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Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

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ELECTROCHEMICAL NOISE BASED CORROSION MONITORING: HANFORD SITE PROGRAM STATUS

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ABSTRACT

The Hanford Site near Richland, Washington has 177 underground waste tanks that store approximately 253 million liters of radioactive waste from 50 years of plutonium production. Prior to 1995 no online corrosion monitoring systems were in place at Hanford to facilitate the early detection of the onset of localized corrosion should it occur in a waste tank. Because of this, a program was started in 1995 to develop an electrochemical noise (EN) corrosion monitoring system to improve Hanford's corrosion monitoring strategy. Three systems are now installed and operating at Hanford. System design, performance history, data and the results of a recent analysis of tank vapor space data are presented.

Keywords: electrochemical noise, corrosion monitoring, nuclear waste tank, vapor space corrosion

INTRODUCTION

The Hanford Site has 177 underground waste tanks that store approximately 253 million liters of radioactive waste from 50 years of plutonium production. Twenty-eight tanks have a double shell and are constructed of welded ASTM A537-Class 1 (UNS K02400), ASTM A515-Grade 60 (UNS K02401), or ASTM A516-Grade 60 (UNS K02100) material. The inner tanks of the double-shell tanks (DSTs) were stress relieved following fabrication. One hundred and forty-nine tanks have a single shell, also constructed of welded mild steel, but not stress relieved following fabrication. Tank waste is in liquid, solid, and sludge forms. Tanks also contain a vapor space above the solid and liquid waste regions. The composition of the waste varies from tank to tank but generally has a high pH (>12) and contains sodium nitrate, sodium hydroxide, sodium nitrite, and other minor radioactive and non-radioactive constituents resulting from plutonium separation processes. Leaks began to appear in the single-shell tanks (SSTs) shortly after the introduction of nitrate-based wastes in the 1950s. Leaks are now confirmed or suspected to be present in a number of SSTs.[1] The probable modes of corrosion failures for the SSTs are nitrate induced stress corrosion cracking (SCC) and pitting.[2] No leaks have ever been confirmed in the DSTs, but a study in 1996 identified SCC and pitting as the greatest potential threats to the long-term integrity of these tanks.[3]

Corrosion monitoring and control of the DSTs at Hanford has historically been provided through a waste chemistry sampling and analysis program. In this program, waste tank corrosion is inferred by comparing waste chemistry samples taken periodically from the DSTs with the results from a series of laboratory tests done on tank steels immersed in a wide range of normal and off-normal waste chemistries.[4] This method has been effective, but is expensive, time consuming, and does not yield real-time data. In 1996, the Department of Energy Tanks Focus Area launched an effort to improve Hanford's DST corrosion monitoring strategy and to help address questions concerning the remaining useful life of these tanks. Several new methods of on-line localized corrosion monitoring were evaluated. The EN technique was selected for further study based on numerous reports that showed this technique to be the most appropriate for monitoring and identifying the onset of localized corrosion.

EN proof-of-principle tests were conducted on UNS K02400 steel specimens immersed in a variety of simulated normal and off-normal DST waste solutions at Oak Ridge National Lab (ORNL) and Pacific Northwest National Lab (PNNL) in 1996.[5] Based on these studies, a 3-channel prototype field probe was designed, constructed and deployed in DST 241-AZ-101 in August 1996.[6] Following the demonstration of the prototype for approximately a year, a longer more advanced 8-channel system was designed and installed in DST 241-AN-107 in September 1997.[7] Figure 1 shows the installation of this system. Unlike the prototype, the in-tank probe on this system reached from tank top to tank bottom exposing two channels of EN electrodes in the sludge at tank bottom, four channels in the tank supernate, and two channels in the tank vapor space. Four additional systems of similar design have been installed into other DSTs in the 241-AN tank farm.[8-11] The three most recently installed systems remain in operation at the site.

SYSTEM DESIGN

In its simplest form, the application of EN to corrosion monitoring in aqueous environments involves the immersion of electrodes made of a material of interest in an environment of interest. Typical EN monitoring systems require three nominally identical electrodes: one working electrode, one counter electrode, and one pseudo-reference electrode per channel. If the measurement environment is conductive enough (as it is in DST waste), fluctuations in current and voltage caused by the corrosion of the electrodes can be passively recorded by the EN equipment. Unlike electrical resistance (ER), linear polarization resistance (LPR), and other more traditional corrosion monitoring techniques, no external voltage or current is artificially applied to the electrodes during EN measurements. Data analysis techniques vary widely and are beyond the scope of discussion in this work. However, all available methods of analysis are dependent on the fact that different forms of corrosion create different fluctuation patterns in current and voltage. Analyses of these fluctuations reveals information about the type of corrosion occurring, specifically for Hanford, if pitting or stress corrosion cracking has initiated. Traditional uniform corrosion monitoring techniques such as ER and LPR do not offer this capability.

