 0.0 | 0.0 | 0.0 |
| 6 | 71.7 | 5.8 | 1.0 | 18.2 | 0.0 | 0.0 | 2.8 | 0.6 | 0.0 | 0.0 | 0.0 | 0.0 |
| 7 | 76.9 | 2.1 | 0.7 | 17.5 | 0.0 | 0.0 | 1.6 | 0.8 | 0.0 | 0.0 | 0.5 | 0.0 |
| 8 | 69.6 | 3.5 | 1.0 | 23.5 | 0.0 | 0.0 | 1.9 | 0.4 | 0.0 | 0.0 | 0.0 | 0.2 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 15.9 | 2.9 | 65.8 | 0.0 | 0.0 | 11.2 | 3.3 | 0.0 | 0.0 | 1.0 | 0.0 |
| 2 | - | 16.6 | 3.4 | 66.4 | 0.0 | 0.0 | 13.7 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 3 | - | 15.0 | 2.8 | 67.7 | 0.0 | 0.0 | 9.1 | 3.4 | 0.0 | 0.0 | 2.1 | 0.0 |
| 4 | - | 13.0 | 3.8 | 70.9 | 0.0 | 0.0 | 9.5 | 0.0 | 0.0 | 0.0 | 2.9 | 0.0 |
| 5 | - | 16.7 | 3.7 | 64.5 | 0.0 | 0.0 | 11.3 | 2.8 | 0.0 | 0.0 | 1.0 | 0.0 |
| 6 | - | 19.8 | 3.3 | 61.8 | 0.0 | 0.0 | 11.1 | 2.3 | 0.0 | 0.0 | 1.9 | 0.0 |
| 7 | - | 8.7 | 2.8 | 74.1 | 0.0 | 0.0 | 7.7 | 4.1 | 0.0 | 0.0 | 2.6 | 0.0 |
| 8 | - | 11.2 | 3.0 | 74.8 | 0.0 | 0.0 | 7.1 | 1.6 | 0.0 | 0.0 | 1.5 | 0.9 |
| As fabricated EDS data from (Olson 2012) | | | | | | | | | | | | |
| - | - | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| - | - | 12.0 | 2.3 | 78.8 | 0.5 | 2.8 | 1.8 | 0.6 | 0.2 | 0.5 | 0.0 | 0.7 |

The as fabricated phase data from an earlier report (Olson 2012), is shown in Table 30 for quick reference and comparison to the EDS point scans in this section.

Table 30 Phase data taken from a previous report to aid in discerning oxidized phases.

| 1U:1Zr | | | | | | | | | | | | |
|----------------------------------|------|-----|------|-----|------|-----|-----|------|------|-----|------|--|
| Phase | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U | |
| Steel | 12.2 | 2.3 | 80.4 | 0.4 | 0.0 | 1.8 | 1.2 | 0.5 | 0.3 | 0.9 | 0.0 | |
| (Zr, U)Fe ₂ | 2.2 | 1.0 | 55.3 | 1.7 | 19.9 | 1.4 | 1.1 | 7.2 | 2.9 | 0.0 | 7.5 | |
| (Zr, U)Pd ₂ | 0.0 | 0.0 | 4.9 | 1.8 | 12.5 | 0.9 | 0.0 | 0.0 | 61.1 | 0.0 | 18.8 | |
| (Zr, U)(Fe, Pd, Rh) ₃ | 1.9 | 0.6 | 39.2 | 3.8 | 10.2 | 0.0 | 0.0 | 12.7 | 11.7 | 0.0 | 19.8 | |

| Zr Rich | 1.2 | 0.1 | 6.3 | 0.1 | 87.3 | 4.8 | 0.0 | 0.0 | 0.0 | 0.0 | 0.2 |
|---------|-----|-----|-----|-----|------|-----|-----|-----|-----|-----|-----|
|---------|-----|-----|-----|-----|------|-----|-----|-----|-----|-----|-----|

Figure 41, Figure 42 and Table 31 show a region that has evidence of corrosion and oxides forming on the steel phases, with either the oxides not forming on the high Z phases, or the oxides that do form peeling off above them. Figure 42, which shows more of the surface relief, shows that the effect on the oxide scale of the high Z compositions may not be evident without back scattered electron imaging.

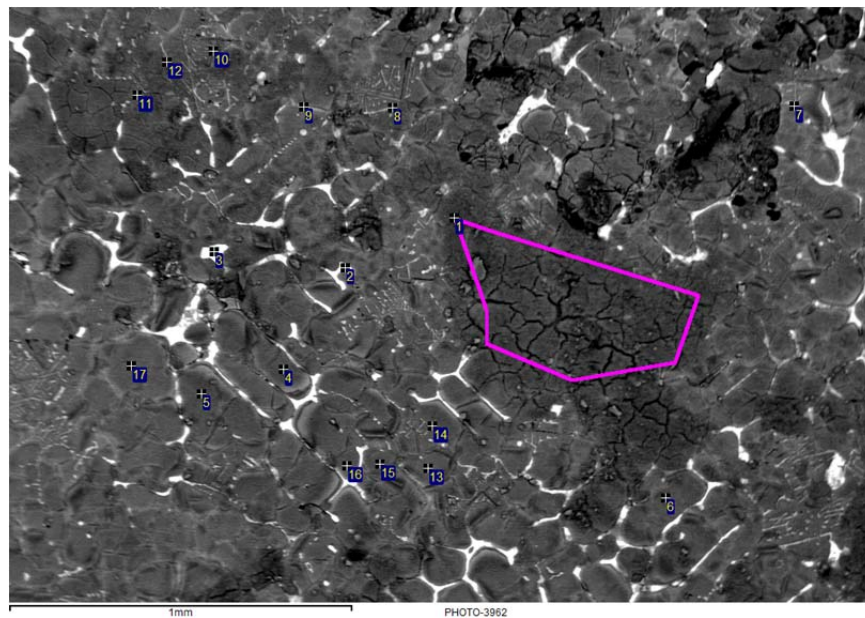


Figure 41 SEM image of 1U:1Zr surface, showing crevices or pits in the surface as well as oxides.

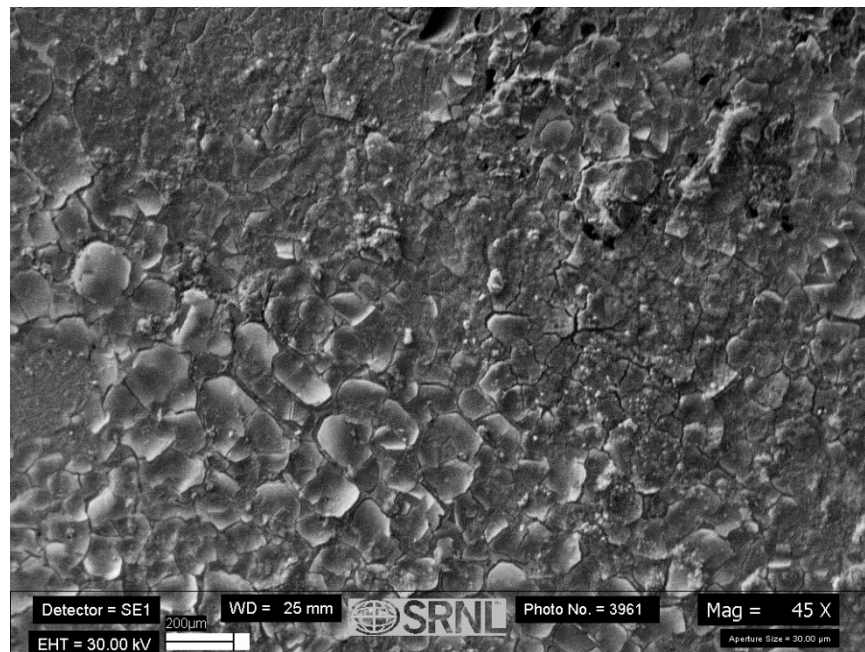


Figure 42 Secondary electron image of same region in Figure 41.

