SPENT FUEL MANAGEMENT AT THE SAVANNAH RIVER SITE

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ABSTRACT

Spent nuclear fuels are received from reactor sites around the world and are being stored in the L-Basin at the Savannah River Site (SRS) in Aiken, South Carolina. The predominant fuel types are research reactor fuel with aluminum-alloy cladding and aluminum-based fuel. Other fuel materials include stainless steel and Zircaloy cladding with uranium oxide fuel. Chemistry control and corrosion surveillance programs have been established and upgraded since the early 1990's to minimize corrosion degradation of the aluminum cladding materials, so as to maintain fuel integrity and minimize personnel exposure from radioactivity in the basin water. Recent activities have been initiated to support additional decades of wet storage which include fuel inspection and corrosion testing to evaluate the effects of specific water impurity species on corrosion attack.

INTRODUCTION

Basin Storage Of Spent Fuel At The Savannah River Site

Spent nuclear fuels provided by the United States Department of Energy and used in foreign and domestic research and test reactors are being accepted and stored at the Savannah River Site (SRS), now exclusively in the L-Basin. Production reactor fuels were previously stored in K-, L-, P-

Reactor basins, and the research and test reactor fuels were stored in the Receiving Basin for Offsite Fuel (RBOF) facility.

The L-Basin is a multi-million gallon structure constructed of reinforced concrete and coated with vinyl paint. The L-Basin fuel storage configuration consists of vertical and horizontal tube storage, and bucket storage. Oversize cans are used to store damaged and failed fuel and fuel pieces. Damaged or degraded cladding or structural features of a fuel assembly, if significant, can result in radiological, criticality safety, waste, and accountability issues.[1] The recent L-Basin inventory includes thousands of Materials Test Reactor Equivalent Fuel assemblies with aluminum cladding and a lesser number of assemblies with stainless steel or zircaloy cladding.

The storage experience at SRS, and experience from basin storage at many sites worldwide, shows that storage of aluminum-based fuel materials in water basins is most challenging because of the need to avoid conditions aggressive to corrosion of the aluminum. Corrosion control and surveillance activities were developed at SRS to minimize aluminum cladding corrosion degradation through water chemistry operational limits and storage configurations to avoid galvanic couple incompatibilities.

Mechanisms Of Aluminum Corrosion In Wet Storage

Identification of the mechanisms that cause corrosion provides a basis for the actions required to minimize corrosion in wet storage. The types of corrosion that can be expected for aluminum in wet storage are localized corrosion attack in the form of pitting or crevice corrosion, galvanic corrosion, and general corrosion. The aluminum cladding typically has an initial film of an aluminum oxyhydroxide due to reactor irradiation or pretreatment.

Aluminum is thermodynamically active in water, leading to the formation of oxyhydroxide films with various structures and degrees of hydration.[2-5] The formation of oxyhydroxide films is dependent on the temperature and pH of the water. Aluminum is amphoteric and its oxyhydroxides will dissolve at pH levels below about 4 and above about 10.

The chemical precursor of the crystalline oxyhydroxide film structures is a gelatinous pseudo-boehmite. The gelatinous film ages to form a tri-hydroxide with the structure of Gibbsite (hydrargillite) [γ-Al(OH)$_3$] if the pH is lower than 5.8 or higher than 9, and Bayerite [α-Al(OH)$_3$] if the pH is between 5.8 and 9 at temperatures below about 80°C. Boehmite, a monohydroxide, [γ-AlOOH] forms at temperatures above about 80°C. These passive films (Bayerite and Boehmite) are stable (limited oxide dissolution) for waters with pH levels between about 4 and 10 and Boehmite provides high resistance to continued corrosion. Autoclave treatment to form a passivating, adherent Boehmite film on the clad surfaces of the fuel assemblies has been used prior to reactor irradiation at several sites. Boehmite films also form on the cladding over the high-temperature fuel regions during reactor operation if no pretreatment is applied.

General corrosion results in a consumption of aluminum cladding. At pH conditions in which the oxyhydroxide layer is stable, the corresponding growth of the hydrated oxide layer with time in general corrosion can be expressed as:[6]

Al consumption or oxyhydroxide film growth = A + B ln(time) ……….(1),

where the coefficients A and B are dependent on the temperature and water flow rate. Using distilled, deionized water, with a conductivity of 0.71 microSiemens per centimeter (µS/cm) and low flow conditions, Draley et al.[6] determined the values of A and B for temperatures of 40, 70, and 90 °C for the weight loss of aluminum alloy 1100 in both helium-saturated and oxygen-saturated water. As an estimate using the helium-saturated coefficients at 70°C, and neglecting the dissolution of the hydrated oxide, consumption of only 2 µm of aluminum metal would be expected for a 10-year exposure. Since the initial cladding thicknesses on aluminum fuels are typically at least 250 µm

thick, cladding thinning by general corrosion is not significant to impact any desired basin storage period up to many decades.

The body of experimental work on pitting of aluminum with an oxyhydroxide film has been reviewed within a mechanistic framework for localized corrosion by Foley[7] and Szklarska-Smialowska.[8] Fundamental steps in the process of pitting attack are adsorption of reactive anions on the film, formation of soluble compounds, dissolution of the film, and direct attack of the exposed aluminum by the anion. There is substantial experimental evidence that adsorption of halides, particularly chloride, a common impurity, is the preliminary step to pitting. The aggressive anion (e.g. chloride ion) is adsorbed "competitively" with hydroxyl ions or water molecules that would promote passivity.

A category of pitting corrosion which leads to rapid attack is nodular pitting in which an occluded cell is developed at the pit. Porter and Hadden[9] reported that this form of pitting develops when calcium bicarbonate, chlorides, copper salts, and dissolved oxygen are present together in the water. Davies[10] and Rowe and Walker[11] have also reported that solutions containing soluble salts of copper, chloride, and bicarbonate have an accelerating effect on the corrosion of aluminum.

The distribution of pits in aluminum exposed to aggressive water has been observed to follow an extreme value statistical distribution in which, after initiation and development of the pit distribution, only a small percentage (~1%) continue to propagate.[12] The growth rate (pitting rate) of these deepest pits has been reported by the authors to follow a power law:[13]

$$\text{Maximum Pit Depth} = A\,(\text{time})^{1/3} \dots\dots\dots\dots\dots\dots\dots\dots(2),$$

where A, in general, depends on the alloy and water quality.

Component immersion tests were conducted in SRS basins from 1992-2000. Pitting through a 30-mil clad target tube occurred in as little as 45 days when conductivity was 180 µS/cm with a chloride level of 8 ppm (Figure 1). This was a warning sign that increased use of deionizers was necessary to eliminate aggressive water conditions.