Measuring EN in gasseous or vapor space environments is more complicated than measuring EN in aqueous environments. If the solution resistance between the electrodes installed in a gasseous environment is infinite, as in a perfectly dry environment, it is impossible to measure any current fluctuations since this would imply an infinitely large change in the potential (i.e.: voltage) difference between the working electrodes. Similarly, there would be no potential fluctuations to measure, since no current can pass between the working and pseudo-reference electrodes. In a more realistic case where some electrical conduction through surface films is possible, measurement of EN is also theoretically possible. In this case, the key question becomes whether the resistance between the electrodes is low enough to allow something sensible to be measured. If it is, then EN can be measured. However, *what* is measured will depend on the details of the probe configuration and the instrumentation. More

specifically, it must be recognized that all measurements must account for the electrical connection through films over the probe housing, rather than directly between the electrodes, and this demands a more careful analysis on a system-by-system basis.[12]

Like most EN based corrosion-monitoring systems, the active Hanford systems monitor EN on channels composed of three nominally identical electrodes immersed in the tank waste. Each system is composed of an in-tank probe and ex-tank data collection hardware. The in-tank probe is fabricated from a ~55 foot long (~17 m) piece of 1 inch (2.5 cm) diameter stainless steel tubing. Eight three-electrode channels are distributed along the probe body. Electrodes are fabricated from UNS K02400 steel that has been heat treated to match the tank wall heat treatment. Four channels on each probe are formed from sets of bullet-shaped electrodes (25 cm^2/per electrode). Four channels are formed from sets of thick-walled C-rings (47 cm^2/electrode). Figure 2 shows two channels on the most recent 241-AN-107 probe. The unstressed bullet-shaped electrodes are used for pitting and uniform corrosion detection. The working electrode on each C-ring channel is notched, pre-cracked and stressed to yield prior to installation to facilitate the monitoring of SCC should tank chemistry conditions change to allow the onset of cracking. The other two C-rings on each C-ring channel are not stressed. Bullet and C-ring channels alternate up the length of the probe. Current DST waste levels in monitored tanks immerse three channels of bullet-shaped electrodes and three channels of C-ring electrodes.

Data collection from each in-tank probe is handled by Amulet[(1)] software from Corrosion & Condition Control, Ltd. and CIS400[(2)] hardware from Capcis Systems, Ltd. Individual 20 gauge radiation-resistant insulated conductors are attached to each electrode and lead up through the stainless steel probe body to the probe top. Belden 8162[(3)] data cable is used to transmit signal from the probe electrodes to the data collection hardware. MTL Model 755-AC[(4)] intrinsic safety barriers are used on all tank-penetrating leads. Simultaneous current and potential EN measurements are made every other second on all eight channels. The sign convention on the equipment is such that voltage values move in the negative direction as the counter/working electrode assembly becomes more anodic (corroding) than the pseudo-reference electrode (i.e.: where V_{ref} equals the potential of the pseudo-reference electrode and V_{work} equals the potential of the working/counter electrode assembly, the equipment records the measurement of $V_{ref} - V_{work}$). In addition to EN, the current system can be configured to periodically run LPR scans to obtain uniform corrosion rate information.

Because of limited riser space a variety of additional pieces of equipment have been added to the corrosion probes installed in the field. In addition to corrosion monitoring electrodes, each in-tank probe is fitted with a water lance to help facilitate its installation into the tank sludge, an array of 22 thermocouples, a movable verification thermocouple, a tank waste high level detector, ports for pressure/gas sampling, and a set of strain gauges to monitor the effects of tank operations on the downhole instrumentation. These features add a great deal of functionality to the probes, provide for a better understanding of the relationship between corrosion and other tank operating parameters, and optimize the use of risers in the tanks.

AQUEOUS DATA

The majority of EN data being collected at Hanford come from aqueous or semi-solid sludge regions of the DSTs. Data are currently being collected from the 241-AN-105, 241-AN-104 and 241-

(1) Amulet is a registered trade name of Corrosion & Condition Control, Ltd., Dingwall, Ross-Shire, England
(2) CIS400 is a registered trade name of Petroleum Research and Production, Witney, Oxfordshire, England
(3) 8162 is a registered trade name of Belden Inc., St. Louis, Missouri
(4) MTL 755-AC is a registered trade name of Measurement Technology, Ltd., Luton, Bedfordshire, England

AN-107 systems installed in the 241-AN tank farm. Data from each system are analyzed each day and downloaded and stored once each week. The vast majority of EN data from the systems are indicative of extremely low (nearly immeasurable) rates of uniform corrosion. This finding is consistent with tank design considerations and with laboratory proof-of-principle testing at PNNL at the start of the program. LPR data collected periodically from these tanks have also indicated a corrosion rate of less than 1 mpy. A typical week of EN data showing uniform corrosion from the 241-AN-107 system is shown in Figure 3. Thousands of data files similar to Figure 3 have been collected from all the active systems.