Table 31 EDS point analysis corresponding to points in Figure 41.

| With Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|---|------|------|-----|------|-----|-----|------|-----|-----|-----|-----|-----|
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | 71.4 | 5.0 | 1.0 | 19.0 | 0.0 | 0.0 | 3.0 | 0.6 | 0.0 | 0.0 | 0.0 | 0.0 |
| 2 | 88.1 | 0.6 | 0.0 | 2.2 | 0.0 | 1.8 | 1.1 | 0.0 | 0.0 | 0.0 | 0.0 | 6.1 |
| 3 | 90.8 | 0.0 | 0.0 | 2.2 | 0.5 | 1.9 | 0.9 | 0.0 | 0.0 | 0.0 | 0.0 | 3.8 |
| 4 | 64.9 | 4.1 | 0.9 | 28.2 | 0.3 | 0.0 | 1.6 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 5 | 64.6 | 4.2 | 1.1 | 28.3 | 0.0 | 0.0 | 2.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 6 | 57.5 | 4.3 | 1.1 | 34.8 | 0.5 | 0.0 | 1.8 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 7 | 65.2 | 3.5 | 0.9 | 27.4 | 0.0 | 0.9 | 1.6 | 0.6 | 0.0 | 0.0 | 0.0 | 0.0 |
| 8 | 60.3 | 4.9 | 1.0 | 31.9 | 0.0 | 0.0 | 1.9 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 9 | 69.5 | 3.9 | 1.0 | 23.9 | 0.0 | 0.0 | 1.8 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 10 | 71.8 | 3.5 | 0.8 | 21.9 | 0.3 | 0.0 | 1.8 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 11 | 65.2 | 3.8 | 0.7 | 28.5 | 0.0 | 0.0 | 1.8 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 12 | 59.7 | 4.8 | 1.4 | 32.4 | 0.0 | 0.0 | 1.3 | 0.5 | 0.0 | 0.0 | 0.0 | 0.0 |
| 13 | 61.7 | 4.2 | 1.1 | 30.8 | 0.0 | 0.0 | 1.8 | 0.4 | 0.0 | 0.0 | 0.0 | 0.0 |
| 14 | 63.0 | 4.0 | 1.0 | 30.0 | 0.4 | 0.0 | 1.6 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 15 | 63.4 | 4.1 | 1.0 | 29.1 | 0.0 | 0.0 | 1.8 | 0.5 | 0.0 | 0.0 | 0.0 | 0.0 |
| 16 | 61.7 | 4.4 | 1.0 | 30.3 | 0.4 | 0.0 | 1.8 | 0.5 | 0.0 | 0.0 | 0.0 | 0.0 |
| 17 | 58.4 | 4.3 | 1.4 | 33.8 | 0.0 | 0.0 | 2.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 16.9 | 3.5 | 65.6 | 0.0 | 0.0 | 11.4 | 2.5 | 0.0 | 0.0 | 0.0 | 0.0 |

| | | | | | | | | | | | | |
|----|---|------|-----|------|-----|------|------|-----|-----|-----|-----|------|
| 2 | - | 5.3 | 0.0 | 17.8 | 0.0 | 15.8 | 9.9 | 0.0 | 0.0 | 0.0 | 0.0 | 51.2 |
| 3 | - | 0.0 | 0.0 | 22.1 | 4.6 | 21.2 | 10.2 | 0.0 | 0.0 | 0.0 | 0.0 | 42.0 |
| 4 | - | 11.6 | 2.5 | 78.7 | 0.8 | 0.0 | 5.1 | 0.0 | 0.0 | 0.0 | 1.3 | 0.0 |
| 5 | - | 11.6 | 2.9 | 77.8 | 0.0 | 0.0 | 6.3 | 0.0 | 0.0 | 0.0 | 1.4 | 0.0 |
| 6 | - | 10.0 | 2.6 | 80.2 | 1.2 | 0.0 | 4.7 | 0.0 | 0.0 | 0.0 | 1.2 | 0.0 |
| 7 | - | 9.9 | 2.5 | 76.5 | 0.0 | 2.9 | 5.1 | 2.0 | 0.0 | 0.0 | 1.2 | 0.0 |
| 8 | - | 12.1 | 2.5 | 78.0 | 0.0 | 0.0 | 5.5 | 0.0 | 0.0 | 0.0 | 1.9 | 0.0 |
| 9 | - | 12.4 | 3.2 | 76.0 | 0.0 | 0.0 | 6.8 | 0.0 | 0.0 | 0.0 | 1.6 | 0.0 |
| 10 | - | 12.1 | 2.6 | 75.2 | 1.1 | 0.0 | 7.3 | 0.0 | 0.0 | 0.0 | 1.9 | 0.0 |
| 11 | - | 10.7 | 2.0 | 79.9 | 0.0 | 0.0 | 6.0 | 0.0 | 0.0 | 0.0 | 1.5 | 0.0 |
| 12 | - | 11.9 | 3.3 | 77.8 | 0.0 | 0.0 | 3.5 | 1.4 | 0.0 | 0.0 | 2.1 | 0.0 |
| 13 | - | 10.9 | 2.8 | 78.8 | 0.0 | 0.0 | 5.2 | 1.3 | 0.0 | 0.0 | 1.1 | 0.0 |
| 14 | - | 10.8 | 2.7 | 79.8 | 1.1 | 0.0 | 4.8 | 0.0 | 0.0 | 0.0 | 0.8 | 0.0 |
| 15 | - | 11.1 | 2.8 | 77.9 | 0.0 | 0.0 | 5.6 | 1.5 | 0.0 | 0.0 | 1.2 | 0.0 |
| 16 | - | 11.4 | 2.4 | 77.3 | 1.0 | 0.0 | 5.3 | 1.4 | 0.0 | 0.0 | 1.2 | 0.0 |
| 17 | - | 10.3 | 3.4 | 80.0 | 0.0 | 0.0 | 5.4 | 0.0 | 0.0 | 0.0 | 0.9 | 0.0 |

3.6.6 1U:2Zr 300 mV Acidic Brine Potentiostatic Hold Hybrid Test SEM/EDS

Figure 43 and Table 32 show EDS large area analysis of the 1U:2Zr electrode after the acidic brine 300 mV potentiostatic hold hybrid test. Little change is observed between pre and post hybrid test large area EDS analysis. Ru, Rh, and Pd are not detected, but the accuracy of EDS semi-quantitative data is generally considered accurate to within a few percent.

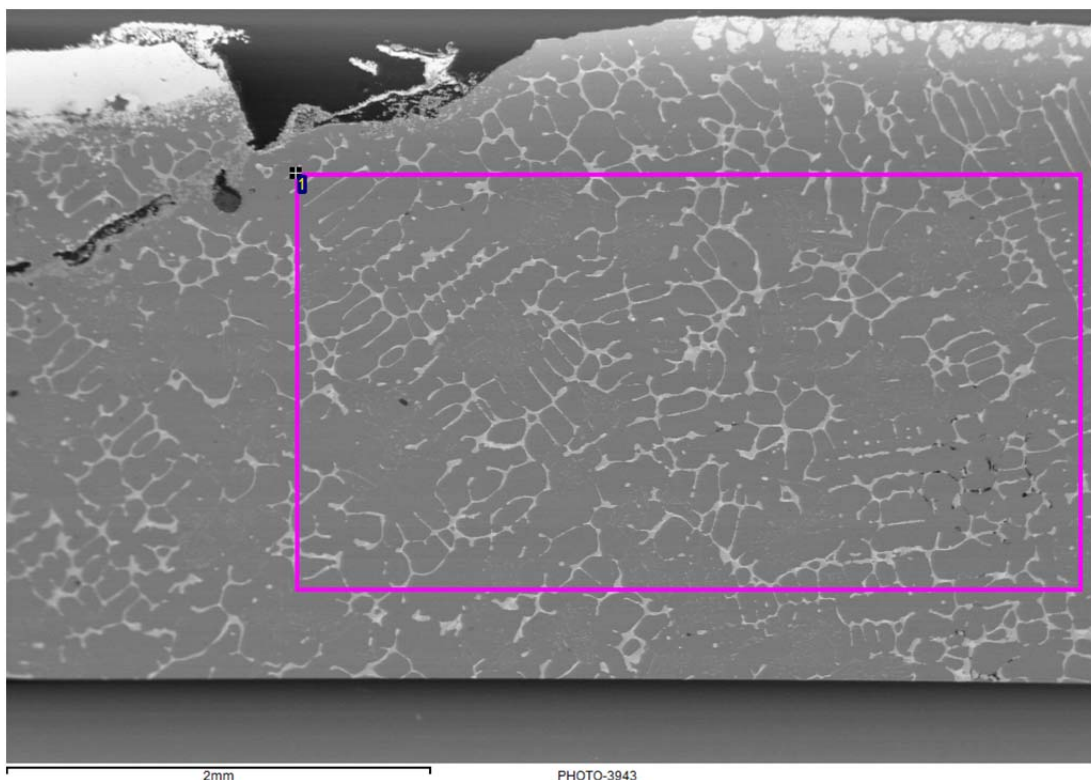


Figure 43 Large area EDS scans of several locations on the 1U:2Zr alloy after the 300 mV potentiostatic hold hybrid test.

Table 32 Large area EDS scans from the 1U:2Zr electrode showing mean oxygen concentrations on the surface. Area number corresponds to number in Figure 43.

| With Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|---|------|------|-----|------|-----|-----|-----|-----|-----|-----|-----|-----|
| Area | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | 35.5 | 6.5 | 2.0 | 51.9 | 0.6 | 1.6 | 1.2 | 0.0 | 0.0 | 0.0 | 0.0 | 0.8 |
| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 10.0 | 3.0 | 78.7 | 0.9 | 2.6 | 2.0 | 0.0 | 0.0 | 0.0 | 1.5 | 1.3 |
| As fabricated EDS data from (Olson 2012) | | | | | | | | | | | | |
| - | - | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| - | - | 11.5 | 0.6 | 76.2 | 0.5 | 2.3 | 1.8 | 1.1 | 1.1 | 1.1 | 1.1 | 1.2 |

The as fabricated phase data from an earlier report (Olson 2012), is shown in Table 33 for quick reference and comparison to the EDS point scans in this section.