Site experience has shown that aggressive water quality conditions can cause pitting and penetration of the claddings especially if the protective Boehmite film is ruptured;[14-16] however, fuel stored in high quality (high purity) water has not shown significant corrosion even after many years of storage. Conductivity of the basin water has been observed to be a primary factor in the incidence of pitting in the storage of aluminum-clad fuels. Galvanic couples between the cladding and stainless steel equipment used in storage have enhanced this corrosion.

Rapid, significant degradation of aluminum has not been observed and would not be anticipated in high quality (high purity) water. Chemistry monitoring and controls, in conjunction with a surveillance program, have been used at the SRS to minimize corrosion degradation during fuel storage. The specific levels of impurities and their combination in water that create conditions "aggressive" to cause pitting in aluminum over extended exposure periods have not been rigorously determined. Rather, separate effects of aggressive species from literature information and service experience have formed the basis for water chemistry control. Reference 17 provides a guide describing water chemistry conditions to avoid corrosion damage to aluminum-clad fuel.

Chemistry Monitoring and Control Program

A Basin Water Chemistry Control Program was established in the early 1990's and upgraded in 1995. The program is designed to minimize personnel exposure from radioactivity in the basin water and corrosion degradation of spent nuclear fuel in the basin. The chemistry control program establishes and maintains chemistry limits to ensure that the basin water quality is controlled to minimize initiation and progression of corrosion. The chemistry control program activities include sampling and control of basin water quality parameters such as pH, temperature, and conductivity,

and species such as chloride, mercury, iron, aluminum, microbes, and Cs-137 activity (see Table 1).

The overall water quality in L-Basin has been maintained since the early 1990's within chemistry program limits. Previous to July 1995, a portable deionizer was rotated among C-, K-, L-, and P-Reactor basins to control basin chemistry. A temporary deionizer was installed in L-Basin in mid-1995 when the basin had an initial conductivity approximately 100 µS/cm. By the end of October 1995, conductivity values decreased to 3 µS/cm and below. A dedicated deionizer system was placed in service in June 1996 to maintain this high water quality.

Recent trend data (2005-2006) for activity, conductivity and pH levels are shown in Figures 2 and 3. Basin activity levels have peaked during the recent storage period due to deionizer shutdown and dropped after startup. Figure 4 shows that prior increases in Cs-137 activity levels did not affect pH and conductivity levels over the time period from late 2002 to early 2004 when the fuels were received from RBOF and placed into L-Basin. This suggests that corrosion processes are not strongly affected by increases in Cs-137 activity levels. Water chemistry records show that the L-Basin water is being maintained within operating limits since 1996, except during a few periods of fuel transfer.

Corrosion Surveillance

A corrosion surveillance program was initiated in the L-Basin facility in 1993. Since then, the L-Basin Corrosion Surveillance Program continues to demonstrate the resistance of the fuel claddings to corrosion in the basin water and to provide assurance of continued safe, interim storage. The corrosion surveillance program monitors susceptibility to pitting, crevice, galvanic, and general corrosion. The corrosion surveillance specimens include coupons and other test specimens in the basin to provide early detection of corrosion susceptibility in the basin water environment. The corrosion specimens include sets of standard corrosion coupons (70 mm and 32 mm diameter disks), that include materials representative of the aluminum cladding materials and storage rack materials used at SRS and in the U.S origin research and test reactors fuel. Standard sets of coupons were developed through a cooperative, Department of Energy complex-wide assessment of basin corrosion and surveillance programs initiated in 1995.[16]

Recent surveillance program results show that corrosion can occur even under good water quality conditions if specimen configurations are present that may facilitate the development of localized volumes of water that may be aggressive to metals, e.g., intimate contact of dissimilar metals (galvanic corrosion) or in crevices and pits. Corrosion of the surveillance coupons is expected, and is the result of the coupon design that promotes the evolution of aggressive localized chemistry conditions.

Corrosion Surveillance Coupons

Sets of standard corrosion coupons, including coupons representative of the aluminum cladding materials and storage rack materials used at SRS and in the research and test reactors were placed in the SRS basins between 1995 and 1999. The surveillance coupons had a finish of 120 grit and were not pretreated to increase the thickness of the air-formed oxide before being placed in the basins. The use of an air-formed film provides a surface that is more susceptible to corrosion than the irradiated fuel cladding because high temperature reactor operations enhanced the protective quality of the oxide film.[15] The use of these non-irradiated coupons allows early detection of corrosion in advance of potential effects on fuel cladding and provides the opportunity to associate that corrosion with changes in the basin water chemistry. Aluminum alloy coupons of 1100, 6061, and 6063, listed in Table 2, were used to represent various fuel cladding and furniture rack alloys.

A "Ray Gun" coupon assembly that has 70 coupons is shown in Figure 4. All "Ray Gun" coupons have been evaluated except for one assembly for evaluation upon basin closure. A "Jr.

Ray Gun" coupon assembly has 36 coupons, and does not contain the 70 mm disk stainless steel coupons on the Ray Gun assembly, but does contain the 1:1 (area ratio) stainless steel to aluminum coupon galvanic couple specimens. A sketch showing the configuration of the galvanic coupons on the Ray Gun and Junior Ray Gun is shown in Figures 5 and 6. Additional specimen types in the surveillance program include "furniture rack" specimens that are flat plates and U-channel specimens of 6061-T5 and 6063-T5 (fuel storage rack) materials, respectively with R4043 weld beads which are shown hanging above the basin in Figure 7.

Evaluation Of Corrosion Specimens

Coupon assemblies have been withdrawn and the corrosion reported on an annual schedule designed to allow corrosion surveillance until the basin closes. The corrosion evaluation of the coupons consists of:

- Photographs of both sides
- Weight gains of all coupons
- Cleaning of selected coupons to remove oxides for detailed pit analysis
- Pit characterization in accordance with ASTM G-46.[19]

The reported information includes coupon photographs including pitting at a high magnification. Comparisons with past coupon surveillance photographs may be reported. The pitting density, and average (of the 10 deepest pits) and maximum pit depths are reported for the disk face regions and crevice regions. Pit depth growth rates are evaluated. Comparison of the information with the chemistry data from the storage period is reported, and the trend of data from previous coupon surveillance evaluations is reported. A recent literature paper containing details of the 2003 corrosion surveillance results is contained in Reference 19. Several of the results from the surveillance coupons from 2006 are summarized below.