Though most data from the systems have indicated uniform corrosion, the data occasionally show sporadic EN voltage transients much larger than the background level of voltage noise, particularly on Channels 7 and 8 in the sludge layer at the bottom of the tanks. An example is shown in Figure 4. Most of the time, these discrete voltage transients have not been accompanied by simultaneous current transients. Since the laboratory development work for these systems and work by others show that pitting initiation creates numerous short-lived simultaneous spikes in current and voltage noise, the field data do not seem to indicate pitting. Similarly, the unknown transients have appeared at different times on both bullet electrode and C-ring electrode channels so transients do not seem to indicate SCC on the C-rings. Because the majority of these type transients are from electrodes near the bottom of the tank, it is possible that these transients are the result of hydrogen bubble formation and collision with the electrodes in the tank sludge or bubble formation and release from the electrode surfaces. Radiolysis in all of the tanks in the 241-AN farm produces hydrogen over time, particularly at the bottom of the tanks. However, no laboratory work has been performed to verify this hypothesis. Until such work is performed, the meaning and significance of these discrete voltage transients will likely remain unclear.

The only off-normal corrosion data that matched laboratory test data from waste simulants were recorded on the bullet electrodes on Channel 4 in the 241-AN-107 system in late December, 2001. On the morning of December 28th, the EN signature on Channel 4 began to gradually change from uniform corrosion to pitting corrosion. This behavior persisted for several weeks before returning on its own back to uniform corrosion. Several short periods of similar data have been recorded since this initial event. It is still unclear why the change in behavior took place. Figure 5 shows the week of data from December 2001 that captured the transition from uniform to pitting corrosion.

The latest 241-AN-107 system was the focus of attention in February, 2002 when approximately 37,000 gallons (140,000 L) of 19M sodium hydroxide solution was added to the tank to bring it back within the range of the site's waste tank chemistry specifications. Corrosion rates determined by running LPR on the EN electrodes in the tank prior to the addition were well under 1 mpy, but since the EN systems have not yet been officially incorporated into the site's official operating procedures, their data could not be used to impact the formal decision to make the addition. On February 20, 2002 the mixer pump in 241-AN-107 was started. Between February 21st and February 28th the sodium hydroxide solution was added to the tank. On March 5, 2002 the mixer pump was stopped. Corrosion rates were measured by LPR before, during and after the addition. For the six channels of electrodes immersed in the tank, the uniform corrosion rate prior to the caustic addition averaged 0.1 mpy. The uniform corrosion rates averaged 0.4 mpy during the addition with the pump running. The uniform corrosion rates recorded four weeks after pump stopped again averaged 0.1 mpy. During the addition, the lowest readings were on Channels 7 and 8 in the sludge. These two channels showed rates of nearly zero prior to the addition and during the addition, indicating that the sludge was not disturbed during the caustic addition. This behavior of the sludge was predicted prior to the addition. Diffusion will carry the new hydroxide into the sludge layers over time. The highest corrosion rate at any point during the addition was 1.2 mpy on Channel 4.

Although corrosion probe data could not be used to impact the decision to make this hydroxide addition, the data collected during the addition showed that corrosion monitoring could be used as a decision making tool or to monitor the impact of future hydroxide additions or other tank waste-disturbing operations.

VAPOR SPACE DATA

Although the EN technique has been successfully applied to corrosion monitoring in a wide variety of industrial environments and to aqueous nuclear waste at Hanford, ORNL, and the Savannah River Site, the technique has never been applied to nuclear waste tank vapor space environments.[13-30] Shortly after the installation of each of the Hanford probes, the EN channels in the tank vapor spaces began producing unexpected data, most often with surprisingly high current readings. High EN currents are often associated with accelerated corrosion - not an expected problem in the vapor spaces of the actively ventilated DSTs at Hanford. EN current levels reached levels high enough to corrupt data on adjacent channels on the probes by early 2002. On March 5, 2002 the vapor space channels were disconnected from system instrumentation to preserve data from the remaining supernate and sludge channels.

In 2003 efforts got underway to interpret the Hanford DST vapor space data. The initial phases of this work focused on a review of historical applications of EN to vapor space environments and the analysis that was performed on a set of vapor space electrodes removed from the original 241-AN-107 probe in 2001. Additionally, a simple DST vapor space simulant test program using Hanford style electrodes and equipment was put in place.[31]

The majority of data from the laboratory simulant tests showed little if any corrosion activity on the vapor space electrodes. Figure 6 shows a week of data from the 25 cm^2 bullet-shaped vapor space electrodes shortly after the start of testing. Figure 7 shows another week of data from the 25 cm^2 bullet-shaped vapor space electrodes near the conclusion of testing. Data collected during the balance of testing are very similar to that in Figures 6 and 7. Figures 8 and 9 show the 25 cm^2 bullet-shaped vapor space electrodes at the conclusion of testing. As the data would predict, no significant corrosion damage is visible on the exposed electrode surfaces (Figure 8) or at the electrode/gasket interfaces (Figure 9). All of these results obviously point to little or no corrosion activity on the electrodes.