Table 33 Phase data taken from a previous report to aid in discerning oxidized phases.

| 1U:1Zr | | | | | | | | | | | |
|--------|----|----|----|----|----|----|----|----|----|----|---|
| Phase | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |

| | | | | | | | | | | | |
|----------------------------------|------|-----|------|-----|------|-----|-----|------|------|-----|------|
| Steel | 12.2 | 2.3 | 80.4 | 0.4 | 0.0 | 1.8 | 1.2 | 0.5 | 0.3 | 0.9 | 0.0 |
| (Zr, U)Fe ₂ | 2.2 | 1.0 | 55.3 | 1.7 | 19.9 | 1.4 | 1.1 | 7.2 | 2.9 | 0.0 | 7.5 |
| (Zr, U)Pd ₂ | 0.0 | 0.0 | 4.9 | 1.8 | 12.5 | 0.9 | 0.0 | 0.0 | 61.1 | 0.0 | 18.8 |
| (Zr, U)(Fe, Pd, Rh) ₃ | 1.9 | 0.6 | 39.2 | 3.8 | 10.2 | 0.0 | 0.0 | 12.7 | 11.7 | 0.0 | 19.8 |
| Zr Rich | 1.2 | 0.1 | 6.3 | 0.1 | 87.3 | 4.8 | 0.0 | 0.0 | 0.0 | 0.0 | 0.2 |

Figure 44 shows a region in SEM images that has evidence of localized attack on the high Z phases as observed for the alkaline brine tests using the 1U:2Zr electrode. The electrode used in the acidic brine tests was made from a different piece of the 1U:2Zr alloy then was used for earlier alkaline brine testing, so this electrode surface is completely independent from the earlier tests. A piece of fiber, likely from a cotton ball used to pack the electrode for transport is also seen to have caught on the electrode surface, and is believed to be non-relevant.

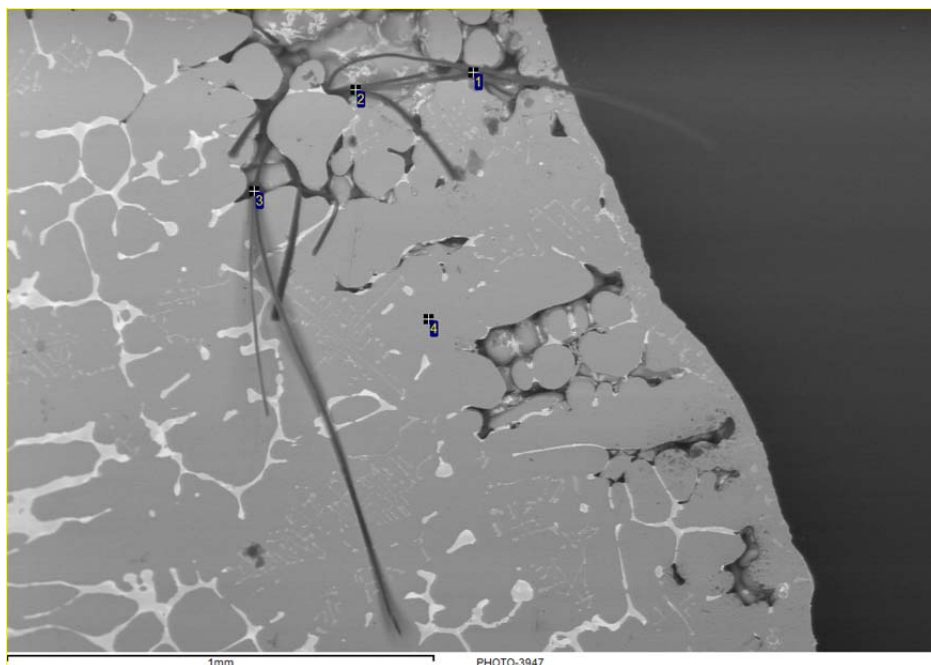


Figure 44 SEM image showing preferential attack of Zr rich phases in the alloy electrode.

Figure 45 shows a region of the surface of the 1U:2Zr electrode that has an oxide layer present, and Table 34 shows EDS analyses from that region. The oxide layer characteristics are shown in more detail in Figure 46 which shows the region in both BSE and SE imaging.

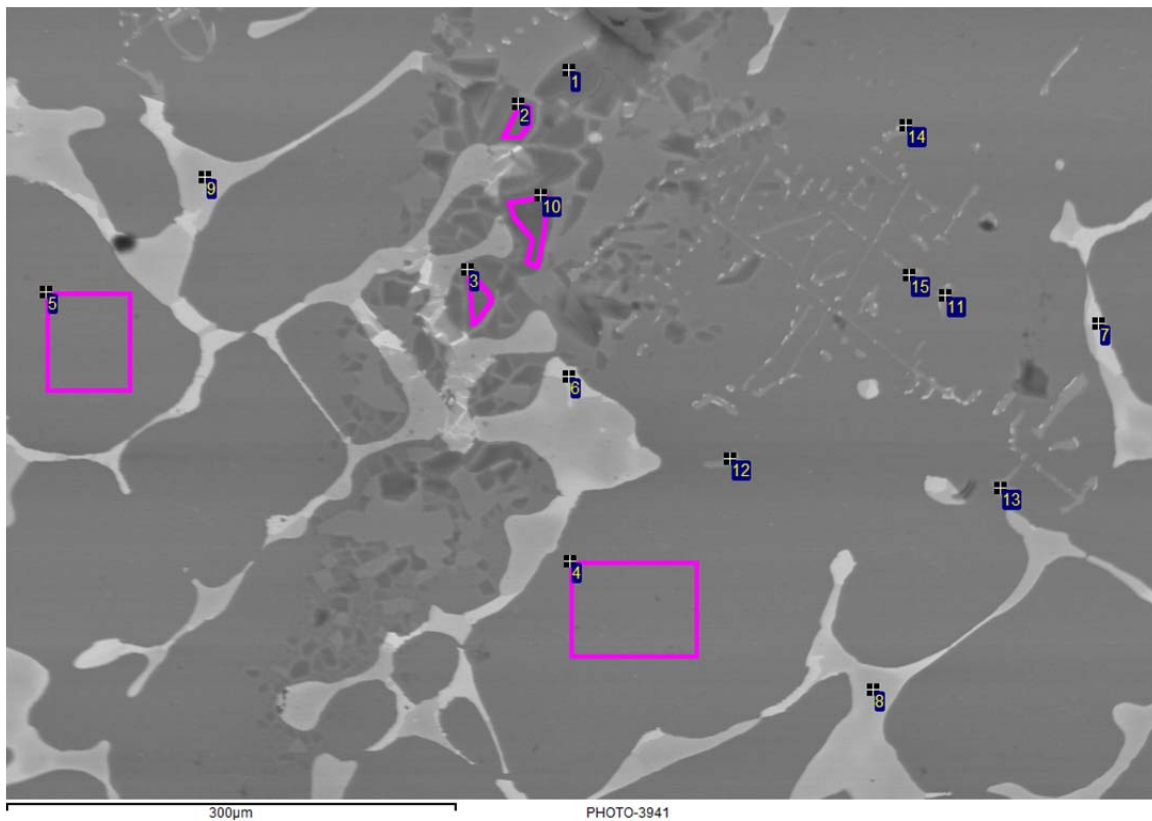


Figure 45 BSE image of oxidized region of 1U:2Zr electrode after being held at 300 mV for several hundred hours in acidic brine.

Table 34 EDS point analysis corresponding to points in Error! Reference source not found..

| With Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|--|------|-----|-----|------|-----|------|-----|-----|-----|-----|-----|-----|
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | 61.9 | 5.0 | 1.0 | 30.1 | 0.0 | 0.0 | 1.5 | 0.6 | 0.0 | 0.0 | 0.0 | 0.0 |
| 2 | 51.8 | 5.9 | 1.4 | 38.2 | 0.5 | 0.0 | 1.6 | 0.6 | 0.0 | 0.0 | 0.0 | 0.0 |
| 3 | 55.2 | 5.4 | 1.6 | 35.8 | 0.4 | 0.0 | 1.7 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 4 | 38.9 | 6.7 | 1.8 | 51.3 | 0.6 | 0.0 | 0.8 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 5 | 39.6 | 6.2 | 1.9 | 50.9 | 0.7 | 0.0 | 0.8 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 6 | 85.7 | 1.2 | 0.0 | 2.6 | 0.0 | 2.6 | 1.0 | 0.0 | 0.0 | 1.8 | 0.0 | 5.1 |
| 7 | 83.9 | 0.0 | 0.0 | 2.6 | 0.0 | 2.1 | 1.7 | 0.0 | 0.0 | 0.0 | 0.0 | 9.7 |
| 8 | 59.8 | 1.1 | 1.0 | 24.2 | 1.2 | 6.9 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 5.8 |
| 9 | 59.8 | 1.1 | 0.8 | 24.4 | 1.0 | 6.5 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 6.3 |
| 10 | 49.5 | 6.7 | 1.5 | 40.2 | 0.4 | 0.0 | 1.1 | 0.7 | 0.0 | 0.0 | 0.0 | 0.0 |
| 11 | 65.4 | 2.5 | 0.6 | 15.3 | 0.0 | 14.9 | 1.3 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 12 | 76.3 | 0.3 | 0.0 | 1.0 | 0.0 | 20.7 | 1.7 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 13 | 73.3 | 0.4 | 0.0 | 1.3 | 0.0 | 23.0 | 2.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 14 | 77.2 | 1.4 | 0.0 | 7.2 | 0.0 | 10.4 | 1.1 | 0.0 | 0.0 | 0.0 | 0.0 | 2.7 |
| 15 | 75.2 | 0.4 | 0.0 | 1.7 | 0.0 | 20.1 | 1.8 | 0.0 | 0.0 | 0.0 | 0.0 | 0.8 |