Selected images of individual surveillance coupons retrieved from the basin in 2006 after 7 years immersion are shown in Figure 8. These coupons appear slightly different when visually compared to coupons pulled in 2001, 2002, 2003, and 2004. The individual coupons from all past years of evaluation are undergoing uniform corrosion with a buildup of an oxyhydroxide layer. In 2005 and 2006, the coupons had sand deposits in addition to the oxyhydroxide buildup. In mid 2004, the sand filter was started up and a cloud of sand filter fines was released to the basin. The 2005 and 2006 coupons exhibited a yellow/tan color on exposed surfaces of the horizontally mounted coupons. Deposit studies from the literature[20] have indicated increased corrosion rates due to foreign particle deposition on the surface of water immersed aluminum coupons. The particles can act as cathodes (depending on particle composition) on the surface and the surrounding metal (anode) corrodes. Thus, energy dispersive spectroscopic (EDS) analysis was performed on selected 2006 corrosion coupons prior to cleaning to confirm the existence of foreign particles. EDS confirmed that sand particles containing Si, Ti, and Fe from the sand filter release were present on aluminum coupon surfaces. In a few instances, pits were observed in the as-received coupon and locations confirmed after acid cleaning (Figure 9).

The most appreciable oxide buildup observed is in galvanically-coupled specimens[21] marked by heavy oxidation between the samples of dissimilar metals (see Figure 10). Similar behavior was noted in the previous examinations in 2001, 2003, 2004 and 2005. Pitting was apparent when the oxides were removed by the cleaning of these specimens in 16M nitric acid solution.

Pit depths were measured on all galvanically-coupled aluminum alloy coupons and summarized in Figure 11 with a display of average and maximum pit depths. Figure 11 shows a 15 mil average pit depth on a 6063 coupon and 33 mil maximum pit depth on a 6061 coupon. Both were coupled with a 304 stainless steel coupon. The pit formation is attributed to a combination of galvanic corrosion and end grain effects (6063). Similar results from the 6063 and 6061/304 coupled

coupons were observed in past coupon evaluations. Maximum pit depths on the aluminum alloy couples were located near the PTFE washer circumference and on the ID and OD of the samples.

Average and maximum growth rates for the galvanically-coupled coupons are shown in Figure 12. The average growth rate is less than 1.8 mils per year (mpy), based on immersion for approximately 7 years. The 6063 galvanic coupons show the highest maximum growth rate (~4.7 mpy). The galvanic coupons display substantially greater growth rates than the other coupon types due to electrochemical potential differences between 304 stainless steel and aluminum.

Figure 13 shows the pronounced pitting on the weld-filler material on the U-Channel specimens removed in 2003 and 2004 and cleaned to remove the oxide deposits. Average pit depths in the weld bead were less than 4.5 mils while maximum pit depths (weld toe) were less than 7 mils.

The following are general conclusions from the surveillance program from specimens removed in 2003-2006 that had been exposed to high-quality water, within the limits of Table 1:

- Uniform surface corrosion was observed on the aluminum surveillance coupons. No significant difference in general corrosion performance was observed among the aluminum alloy corrosion surveillance disk coupons (i.e., 1100, 6061 and 6063). It appears that oxide growth was initially high and has grown at a much slower rate or has stabilized.
- Pitting occurred in several regions, including the inner diameter and outer diameter circumferences (due to end grain attack), in crevices beneath the PTFE insulating washers, and in crevices between galvanically-coupled stainless steel and aluminum, between galvanically-coupled aluminum coupons, and in the weld toe and bead of the welded samples.

These results show that basin storage in low-impurity, high-quality water provides for only limited, slow corrosion of aluminum cladding and structural materials. It is emphasized that the corrosion surveillance coupons were deliberately designed to be susceptible to corrosion attack, and that the aluminum cladding materials are less susceptible to corrosion attack due to protective oxyhydroxides and limited galvanic couples in the basin storage configurations.

Basin Storage Life Extension

A strategy to extend the storage life of fuels in the SRS basins is based on the management of corrosion degradation of aluminum-clad fuels through several program elements. These are:

- Development of water quality standards to minimize corrosion of fuel and fuel storage structures
- Validation of the water quality standards through detailed examination of surveillance specimens exposed to basin conditions
- Monitoring of the basin conditions (water quality, radioactivity, physical condition), and the fuel condition in an inspection program to verify acceptable conditions are maintained
- Specification of an acceptable fuel condition (i.e., maximum extent of loss in net section and maximum release rate of radionuclides) to remain in direct basin storage. Fuels exceeding the acceptance criteria would be placed in isolation canisters
- Development of water-quality-dependent corrosion models to allow prediction of the general condition of the fuel and basin storage structures as a function of time

Several elements of this strategy have been previously implemented including ongoing corrosion specimen surveillances and monitoring of basin conditions. Additional tasks are in progress to provide the technical bases to enable additional decades of safe basin storage. These tasks are outlined below.

Chemistry Envelope Definition For Corrosion Minimization

Specifications and operating standards are in place to govern water temperature, basin level, chemistry, conductivity, and water radioactivity for the L-Basin. These standards are primarily based on empirical observations. As part of the activities for extended basin storage, an experimental program is being conducted to provide detailed technical bases for impurity levels for water quality. Water quality standards would be set within the region of water qualities which are non-aggressive to pitting or, at least, lead to a low pitting rate that would not adversely impact the fuel for a desired storage period.

Initial cyclic potentiodynamic polarization (CPP) testing was performed to identify the region of "aggressive" water qualities where corrosion attack would occur if the existing oxide layer were scratched, exposing the bare metal. The testing is based on CPP methods reported in References 22 and 23. Figure 14 shows that a 1 ppm chloride addition to deionized water should not cause pitting, whereas increases to 10 ppm, 50 ppm and 100 ppm chloride solutions clearly showed pitting attack, as verified by post-test metallography on Al 1100 coupons. Increasing chloride additions also increase current densities by up to 50X. The CPP curve in Figure 15 indicates that an addition of 10 ppm sulfate to 10 ppm chloride creates an aggressive pitting solution with current densities close to that of a 50 and 100 ppm chloride solution. However, adding 100 ppm nitrate to a 10 ppm chloride solution appears to act as an inhibitor in CPP results shown in Figure 16. Additional results not shown in this paper show that nitrate at low levels may serve as an inhibitor to aggressive solutions containing both chloride and sulphate. These CPP results exhibit a classic passive regime, a breakdown potential, and a repassivation potential. Future testing will support definition of a "chemistry envelope" for corrosion minimization based on these initial results.

Fuel Inspection Program

The L-Basin storage conditions are expected to provide for minimal degradation of the fuel. Nevertheless, periodic inspection of the wet-stored fuel provides a direct measure of the condition of the fuel with storage time. Direct visual examination of selected fuels in L-Basin over the past several years has shown that the fuel is typically in good condition, and is not degrading noticeably with time in L-Basin storage based on internal reports. The L-Basin has received many different fuel types with various cladding conditions, including through-clad damage that occurred prior to storage in the SRS basins. Aluminum-based fuel with through-clad damage with even moderate leakage is shown to be acceptable for continued direct basin storage.[1,24] Other fuel types (e.g. oxide fuel) with damage are stored in isolation canisters with J-tube vents that are effective at eliminating radioactivity release into the basin water.