Laboratory data were strikingly similar to field data produced by the vapor space electrodes on the corrosion probes in the 241-AN tank farm shortly after installation in the DSTs. Figures 10 and 11 respectively show data collected from 25 cm^2 bullet-shaped electrodes and 47 cm^2 C-ring shaped electrodes shortly after probe installation in the vapor spaces of DSTs 241-AN-104 and 241-AN-107. Data shown in Figures 10 and 11 are typical of other probe vapor space data collected shortly after installation. Both laboratory and field data indicate little or no corrosion activity in the vapor space immediately following installation.

The good correlation between laboratory and field data all but dissappears however at longer exposure times. Surprisingly, the level of current EN (ie: corrosion activity) on most of the field vapor space electrodes rose, most often sharply, within a few months of installation. Figures 12 and 13 show the sharp changes in vapor space electrode behavior in DSTs 241-AN-105 and 241-AN-107. Other tank vapor space electrodes displayed similar behavior. None of the laboratory vapor space electrodes displayed the sharp increase in current.

The increase in current on vapor space electrodes in the field has never been visibly correlated with corrosion damage on vapor space electrodes removed from service. No currently monitored vapor space electrodes (showing high current levels) have ever been removed from service and analyzed for corrosion damage. No correlation was found between the sharp changes in current seen in the field vapor space electrodes and weather related changes or tank farm activities. No sharp changes in the behavior of other probe supernate electrodes immersed in the tank coincided with the sharp changes in vapor space current.

In late 2001, vapor space electrodes were analyzed upon removal of the original 241-AN-107 probe. Pictures of a C-ring and pin-type electrode removed from this probe are shown in Figures 14 and 15. Electrodes removed from this probe show little uniform corrosion, but significant corrosion at metal/metal mating surfaces on C-ring electrodes and near the electrode gasket seating surface on other vapor space electrodes.[32] No significant corrosion product buildup was observed on electrodes immersed in the supernate or sludge in the tank. Heavy corrosion product buildup around joints and othe shared surfaces is a classic sign of crevice corrosion. Corrosion damage shown in Figures 14 and 15 could not be correlated to EN data from this probe because long analog data cables used on this system substantially corrupted all data collected from this probe (newer corrosion monitoring systems in the 241-AN farm do not use long analog data cables as a result of the data collection problems they created on the original 241-AN-107 system).[33]

Crevice corrosion demands a tight space (such as a gasket/metal or metal/metal interface) and moisture to initiate and proceed. In-tank equipment operation, waste transfers, gas release events, or waste disturbances could have caused splashing and provided the moisture necessary to induce crevice corrosion in the vapor space electrodes on the original 241-AN-107 probe (and any of the newer probes). However, condensation is a far more likely source for this moisture since the probe body is likely to be slightly cooler than the tank vapor space in which it is suspended. Once active, the accelerated corrosion at the base of a crevice is capable of producing the high current levels seen on the current probes.

No crevice corrosion (or any other form of corrosion) was observed on vapor space electrodes in the controlled laboratory environment during the recent testing at ORNL. Likewise, no laboratory vapor space electrodes produced high current levels like those seen in the 241-AN DST probes. Although the gasket seating surfaces differ in design between the original 241-AN-107 probe and the more recently installed probes, it seems likely that vapor space electrodes in tank farm service are suffering from condensation induced crevice corrosion at tight metal-to-metal or gasket-to-metal interfaces.

Hanford DST domes and upper wall surfaces are primarily formed from welded steel plates. The bulk of the steel plate surfaces are inherently smooth and crevice-free. Because crevice corrosion appears to be active in crevices on the vapor space EN electrodes, data collected from these electrodes are *not* likely to be applicable to the large flat metal surfaces that make up the bulk of the DST domes and upper walls (by definition crevice corrosion cannot exist on large bare flat metal surfaces). The data do however indicate that conditions in the DST vapor spaces are conducive to accelerated crevice corrosion at creviced areas in the tank vapor space (overlapping joints, riser interfaces, equipment penetrations, etc.). For verification, one of the new EN probes that has produced high vapor space elctrode currents should be removed or temporarily lifted out of the tank to verify the existence of crevice corrosion on the vapor space electrodes.