| Without Oxygen in Scan - All results in atomic% | | | | | | | | | | | | |
|---|---|------|-----|------|-----|------|------|-----|-----|------|-----|------|
| Point | O | Cr | Mn | Fe | Ni | Zr | Mo | Ru | Rh | Pd | Re | U |
| 1 | - | 13.0 | 2.5 | 77.0 | 0.0 | 0.0 | 4.3 | 1.8 | 0.0 | 0.0 | 1.4 | 0.0 |
| 2 | - | 12.2 | 2.9 | 77.6 | 1.0 | 0.0 | 3.6 | 1.4 | 0.0 | 0.0 | 1.4 | 0.0 |
| 3 | - | 11.9 | 3.4 | 78.1 | 0.8 | 0.0 | 4.3 | 0.0 | 0.0 | 0.0 | 1.5 | 0.0 |
| 4 | - | 11.0 | 2.8 | 82.9 | 0.9 | 0.0 | 1.4 | 0.0 | 0.0 | 0.0 | 1.0 | 0.0 |
| 5 | - | 10.2 | 3.1 | 83.0 | 1.2 | 0.0 | 1.4 | 0.0 | 0.0 | 0.0 | 1.2 | 0.0 |
| 6 | - | 8.6 | 0.0 | 17.4 | 0.0 | 18.5 | 7.4 | 0.0 | 0.0 | 12.9 | 0.0 | 35.2 |
| 7 | - | 0.0 | 0.0 | 15.6 | 0.0 | 13.6 | 10.7 | 0.0 | 0.0 | 0.0 | 0.0 | 60.1 |
| 8 | - | 2.8 | 2.6 | 59.2 | 2.8 | 17.8 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 14.9 |
| 9 | - | 2.9 | 2.1 | 59.7 | 2.5 | 16.7 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 16.1 |
| 10 | - | 13.1 | 2.9 | 77.8 | 0.8 | 0.0 | 2.4 | 1.4 | 0.0 | 0.0 | 1.7 | 0.0 |
| 11 | - | 6.9 | 1.7 | 41.8 | 0.0 | 44.1 | 4.2 | 0.0 | 0.0 | 0.0 | 1.3 | 0.0 |
| 12 | - | 1.4 | 0.0 | 4.0 | 0.0 | 85.5 | 9.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 13 | - | 1.5 | 0.0 | 4.8 | 0.0 | 84.1 | 9.6 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| 14 | - | 6.1 | 0.0 | 30.1 | 0.0 | 45.9 | 5.1 | 0.0 | 0.0 | 0.0 | 0.0 | 12.8 |
| 15 | - | 1.7 | 0.0 | 6.6 | 0.0 | 79.3 | 8.7 | 0.0 | 0.0 | 0.0 | 0.0 | 3.8 |

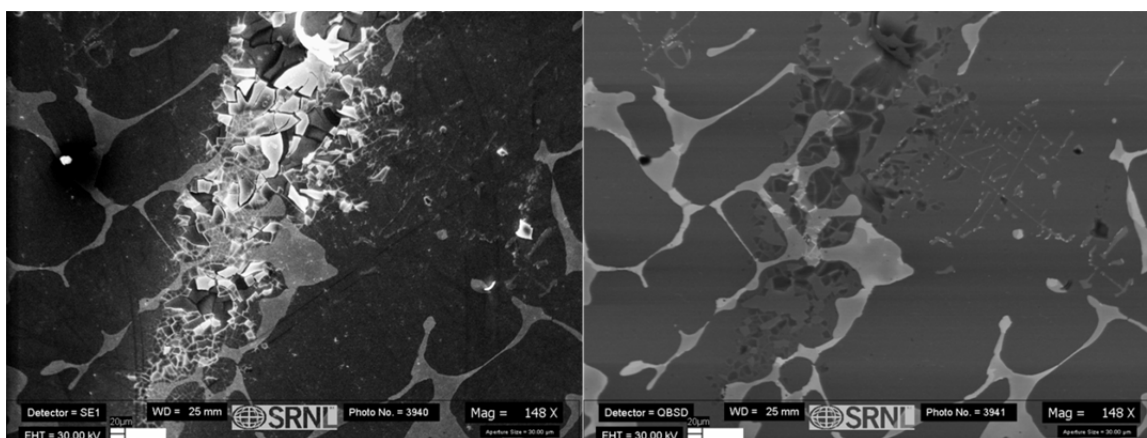


Figure 46 Back Scatter electron image (left) and secondary electron image (right) showing compositional and topographical effects of localized corrosion.

3.7 Solution Analysis

Aliquots from the test solutions were taken and analyzed at specific points during the long-term electrochemical measurements. The following elements were analyzed for: Mo, Re, and U.

3.7.1 1U:2Zr 50 mV Alkaline Brine Solution Analysis

Table 35 and Figure 47 show the electrolyte solution analyses from the alkaline brine 50 mV hybrid test with the 1U:2Zr alloy electrode. There was negligible U, Mo or Re (surrogates for Tc) releases beyond 2 days.

Table 35 Concentration of Mo, Re, and U as a function of test duration.

| 1U:2Zr | | | | |
|-------------------------|-------------------------|-------------|-------------|------------|
| Exposure time (days) | Exposure time (UNIX) | Mo (ppb) | Re (ppb) | U (ppb) |
| 0.00 | 1422294421 | <0.8 | <0.4 | <0.4 |
| 1.84 | 1422453120 | 2.18 | 2.52 | 3.60 |
| 8.89 | 1423062300 | 2.19 | 2.62 | 3.14 |
| 15.89 | 1423667400 | 1.87 | 3.34 | 2.25 |
| 22.84 | 1424268000 | 1.97 | 2.90 | 2.54 |
| 29.82 | 1424871300 | 2.04 | 3.01 | 2.64 |

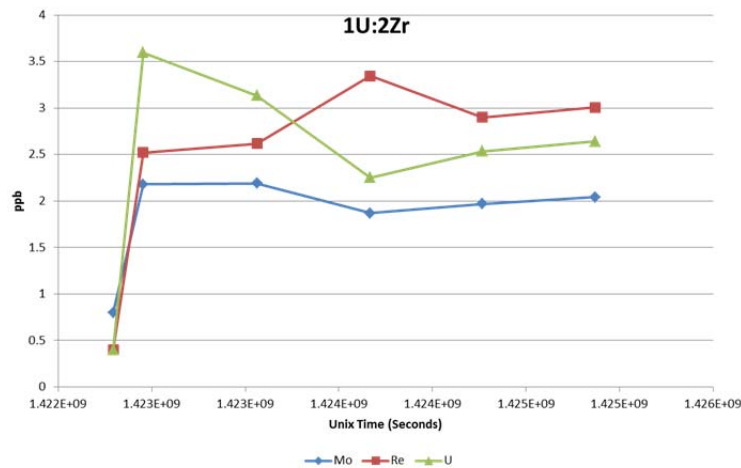


Figure 47 Concentration of Mo, Re, and U as a function of test duration.

3.7.2 1U:2Zr 65 mV Alkaline Brine Solution Analysis

Table 36 and Figure 48 show the electrolyte solution analyses from the alkaline brine 65 mV hybrid test with the 1U:2Zr alloy electrode. Solution concentrations show a slow, but constant Re (surrogate for Tc) and Mo release rate throughout test.

Table 36 Concentration of Mo, Re, and U as a function of test duration.

| 1U:2Zr | | | | |
|-------------------------|-------------------------|-------------|-------------|------------|
| Exposure time (days) | Exposure time (UNIX) | Mo (ppb) | Re (ppb) | U (ppb) |
| 0.00 | 1433935800 | 0.83 | < .4 | 1.88 |
| 7.14 | 1434553200 | 1.64 | 0.98 | 1.43 |

| | | | | |
|-------|------------|------|------|------|
| 14.16 | 1435159200 | 2.29 | 1.59 | 1.83 |
| 21.03 | 1435752960 | 2.39 | 1.98 | 1.99 |
| 27.96 | 1436351400 | 3.03 | 3.12 | 1.61 |

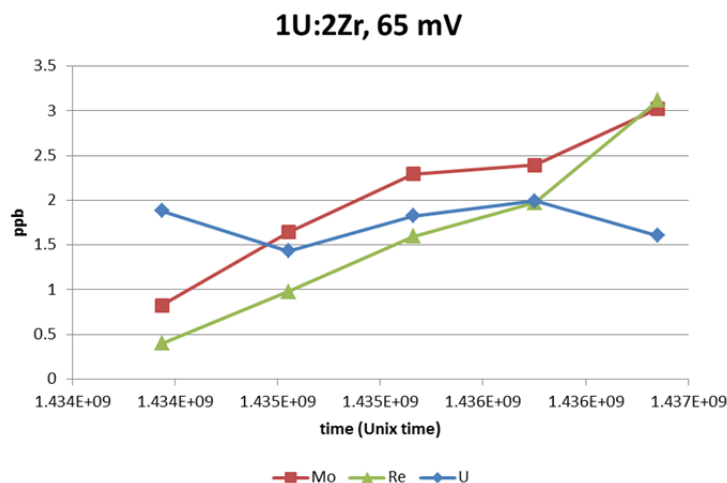


Figure 48 Concentration of Mo, Re, and U as a function of test duration.