An inspection program to characterize, trend, and disposition fuel condition is being planned. Specifications for acceptable fuel conditions to remain in direct basin storage will be included in the plan.

TABLE 1. L-Basin Water Quality Operating Limits

Water Quality Parameters	Operating Limit	Monitoring Frequency
pH	5.5 to 8.5	Weekly
Conductivity	10 µS/cm	Weekly
Activity	Cs-137: 500 dpm/ml	Weekly
Cu Concentration	0.1 ppm	Biannual
Hg Concentration	0.014 ppm	Biannual
Cl Concentration	0.1 ppm	Biannual
Fe	1.0 ppm	Biannual
Al	1.0 ppm	Biannual
Temperature	45°C	Weekly

TABLE 2. Composition of Al Surveillance Coupon Alloys

Al Alloy Designation	Maximum Elemental Composition, %					
	Si	Cu	Mn	Mg	Zn	Other
1100	---	0.20	0.05	0.05	0.10	1.0 Max Si+Fe
6061	0.8	0.40	0.15	1.2	0.25	0.35 Cr
6063	0.6	0.10	0.10	0.9	0.10	0.35 Fe/0.10 Cr
*R4043	6.0	0.30	0.05	0.05	0.10	0.8 Fe

- Welding rod for furniture rack samples

Figure 1. Aluminum clad full size target tubes after immersion in K-Basin.
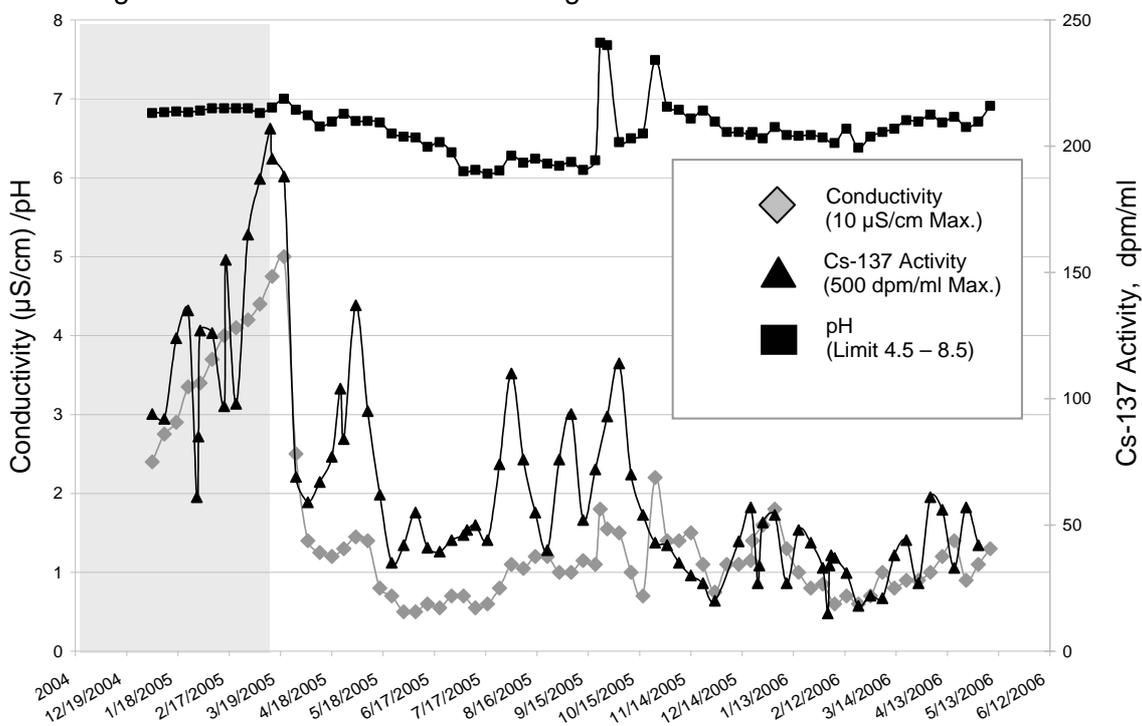
Figure 2. Cs-137 activity levels in L-Basin versus pH and conductivity during December 2004 to the middle of June 2006.
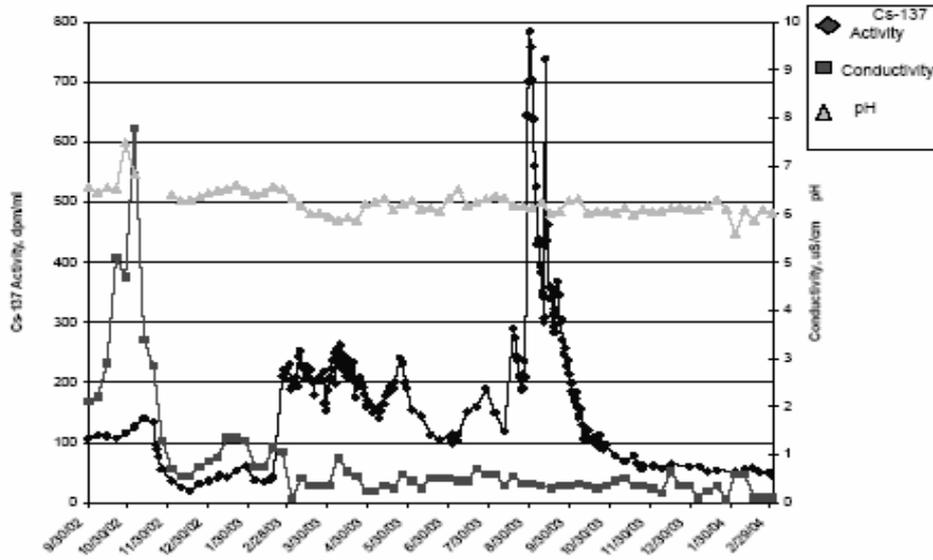
Figure 3. L-Basin Activity, Conductivity and pH from January 2003 until March 2004
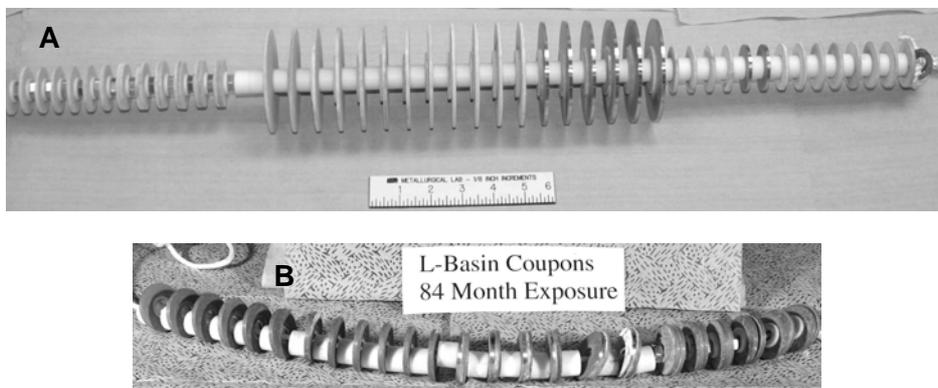
Figure 4. "Ray Gun" (A) and "Jr. Ray Gun" (B) Assemblies: As-received from L-Basin
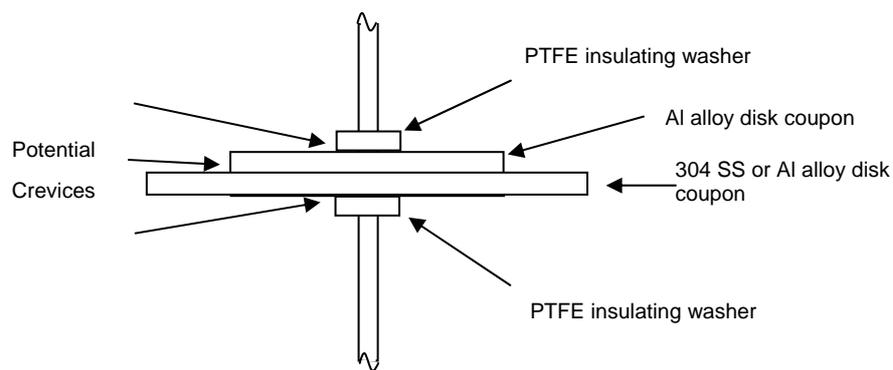
Figure 5. Sketch of Ray Gun Galvanic Coupon Arrangement. The arrangement creates a galvanic cell between the Al and a larger SS coupon (0.5:1 diameter ratio), with potential crevices between each coupon and between the coupon and each washer.
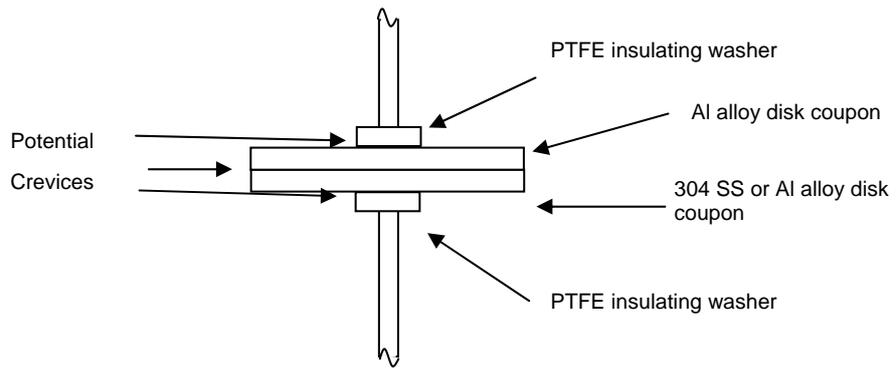
Figure 6. Sketch of Jr. Ray Gun Galvanic Coupon Arrangement. Both coupons are 32 mm in diameter, a 1:1 diameter ratio.
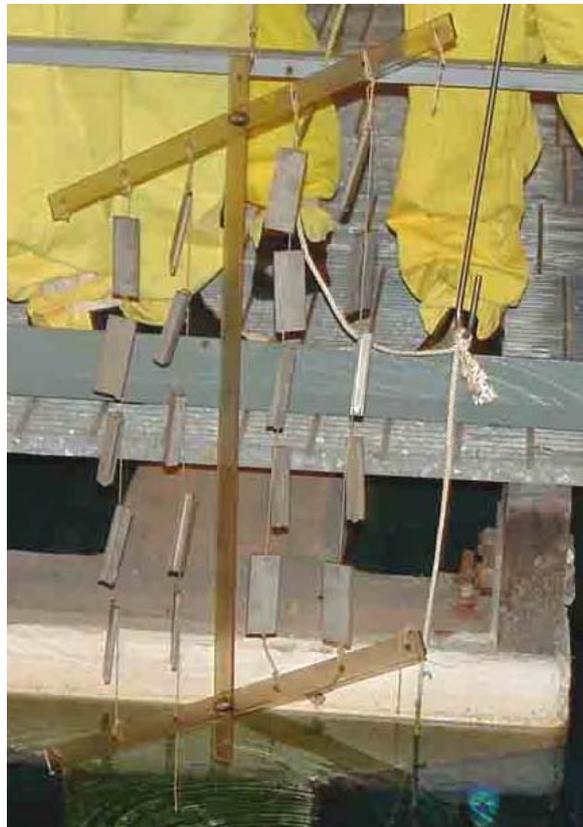
Figure 7. Hanging furniture rack coupons removed from L-Basin.