CONCLUSIONS

The performance of the three corrosion monitoring systems currently in operation at the Hanford Site has demonstrated that the EN technique can be effectively applied to the study of nuclear waste environments. The systems at Hanford have shown that uniform corrosion is the most dominant active form of corrosion in the DSTs. They have also captured data that indicate that pitting corrosion is possible in the tank supernate regions and that crevice corrosion could be active at crevices in the tank vapor space region under high humidity conditions. Moreover, they've demonstrated their usefulness in monitoring the impact of tank waste disturbing activities such as hydroxide additions. It appears that with continued use and some refinements in data interpretation, the EN based corrosion monitoring systems could become a valuable tank integrity management tool for nuclear waste tanks, particularly as these tanks meet or exceed their design life.

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TABLE 1
WASTE SIMULANT COMPOSITION

Anion	Concentration (M)
Nitrite	2
Nitrate	4
Chloride	0.02
Hydroxide	3

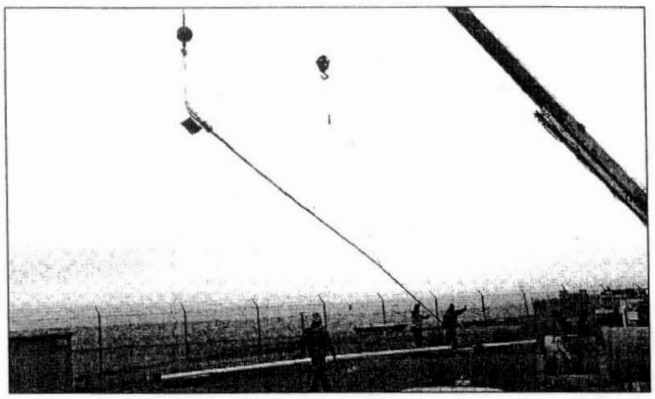

FIGURE 1 - Installation of first full-scale probe into DST 241-AN-107

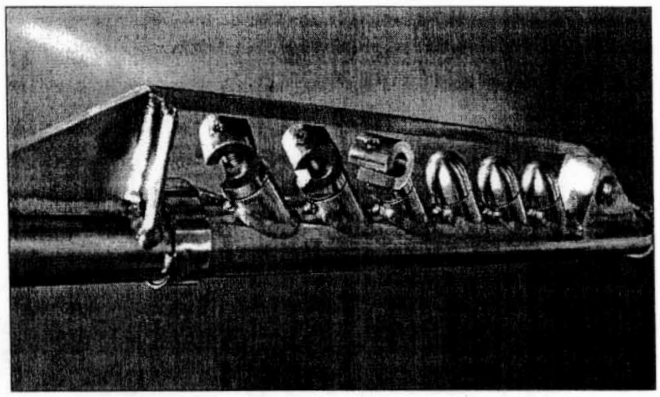

FIGURE 2 - Detail of 25 cm^2 bullet and 47 cm^2 C-ring channel electrodes

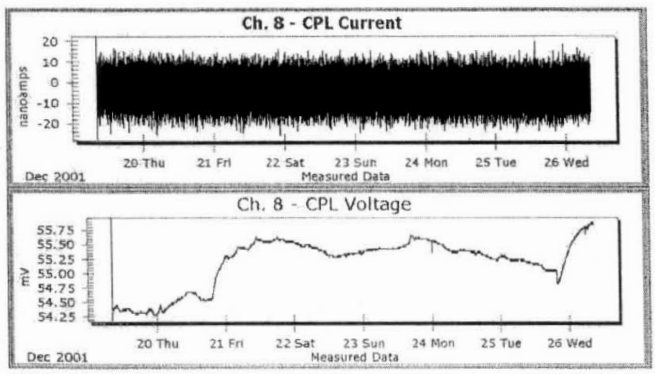

FIGURE 3 - Typical week of EN data showing uniform corrosion in DST 241-AN-107

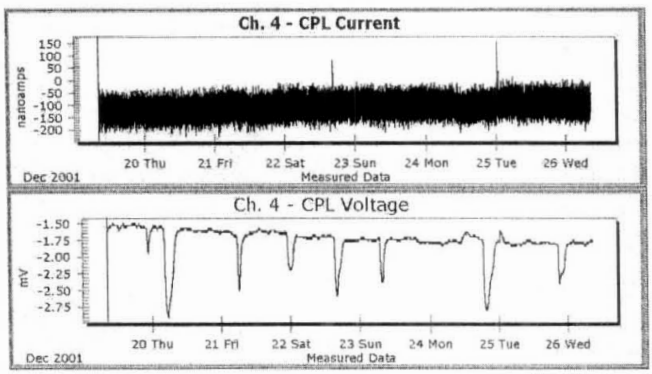

FIGURE 4 - Week of EN data showing unknown transients in DST 241-AN-105

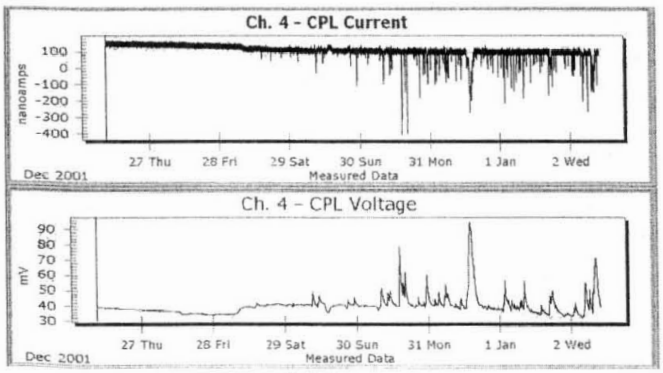

FIGURE 5 - Transition from uniform corrosion to pitting on Channel 4 in DST 241-AN-107

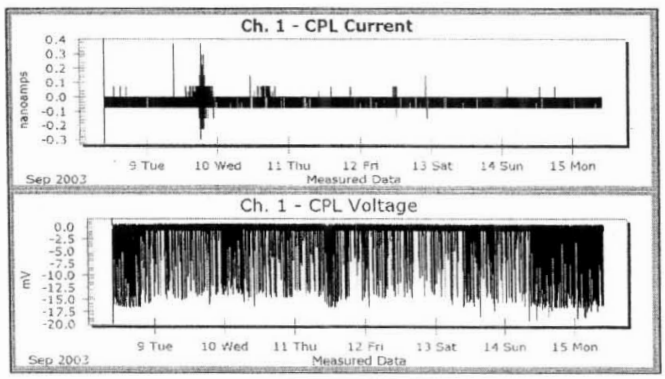

FIGURE 6 - Low corrosion activity at start of testing on 25 cm^2 lab vapor space electrodes

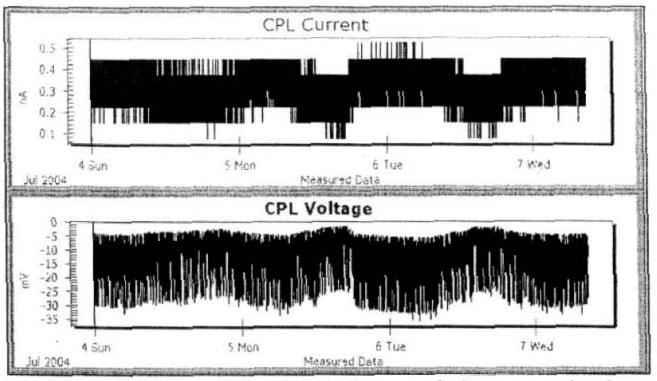

FIGURE 7 - Low corrosion activity at end of testing on 25 cm^2 lab vapor space electrodes

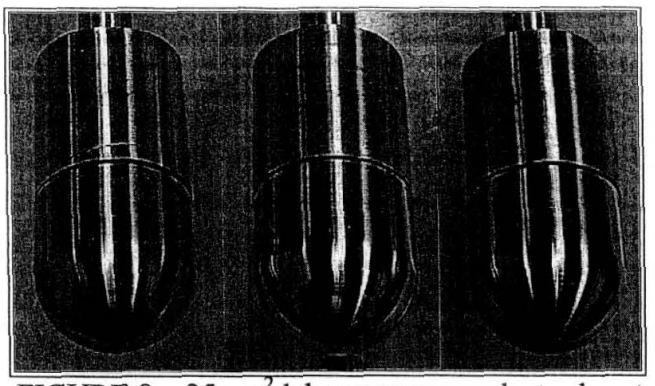

FIGURE 8 – 25 cm^2 lab vapor space electrodes at conclusion of testing showing no damage