3.7.3 3U:1Zr 50 mV Alkaline Brine Solution Analysis

Table 37 and Figure 49 show the electrolyte solution analyses from the alkaline brine 50 mV hybrid test with the 3U:1Zr alloy electrode. Solution concentrations show constant Re release rate throughout test (surrogate for Tc).

Table 37 Concentration of Mo, Re, and U as a function of test duration.

| 3U:1Zr | | | | |
|----------------------|----------------------|----------|----------|---------|
| Exposure time (days) | Exposure time (UNIX) | Mo (ppb) | Re (ppb) | U (ppb) |
| 0.00 | 1422294447 | <0.8 | <0.4 | <0.4 |
| 1.84 | 1422453120 | 5.57 | 13.33 | 14.12 |
| 8.89 | 1423062300 | 10.06 | 48.82 | 1.03 |
| 15.89 | 1423667400 | 7.84 | 88.89 | 0.86 |
| 22.84 | 1424268000 | 5.75 | 125.11 | <0.4 |
| 29.82 | 1424871300 | 2.62 | 154.86 | <0.4 |

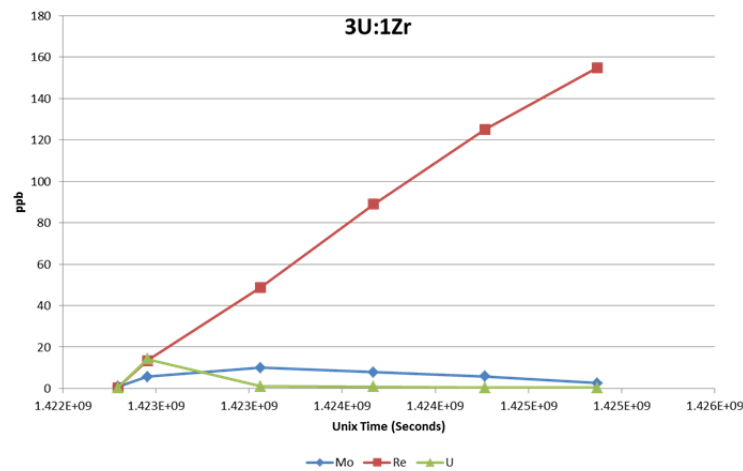


Figure 49 Concentration of Mo, Re, and U as a function of test duration.

3.7.4 3U:1Zr -200 mV Alkaline Brine Solution Analysis

Table 38 and Figure 50 show the electrolyte solution analyses from the alkaline brine -200 mV hybrid test with the 3U:1Zr alloy electrode. Solution concentrations show an initial large release, followed by a more gradual, but constant Re (surrogate for Tc) and Mo release rate throughout test.

Table 38 Concentration of Mo, Re, and U as a function of test duration.

| 3U:1Zr | | | | |
|----------------------|----------------------|----------|----------|---------|
| Exposure time (days) | Exposure time (UNIX) | Mo (ppb) | Re (ppb) | U (ppb) |
| 0.00 | 1433935800 | 0.61 | < 0.4 | < 0.4 |
| 6.97 | 1434553200 | 155.64 | 363.36 | 0.59 |
| 13.99 | 1435159500 | 181.70 | 390.38 | 0.47 |
| 20.86 | 1435753200 | 194.04 | 428.25 | 1.88 |
| 27.78 | 1436351400 | 198.39 | 414.61 | 0.88 |

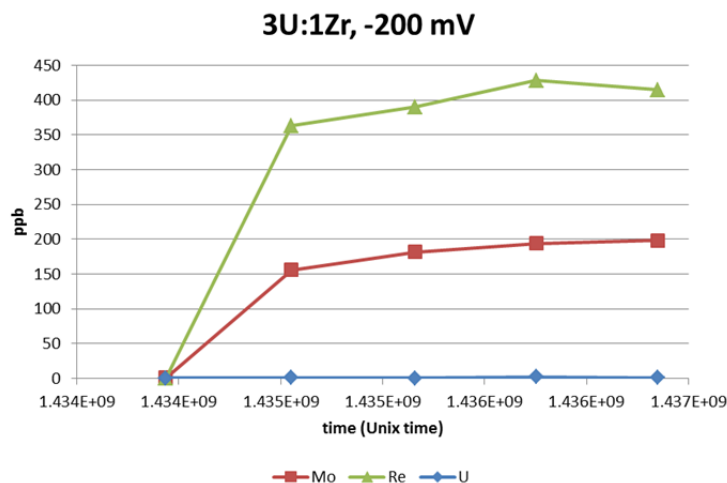


Figure 50 Concentration of Mo, Re, and U as a function of test duration.

3.7.5 1U:1Zr 300 mV Acidic Brine Solution Analysis

Table 39, Figure 51, and Figure 52 show the electrolyte solution analyses from the acidic brine 300 mV hybrid test with the 1U:1Zr alloy electrode. Solution concentrations prior to the electrode short between the WE and RE show an initial large U release after the first week of testing, followed by a smaller U release the second week. The Re has a slower more gradual release over the first two weeks of testing. After the electrode shorting, there is a large initial U and Mo release, but these concentrations decrease in subsequent weeks and Re concentration increases. It is possible the sample with the large U and Mo content included more of a brown “gel-like” precipitate layer near the base of the test vessel; care was taken to avoid this material in subsequent aliquots. It is also seen that the solution pH changed significantly post shorting, going from acidic to neutral.

Table 39 Concentration of Mo, Re, and U as a function of test duration.

| 1U:1Zr | | | | | |
|----------------------|----------------------|----------|----------|----------|------|
| Exposure time (days) | Exposure time (UNIX) | Mo (ppb) | Re (ppb) | U (ppb) | pH |
| 0.00 | 1438787796 | 2.52 | 0.40 | 2.16 | 3.94 |
| 6.71 | 1439367840 | 1.43 | 21.87 | 87.36 | 4.14 |
| 13.68 | 1439969640 | 1.87 | 70.08 | 97.89 | 4.49 |
| 20.81 | 1440585360 | 5789.55 | 9225.33 | 14693.65 | 7.59 |
| 27.98 | 1441205640 | 82.05 | 12681.72 | 277.87 | 6.95 |
| 33.78 | 1441706400 | 482.70 | 14385.38 | 870.76 | 6.72 |

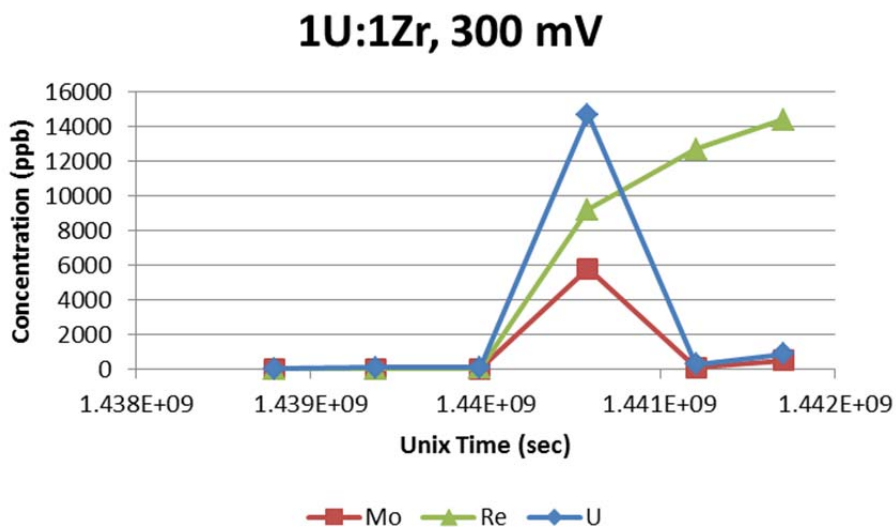


Figure 51 Concentration of Mo, Re, and U as a function of test duration.

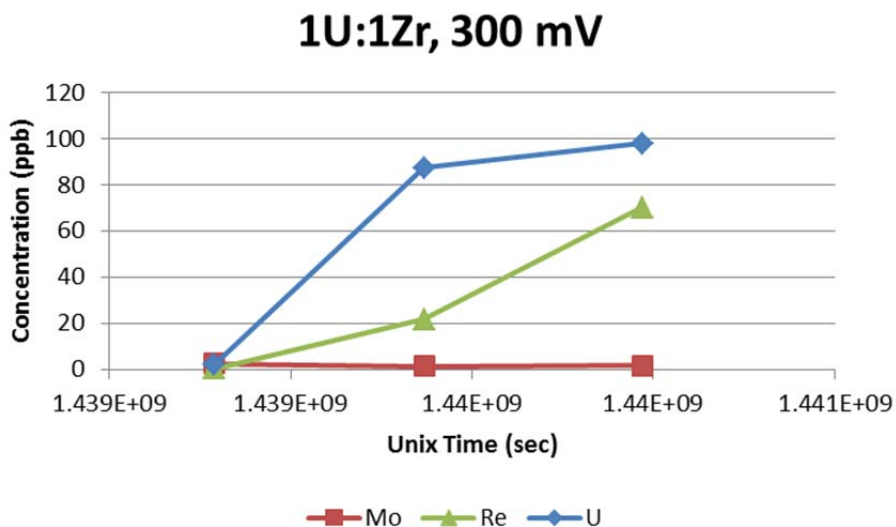


Figure 52 Concentration of Mo, Re, and U as a function of test duration. Results prior to electrode shorting.