As-Removed

Nitric Acid Cleaned

1100   6061   6063
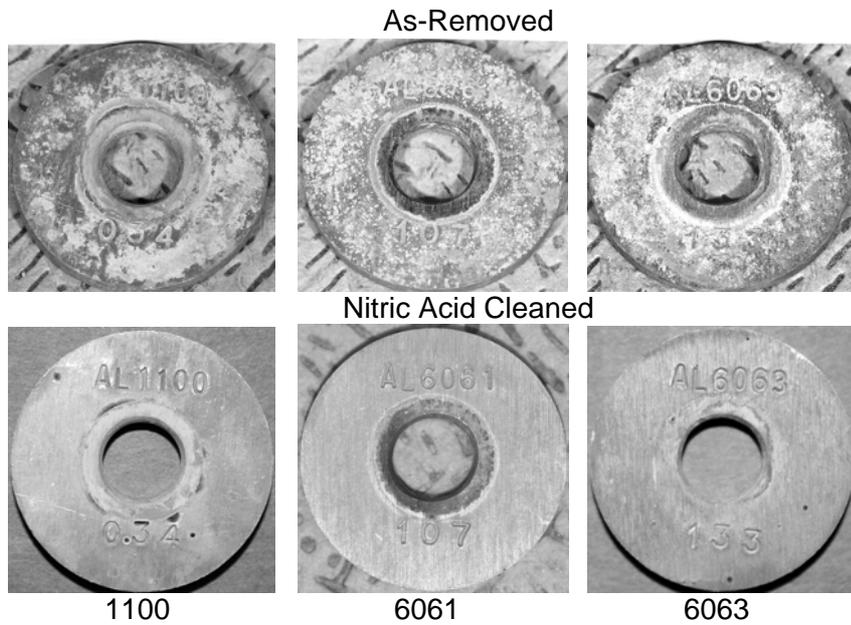
Figure 8. Selected Surveillance Coupons removed from L-Basin in 2006. The as-removed aluminum specimens exhibited a uniform oxide scale plus sand deposits and pitting attack near the crevice region created by the PTFE washer. The nitric acid cleaned coupons revealed a few large pits.