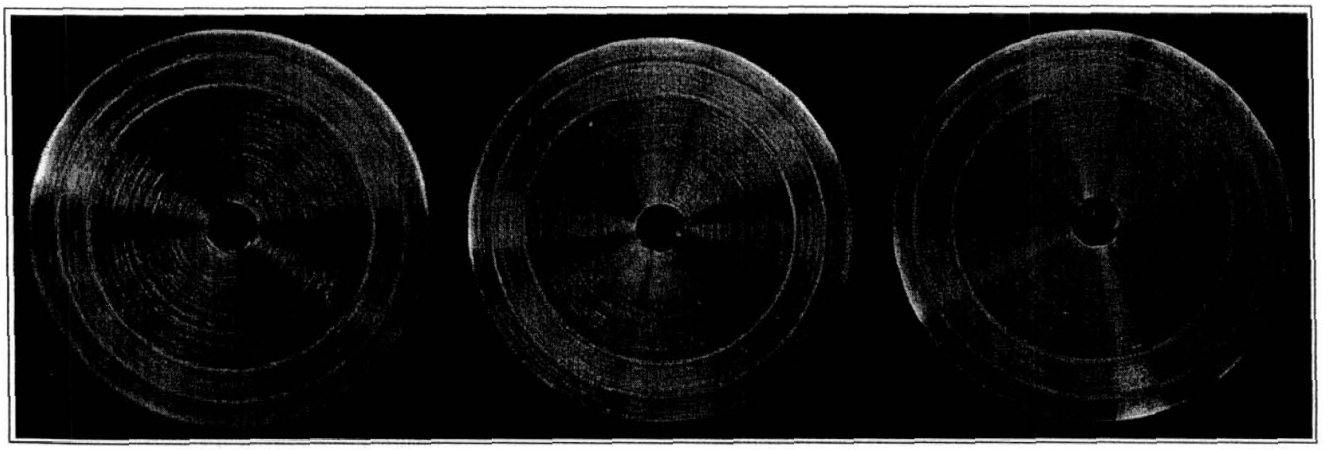

FIGURE 9 - Gasket/electrode interfaces on 25 cm^2 lab vapor space electrodes at conclusion of testing

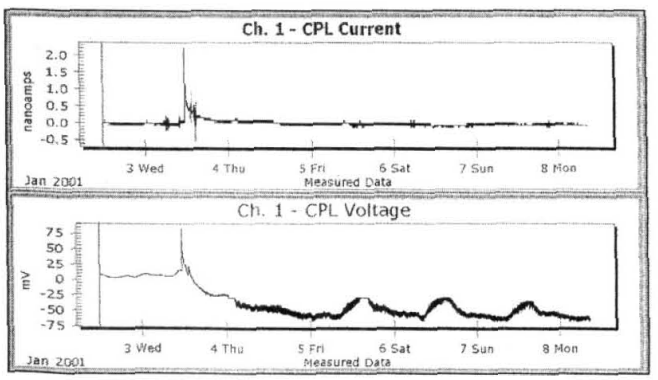

FIGURE 10 - Low corrosion activity on 25 cm^2 vapor space electrodes shortly after installation in DST 241-AN-104 (install at 01/03/01 12:00)

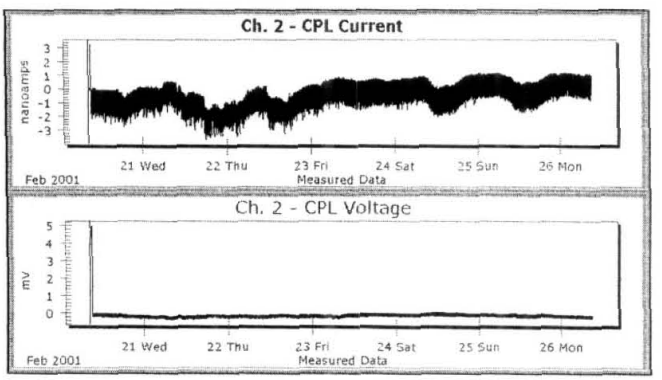

FIGURE 11 - Low corrosion activity on 47 cm^2 vapor space electrodes shortly after installation in DST 241-AN-107 (install right at scan start)

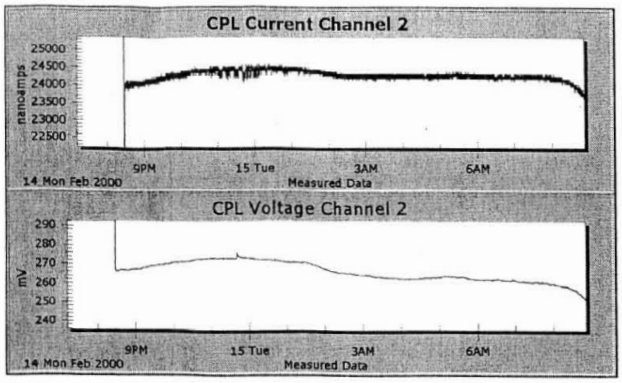

FIGURE 12 - Sharp rise in current EN data on the 47 cm^2 C-ring shaped vapor space electrodes on the 241-AN-105 probe 1 month after install

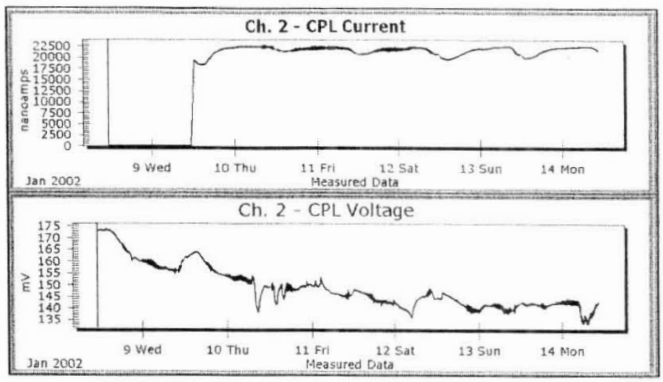

FIGURE 13 - Sharp rise in current EN data on the 47 cm^2 C-ring shaped vapor space electrodes on the 241-AN-107 probe 11 months after install