3.7.6 1U:2Zr 300 mV Acidic Brine Solution Analysis

Table 40 and Figure 53 show the electrolyte solution analyses from the acidic brine 300 mV hybrid test with the 1U:2Zr alloy electrode. Solution concentrations show an initial in U and Re release after the first week of testing, followed by a mediating U release and somewhat steady Re release. The solution pH remains acidic during the hybrid test.

Table 40 Concentration of Mo, Re, and U as a function of test duration.

| 1U:2Zr | | | | | |
|-------------------------|-------------------------|-------------|-------------|------------|------|
| Exposure time (days) | Exposure time (UNIX) | Mo (ppb) | Re (ppb) | U (ppb) | pH |
| 0.00 | 1438787738 | 2.52 | <0.4 | 2.16 | 3.94 |
| 6.71 | 1439367720 | 1.55 | 3.57 | 7.93 | 4.12 |
| 13.68 | 1439969520 | 1.90 | 3.61 | 8.29 | 4.01 |
| 20.80 | 1440585120 | 2.40 | 6.84 | 11.52 | 4.02 |
| 27.98 | 1441205520 | 2.16 | 9.96 | 12.77 | 4.08 |
| 33.78 | 1441706400 | 2.19 | 15.09 | 11.34 | 4.07 |

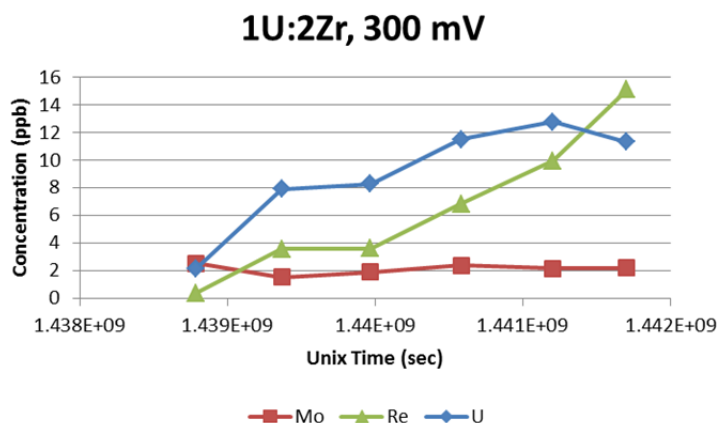


Figure 53 Concentration of Mo, Re, and U as a function of test duration.

4. CONCLUSIONS

Surrogate alloy waste forms of the RAW3-(URe) composition were made into electrodes and examined electrochemically in alkaline and acidic brines. The testing approach was developed in collaboration with Argonne National Laboratory, and consisted of 4 main steps, 1) measure the bare surface corrosion behavior in reference solutions at open circuit over a wide imposed potential range, 2) perform several differing potentiostatic holds to measure the time-evolution of anodic current, evolution of surface properties, and radionuclide concentrations in solution, 3) relate steady-state current to radionuclide (or surrogate) release rates through periodic sampling of test electrolyte, and 4) identify the corroding phase/phases using SEM/EDS. Step 2 could last up to 30 days with mostly fixed electrode potential and periodic potential interruptions for a variety of electrochemical analyses. This combined electrochemical and extended immersion testing is referred to as hybrid testing.

Electrochemical Analysis

The potentiodynamic scans on the fresh surfaces indicated a passive zone in alkaline brine for the 1U:2Zr alloy electrode from between about 50 mV to 100 mV vs SCE. In alkaline and acidic

brines the 1U:2Zr electrodes had larger current densities than the 1U:1Zr and 3U:1Zr electrodes at similar anodic potentials. In the acidic brines, the corrosion potential of the RAW3 alloys was more anodic than pure HT9 alloy, from which the RAW3 alloys are derived.

The current density from the alkaline brine 50 mV potentiostatic hold of the 1U:2Zr alloy indicates a passivating surface. A similar passivation of the surface was observed for the acidic brine 300 mV potentiostatic hold of the 1U:2Zr alloy. The alkaline brine 65 mV potentiostatic hold of the 1U:2Zr alloy indicates a reducing surface condition, indicating that perhaps the diamond polishing of the alloy electrode surface may have been insufficient to refresh the electrode surface back to pre-immersion condition.

The alkaline brine 50 mV potentiostatic hold of the 3U:1Zr alloy current density increased over the first week, and stayed at a relatively high value for the test duration. The current density of the alkaline brine -200 mV potentiostatic hold of the 3U:1Zr alloy was negative, indicating a reducing condition. A reducing condition was one of the expected outcomes for this alloy, although it is also possible a prior test at 50 mV may have affected this alloy in a similar fashion as prior test affects for the 1U:2Zr alloy.

The current density of the acidic brine 300 mV potentiostatic hold of the 1U:1Zr alloy showed a gradual increase prior to the short that occurred around day 13. The current increase for the 1U:1Zr alloy was from $\sim 5 \times 10^{-6}$ A/cm² to $\sim 15 \times 10^{-6}$ A/cm², which indicated that the surface was not passivating.

Tafel testing was used to derive Tafel constants and Stern-Geary coefficients needed for analysis of linear polarization resistance data. The polarization resistance of the 1U:2Zr alloy, during the course of the alkaline brine 50 mV potentiostatic holds first increased gradually before decreasing back to pre-test values, during the course of the alkaline brine 65 mV potentiostatic holds it varied little, and during the course of the acidic brine 300 mV potentiostatic holds of it gradually increased. This 1Z:2Zr alloy behavior indicated passivation in alkaline brines at 50 mV, little passivation at 65 mV, and passivation in acidic brines at 300 mV. The polarization resistance, during the course of the alkaline brine 50 mV potentiostatic holds of the 3U:1Zr alloy decreased over the first couple days and then held relatively constant, during the course of the alkaline brine -200 mV potentiostatic holds it gradually increased. This indicated the 3U:1Zr alloy was not passivated at 50 mV, but the surface may have been at -200 mV. The polarization resistance, during the course of the acidic brine 300 mV potentiostatic holds of the 1U:1Zr alloy at first increased but then decreased compared to the initial value, and continued decreasing for much of the testing indicating an active surface.

EIS indicated the 1U:2Zr alloy surface was passivating in the alkaline brine, but this passivation tenuous in the acidic brines, with passivation decreasing after day 15.

SEM/EDS

The EDS indicated minor apparent corrosion in all the alloy electrodes except for the 1U:1Zr alloy electrode where shorting was present, and a large amount of current was passed that resulted in largescale loss of metal. The 1U:2Zr alloy had localized corrosion in the high Z phases, predominantly the (Zr, U)Fe₂ and the (Zr, U)Pd₂ phases. The localized corrosion was apparent in both the alkaline and acidic brines, in the 3 electrode hold potentials.

The 3U:1Zr alloy electrode had general corrosion in the alkaline brine at 50 mV, and in the alkaline brine at -200 mV localized corrosion similar to that seen in the 1U:2Zr alloy was

observed. The localized attack appeared to start at the interface between the high Z phases and steel phase, and proceed into the high Z phase with increasing time.

The 1U:1Zr alloy electrode was damaged from large amounts of current that resulted from the short between the WE and the RE, and therefore its microstructure as a direct result of the pre-short corrosion is obscured. The surface of the 1U:1Zr electrode showed large scale metal loss and heavy oxide layer build-up. Regions of the electrode, usually near the interface of the electrode and the nonconductive epoxy mount, appeared especially susceptible to localized corrosion, which appeared to manifest itself primarily through oxidation of the steel phase. The determination that it was the steel phase that was attacked was made primarily through analysis of the SEM BSE images. Where the oxide scale flaked, often the underlying phase that was visible in BSE was a high Z phase.

Solution analysis

In all the cases for the 1U:2Zr alloy, both in the alkaline and acidic brines, the release of U, Mo, and Re were minor in comparison to the other electrodes, indicating increased Zr in the alloy increases the overall alloy durability. In the case of the alkaline brine 50 mV 1U:2Zr hybrid test, after an initial release, the detected levels of U, Mo, and Re flattens at about 3 to 2 ppb. In the alkaline brine 65 mV 1U:2Zr hybrid test, the U concentration was flat at about 2 ppb, and the Mo and Re concentration gradually increases from about 0.5 to 3 ppb. In the acidic brine 300 mV 1U:2Zr hybrid test, the Mo concentration was flat at about 2 ppb throughout the test duration, while the U content initially jumped and then increased in a haphazard manner before dropping slightly for the last measurements, and the Re concentration increased in a gradual manner throughout the testing.

The alkaline brine 3U:1Zr alloy electrode hybrid tests both resulted in the largest releases during normal test conditions. In the alkaline brine 50 mV 3U:1Zr alloy electrode hybrid test, the Re concentration increases linearly throughout the test, ending at about 160 ppb, the U increases the first week, before dropping back to below the detection limit of <0.4 ppb, and the Mo content increases the first two weeks to ~10 ppb before dropping back to ~3 ppb. The U concentrations remain flat and around the detection limit, and the Re and Mo content spike initially, before gradually increasing throughout the test to end at ~414 ppb and 198 ppb, respectively, in the alkaline brine -200 mV 3U:1Zr alloy electrode hybrid test. The Re concentration in the -200 mV hybrid test drops during the last measurement.