As-received Face-Up side   After cleaning w/nitric acid
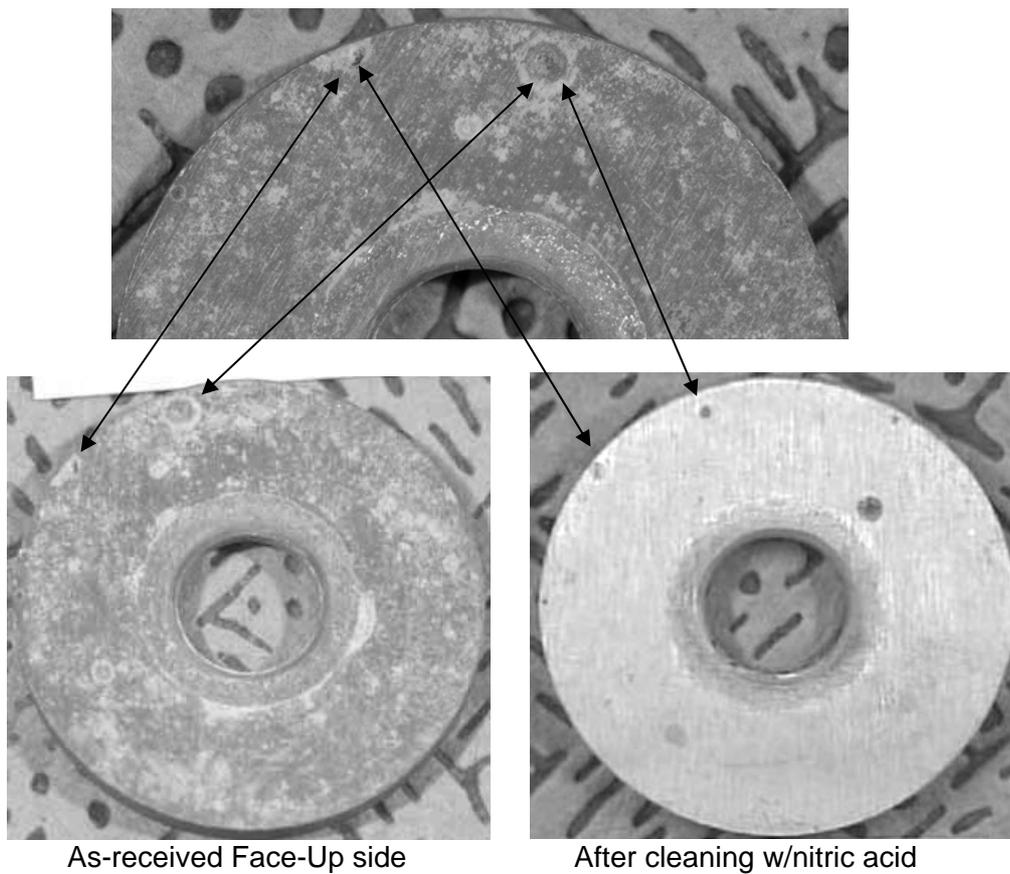
Figure 9. Comparison of individual aluminum coupon with a few deep pits shown after cleaning with the same pits visible in as-received coupon.
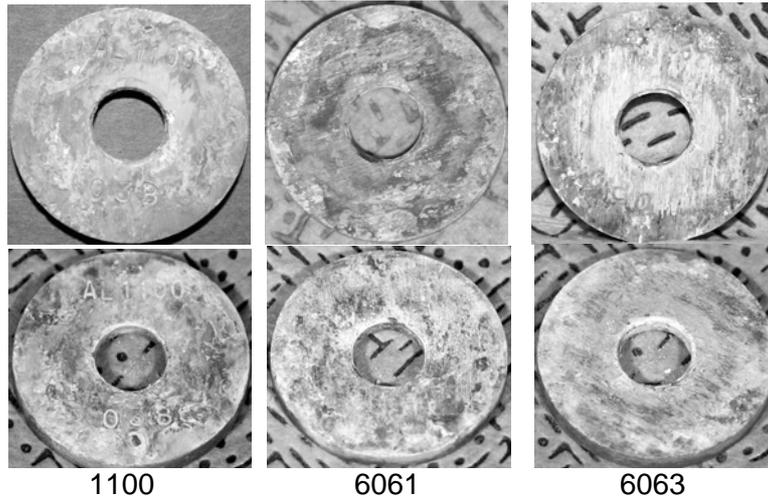
Figure 10. As-received aluminum 1100, 6061, and 6063 coupons galvanically coupled with a 304 stainless steel coupons shown with aluminum mating surfaces in view.
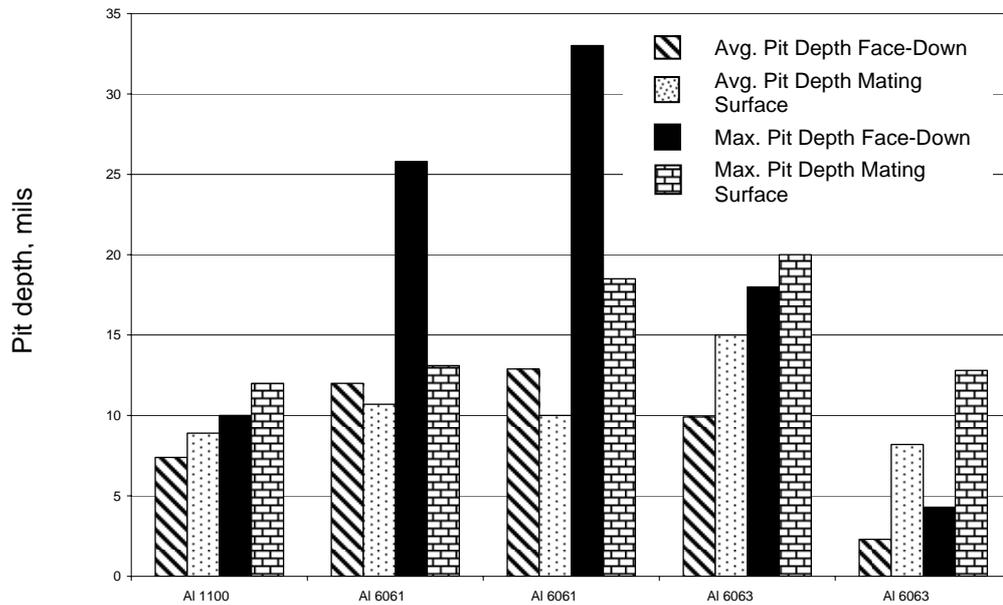