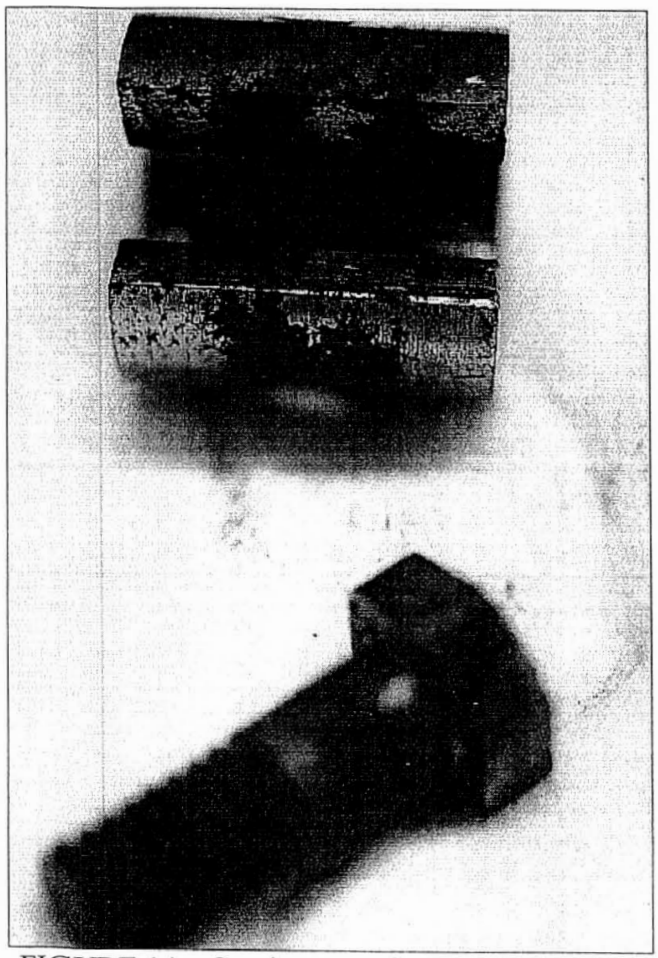

FIGURE 14 – Crevice corrosion on original 241-AN-107 probe C-ring electrode after exposure to DST vapor space

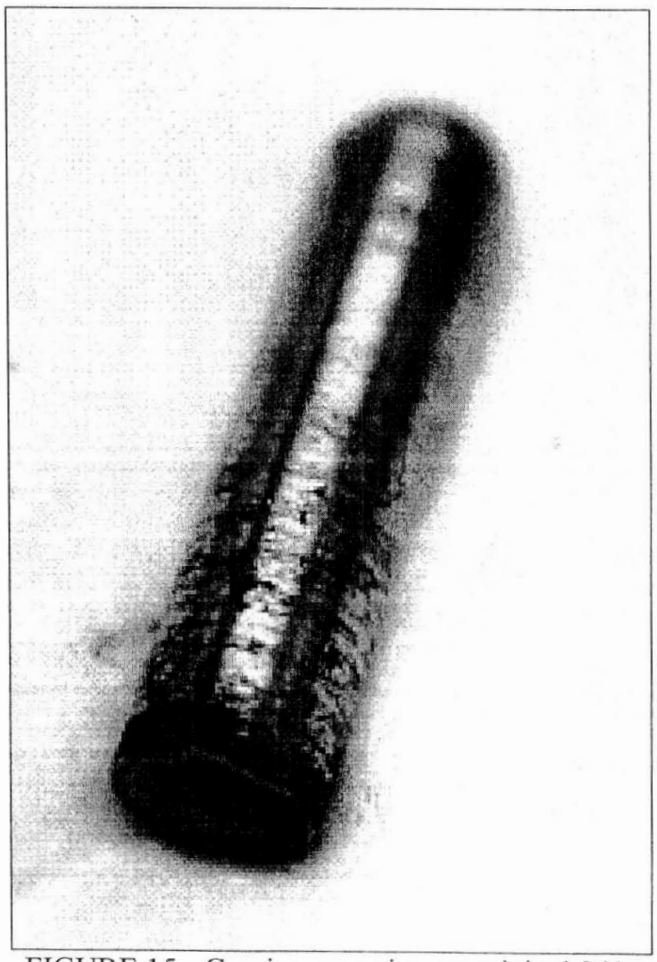

FIGURE 15 - Crevice corrosion on original 241-AN-107 probe pin-type electrode after exposure to DST vapor space