In the acidic brine 300 mV 1U:1Zr hybrid test, only solution samples from the first 2 weeks can be used to judge the corrosion behavior of the alloy electrode in the hybrid test. It is observed that the U content jumps the first week, then increases less the second. The Re content increases slightly in the first week, and then looks to increase its rate of release the second week. After the short and galvanic surge in the electrode, the U concentration increases to ~14700 ppb, Re to ~9230 ppb, and Mo concentration to ~5790 ppb. After the short was corrected, the U and Mo concentrations rapidly decrease, while the Re concentration increases gradually with each sampling period.

Overall Conclusions and Future Recommendations

Based on the Anodic currents, LPR, and EIS data, the 1U:2Zr alloy electrode appeared the most likely of the tested alloys to form a semi-protective passivation layer, although as evident from

SEM/EDS analysis, this alloy still suffered from localized corrosion of the high-Z phases responsible for sequestration of the actinides (U in these alloys). Based on solution analyses of the hybrid tests, the U concentration often initially spikes, but usually then stabilizes or drops as the Re content increases, indicating that the high-Z localized corrosion is either transitory, or that the released corrosion products are non-soluble.

Based on electrochemical tests and SEM/EDS, oxidation of the 1U:1Zr and 3U:1Zr alloy does not appear to form a protective passivation layer. When the 3U:1Zr alloy is held at reducing potentials, significant Re (and Mo in acidic brine) is still released. The 3U:1Zr alloy appears more prone to radionuclide release than the 1U:1Zr alloy. While the high release rates of Re indicate attack of the steel phase, localized attack of the high-Z phases was also visible on the 3U:1Zr alloy, indicating that the overall reducing condition of the alloy may have still been oxidizing for the high-Z phases.

Due to the nature of the localized corrosion affecting specific phases, it is recommended to examine the possibility of adding trim additions that may segregate to the high-Z phases, and either make this phase nobler, or aid in the formation of a protective oxide. It is hypothesized that Ni may perform this function, and future work will test this hypothesis. Previous work has shown Cr has limited solubility in the high-Z phases so Cr trim additions, while perhaps decreasing potential Tc release, will likely have little impact on the localized corrosion.

5. REFERENCES

Olson, L. (2011). FY2011 Status Report: Chromium Content In Iron Based Alloy Waste Forms, Savannah River National Laboratory Technical Document, SRNL-STI-2011-00579 Rev. 0 and FCRD-SWF-2011-000366.

Olson, L. (2012). Formulation of Reference Alloy Waste Form RAW-3, Savannah River National Laboratory Technical Document, SRNL-STI-2012-00534 and FCRD-SWF-2012-000348.

Olson, L. (2014). "Corrosion behavior of electrically coupled phases in bulk alloy waste form material, FCRD-SWF-2014-000245." 64.

6. APPENDIX 1: Electrochemical Measurement Data

Hybrid Testing Electrochemical Measurement Data

Table 41 Material Identification

| | | | | | | |
|--|-------------------------------------|--------|--------|--------|--------|--------|
| Unique material identification label | 1U:2Zr | 3U:1Zr | 1U:2Zr | 3U:1Zr | 1U:1Zr | 1U:2Zr |
| Identify QA level | Scoping | | | | | |
| Intended use of material | Basic analysis and characterization | | | | | |
| Production date(s) | FY12 | FY12 | FY12 | FY12 | FY12 | FY12 |
| Production location | SRNL | | | | | |
| Principle investigator | Luke Olson | | | | | |
| Radionuclide content (mass %) | 4 (depleted uranium) | | | | | |
| Comments by producer | None | | | | | |
| Location of ~500x magnification SEM image of alloy | In section 2.2. | | | | | |

Table 42 Corrosion Cell Set-Up

| | | | | | | |
|---|--|-----------------------|-----------------------|-----------------------|--|-----------------------|
| Unique material identification label | 1U:2Zr | 3U:1Zr | 1U:2Zr | 3U:1Zr | 1U:1Zr | 1U:2Zr |
| Identify QA level | Scoping | | | | | |
| Reference electrode used | RHE in tests, potential changed to SCE for most analysis. RHE is pH dependent. In alkaline brines (9.80 pH initial) after several days exposure to lab air, pH reading was 7.64, and RHE as RE vs. SCE as WE potential was 639.8 mV. In acidic brines, after several days air exposure, pH remained steady and gave reading of 3.94 pH. In acidic brine, RHE as RE and SCE as WE potential was 460.4 mV. | | | | | |
| Counter electrode used | Graphite. 2 x 0.25" graphite rods spaced equidistant from WE (about 2") | | | | | |
| WE surface finish (oxidized, polished to 600 grit...) | 1 micron diamond polish | | | | | |
| WE surface area (cm ²) | 1.026 cm ² | 0.801 cm ² | 1.026 cm ² | 0.801 cm ² | 0.931 cm ² | 0.292 cm ² |
| WE density (g/cm ³) | 9.0 | 9.1 | 9.0 | 9.1 | 9.1 | 9.0 |
| WE equivalent weight (units?) | 27.0 | 27.1 | 27.0 | 27.1 | 27.1 | 27.0 |
| Non-conductive epoxy used for WE | Epofix | | | | | |
| Identify and describe test generating sample (if analyzing a reacted specimen) | Samples were all freshly polished prior to hybrid testing | | | | | |
| Identify calibration of scale(s) used for solution (If current, or date of last calibration) | 08/27/2015 | | 08/27/2015 | | | |
| Identify standard name of electrolyte solution | Alkaline Brine | | | | Acidic Brine | |
| Electrolyte temperature (before, after, during, etc...) | 23 (before) | 23 (before) | 23 (before) | 23 (before) | 23 (before) | 23 (before) |
| Electrolyte Composition | (0.1 mmol NaOH + 10 mmol NaCl)/(1 kg H ₂ O) | | | | (0.1 mmol H ₂ SO ₄ + 10 mmol NaCl)/(1 kg H ₂ O) | |
| Test specific electrolyte conditions (e.g., was the electrolyte made a week before testing and sat out in a covered container for a week prior to measurements, etc...) | -100x concentration made from 0.0421g NaOH chips and 5.8441g NaCl and 100.0230g H ₂ O saturated in Air. Solution made day of initial hybrid tests (01/26/2015) then used for all tests. -round 1 tests: 01/26/2015, 15.1g stock mixed with 1485.0g H ₂ O, then mixed to greene cells as 1U:2Zr=726.7g solution, 3U:1Zr=737.6g solution -round 2 tests: solution mixed 06/04/2015 but tests not started until 06/10/2015, 15.0g stock mixed with 1485.0g H ₂ O, then partitioned to 700.0g solution to each greene cell. | | | | -100x concentration made from 0.1003g 98% concentrated sulfuric acid and 5.8440g NaCl and 100.0046g H ₂ O saturated in Air. Solution made 07/24/2015. -for bare surface tests on 07/24/2015, 15.0g of concentrate added to 1485.0g H ₂ O. 700.0g per green cell. -for hybrid tests starting on 08/04+05/2015, 15.0g concentrate added to 1485.22g H ₂ O, then 700.0g of solution add to each Greene cell. | |
| Test order (Date) | 01/26/2015 | | 06/10/2015 | | 08/03/2015 bare surface measurements & 08/05/2015 hybrid test | |