Figure 11. Average and maximum pit depths of 2006 galvanically coupled coupons. Maximum pit depths were measured near the edge of the PTFE washer area or at the coupon ID.
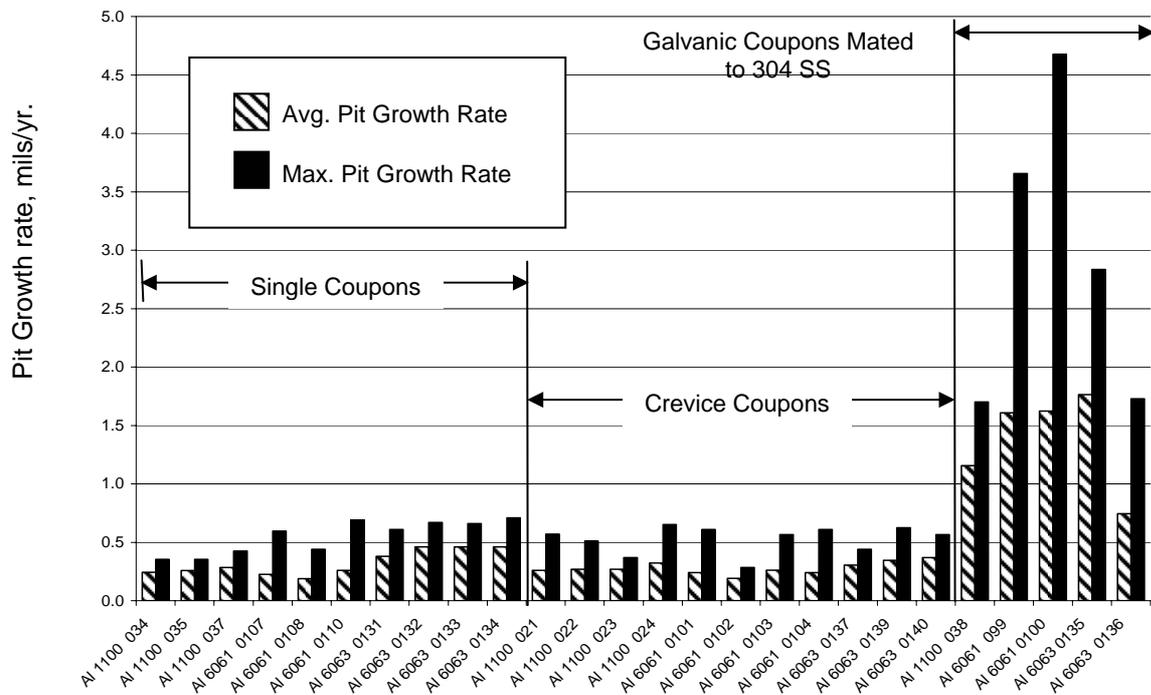
Figure 12. Average and maximum pit growth rates for single, crevice, and galvanically-coupled coupons. Coupon order and sizes are identical to those in Figure 8. The same pit depths were used with depth divided by time in basin to calculate growth rates.
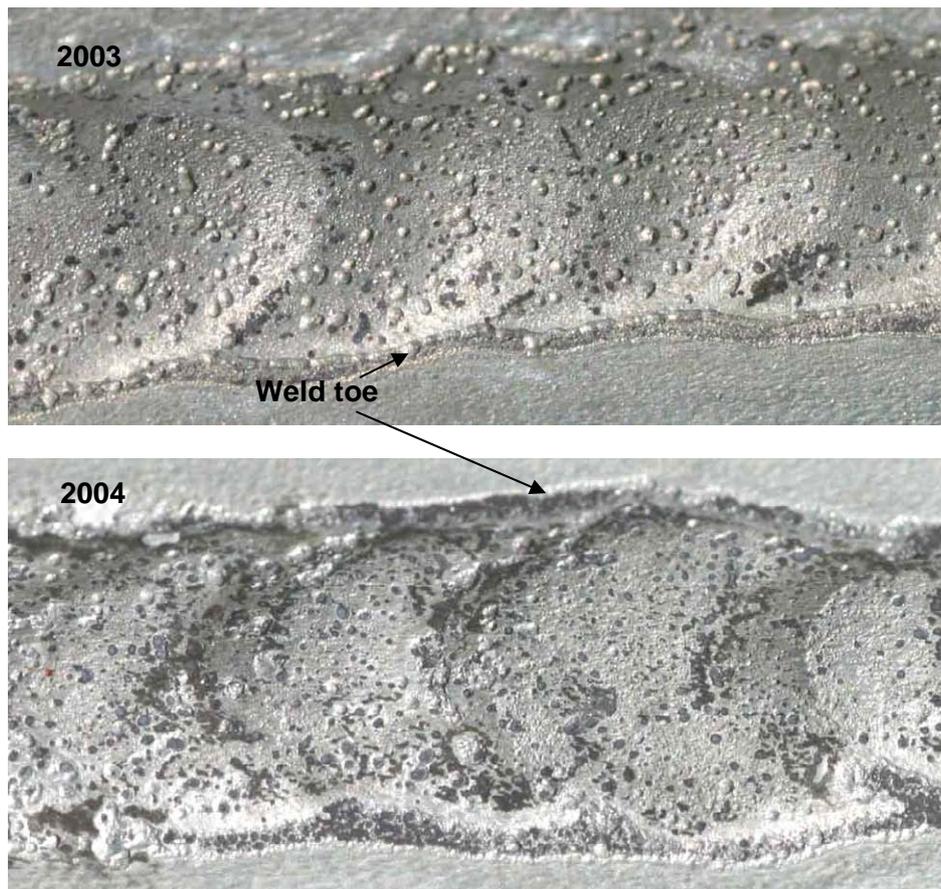

Figure 13. Cleaned R4043 weld beads on 2003 (upper) and 2004 (lower) Al 6063 T5 U-Channel samples after immersion in L-Basin for 85 and 97 months, respectively. Weld bead width is approximately 6.4 mm. Numerous pits are visible in the weld bead and weld toe on each side of bead. Few pits were found in the base metal.