Table 43 Solution Sample Data

| Vial Sample # | greene cell origin | tare weight | solution volume | Date sample taken | Time sample taken: military time (hr:min:sec, use cpu time) | final weight | pH (2-11 measured on 03/16/2015) |
|-----------------|--------------------|-------------|-----------------|-------------------|---|--------------|----------------------------------|
| 1 | blank | 8.4494 | 5 mL | 1/22/2015 | "N.A. | 16.4655 | 6.85 |
| 2 | 1U:2Zr | 8.5188 | 5 mL | 1/28/2015 | 1:52 PM | 15.083 | 6.9 |
| 3 | 3U:1Zr | 8.6852 | 5 mL | 1/28/2015 | 1:52 PM | 13.1271 | 6.98 |
| 4 | 1U:2Zr | 8.6573 | 5 mL | 2/4/2015 | 3:05 PM | 13.224 | 6.89 |
| 5 | 3U:1Zr | 8.6893 | 5 mL | 2/4/2015 | 3:05 PM | 13.1421 | 6.85 |
| 6 | 1U:2Zr | 8.5204 | 5 mL | 2/11/2015 | 3:10 PM | 13.3564 | 7.05 |
| 7 | 3U:1Zr | 8.4621 | 5 mL | 2/11/2015 | 3:10 PM | 13.3187 | 7.12 |
| 8 | 1U:2Zr | 8.5476 | 5 mL | 2/18/2015 | 2:00 PM | 13.3518 | 6.94 |
| 9 | 3U:1Zr | 8.3967 | 5 mL | 2/18/2015 | 2:00 PM | 13.2257 | 6.98 |
| 10 | 1U:2Zr | 8.5168 | 5 mL | 2/25/2015 | 1:35 PM | 13.3001 | 6.86 |
| 11 | 3U:1Zr | 8.533 | 5 mL | 2/25/2015 | 1:35 PM | 13.454 | 6.88 |
| 12 | 1U:2Zr | 8.5224 | 5 mL | 6/10/2015 | 11:30 AM | 13.4275 | 7.08 ^a |
| 13 | 3U:1Zr | 8.5939 | 5 mL | 6/10/2015 | 11:30 AM | 13.542 | 7.1 |
| 14 | 1U:2Zr | 8.5602 | 5 mL | 6/17/2015 | 3:00 PM | 13.6479 | 7.07 |
| 15 | 3U:1Zr | 8.4753 | 5 mL | 6/17/2015 | 3:00 PM | 13.3777 | 7.13 |
| 16 | 1U:2Zr | 8.7403 | 5 mL | 6/24/2015 | 3:20 PM | 13.6926 | 7.06 |
| 17 | 3U:1Zr | 8.7374 | 5 mL | 6/24/2015 | 3:25 PM | 13.7777 | 7.09 |
| 18 | 1U:2Zr | 8.6245 | 5 mL | 7/1/2015 | 12:16 PM | 13.3059 | 7.11 |
| 19 | 3U:1Zr | 8.4808 | 5 mL | 7/1/2015 | 12:20 PM | 13.124 | 7.08 |
| 20 | 1U:2Zr | 8.506 | 5 mL | 7/8/2015 | 10:30 AM | 13.3902 | 7.07 |
| 21 | 3U:1Zr | 8.6291 | 5 mL | 7/8/2015 | 10:30 AM | 13.3457 | 7.04 |
| 22 | 1U:2Zr | 8.6188 | 10 mL | 8/5/2015 | Prior to test and electrode insertion | 18.3547 | 3.94 |
| 23 | - | - | - | - | - | - | - |
| 24 ^b | neither cell | 8.3534 | 10 mL | 7/27/2015 | used to check RE5 | 17.4946 | 3.78 |
| 25 | - | - | - | - | - | - | - |
| 26 | D, 1U:2Zr | 8.7354 | 5 mL | 8/12/2015 | 8:22 AM | 13.3992 | 4.12 |
| 27 | E, 1U:1Zr | 8.5976 | 5 mL | 8/12/2015 | 8:24 AM | 13.4797 | 4.14 |
| 28 | D, 1U:2Zr | 8.4738 | 5 mL | 8/19/2015 | 7:32 AM | 13.2556 | 4.01 |
| 29 | E, 1U:1Zr | 8.4435 | 5 mL | 8/19/2015 | 7:34 AM | 13.1674 | 4.49 |
| 30 | D, 1U:2Zr | 8.4564 | 5 mL | 8/26/2015 | 10:32 AM | 13.3746 | 4.02 |
| 31 | E, 1U:1Zr | 8.5844 | 5 mL | 8/26/2015 | 10:36 AM | 13.228 | 7.59 |
| 32 | D, 1U:2Zr | 8.6871 | 5 mL | 9/2/2015 | 2:52 PM | 13.588 | 4.08 |
| 33 | E, 1U:1Zr | 8.596 | 5 mL | 9/2/2015 | 2:54 PM | 13.411 | 6.95 |
| 34 | D, 1U:2Zr | 8.5471 | 5 mL | 9/8/2015 | 10:00 AM | 13.5596 | 4.07 |
| 35 | E, 1U:1Zr | 8.7281 | 5 mL | 9/8/2015 | 10:00 AM | 13.6913 | 6.72 |

^anew pH meter probe obtained. ISFET design.

^bafter about >4 days in Greene cell and fully oxygenated/air saturated

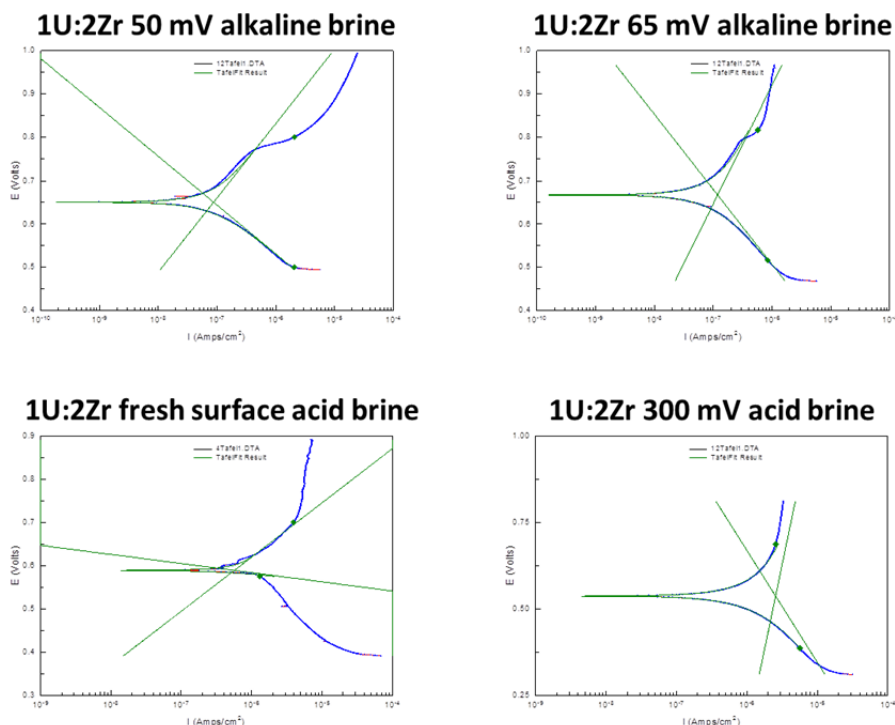
*Testing Notes:

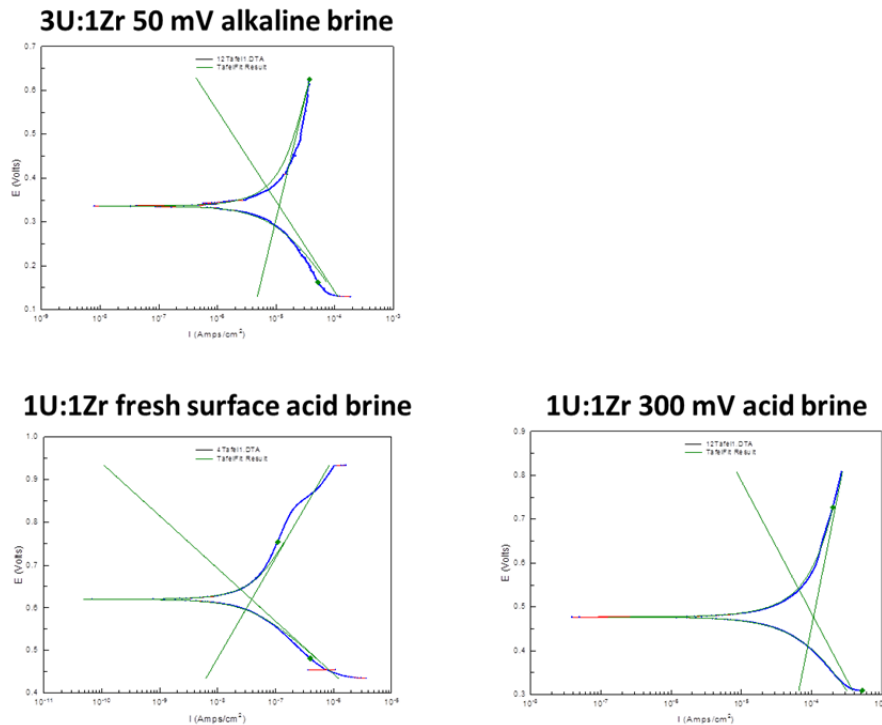
- 01/26/2015
 - Test #1 for FY15 started on 01/26/2015 at -5:30pm
 - electrodes inserted at -4:00pm, test measurements started at -5:30pm, allowing for about 1.5 hrs to stabilize in new solution after movement.
- 01/28/2015
 - EIS of 1U:2Zr had stopped due to a "bad point". The measurement error was ignored and measurement restarted
- 02/04/2015
 - stopped tests on 02/04/2015 to install a UPS

- 1U:2Zr was on (Pot1) step 4: loop(cycle 1 of 5: loop(cycle 3 of 4) substep 1 pot. Scan
- 3U:1Zr (Pot2) step 4: loop(cycle 1 of 5: loop(cycle 3 of 4) substep 1 pot. Scan
- On 08/20/2015 noted solution of 1U:1Zr cell looked brown, and potentiostat was trying potentiostatic EIS with odd looking results. Suspected shorting of wire, which was later verified between RE and WE. Short condition was removed and test allowed to proceed. Suspected all data from start of short is no good. Short likely occurred during solution sample on 08/19/2015 at 7:32 AM.
- on 09/01/2015 noted power was off from 08/27/2015 due to power interruption. Restarted tests at offset points.

Note: all Electrochemical data files were sent to ANL and the DMS for the FCRD program at INL as part of milestone M4FT-15SR0307113 Provide data in EXCEL spreadsheets to ANL for inclusion in data base.

The following is the data fits for the Tafel data:





The following is the data fits for the LPR data:

Due to the volume of data fits required for the LPR data, they have not been included here. The R_p data fits for the LPR measurements, similar to those shown for the Tafel measurements above, were included in the data sets sent to ANL and DMS for the FCRD program at INL as part of milestone M4FT-15SR0307113 *Provide data in EXCEL spreadsheets to ANL for inclusion in data base.*