Cyclic Potentiodynamic Polarization Tests - Cl⁻ Effects on Al 1100
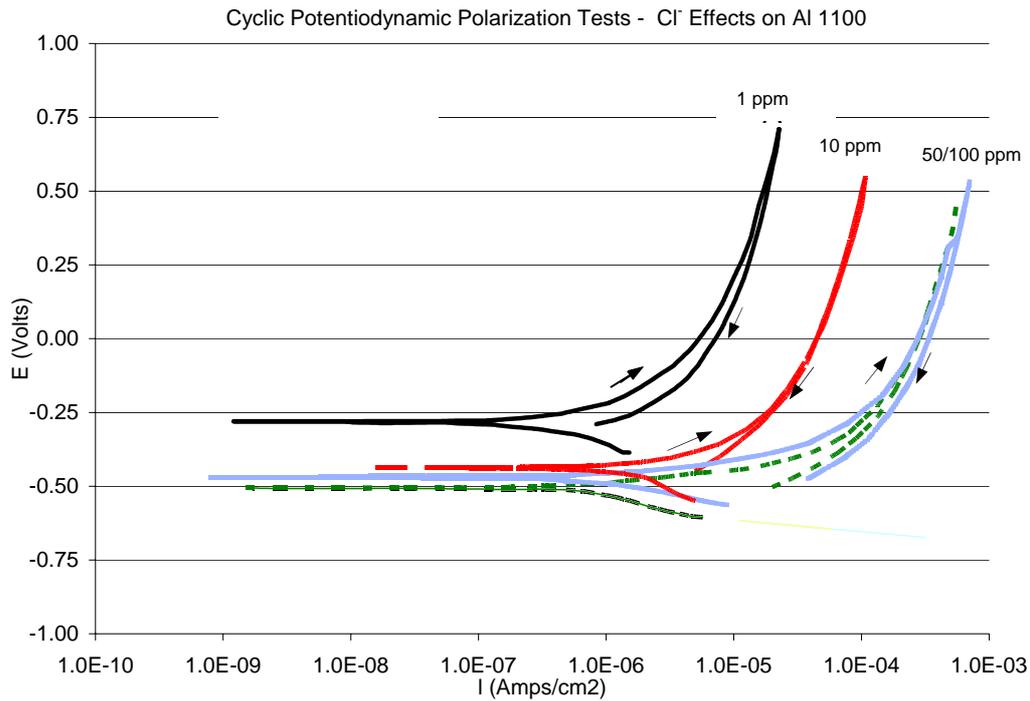
Figure 14. Cyclic Polarization Results for Al 1100 in solutions with increasing chloride levels from 1 ppm to 100 ppm. Increasing current densities is also noted as chloride levels increase.
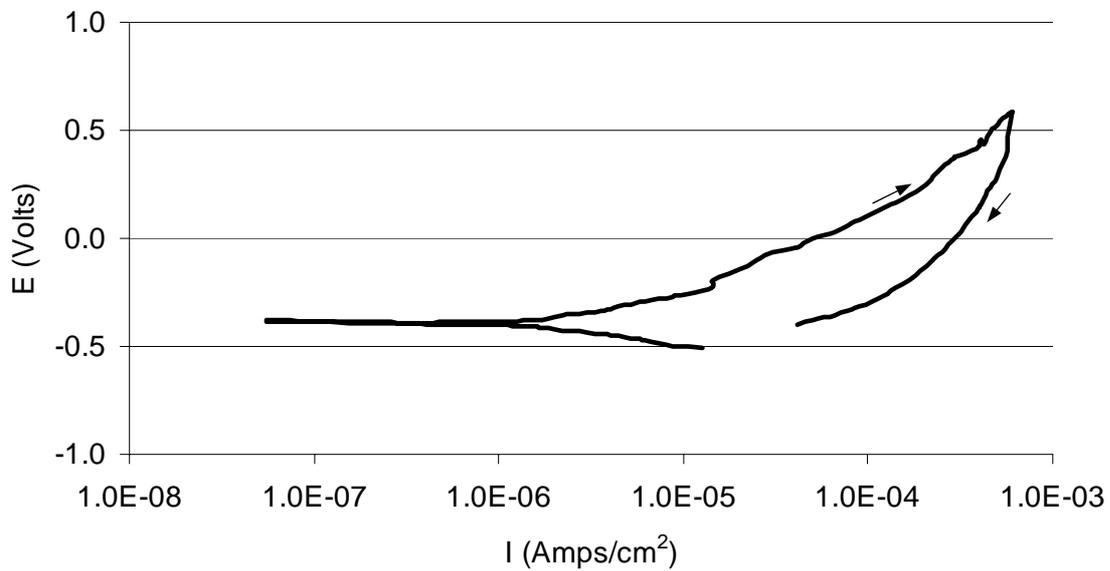

Figure 15. Cyclic Polarization Results for Al 1100 in a solution with 10 ppm Chloride plus 10 ppm Sulfate. The results show an aggressive solution (high current density).
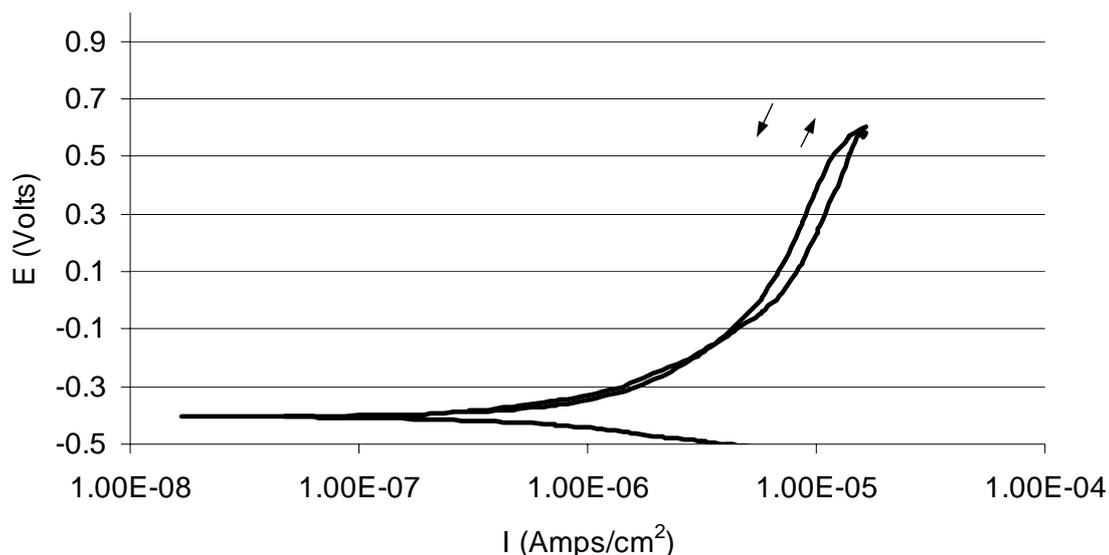
Figure 16. Cyclic Polarization Results for Al 1100 in a solution with 10 ppm Chloride plus 100 ppm Nitrate. The results show a non-aggressive solution when nitrate is added to the chloride.

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