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Hanford Prototype Corrosion Probe Operational Experience

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HANFORD PROTOTYPE CORROSION PROBE OPERATIONAL EXPERIENCE

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ABSTRACT

High-level nuclear wastes at the Hanford Site are stored underground in carbon steel double-shell and single-shell tanks. The installation of a prototype corrosion monitoring system into double-shell tank 241-AZ-101 was completed in August, 1996. The system monitors fluctuations in corrosion current and potential (electrochemical noise) occurring on three electrode arrays immersed in the waste liquid and in the vapor space above the waste. The system also supports the use of Tafel and linear polarization resistance testing. By monitoring and analyzing the data from these techniques, changes in the corrosive characteristics of the waste have been rapidly detected and correlated with operational changes in the tank.

Keywords:

electrochemical noise, pitting, stress corrosion cracking, localized corrosion, nuclear waste tank corrosion, mild steel, nitrates, corrosion surveillance, and corrosion-monitoring.

INTRODUCTION AND BACKGROUND

The Hanford Site has 177 underground waste tanks that store approximately 253 million liters of radioactive waste from 50 years of weapons production.[1] Twenty-eight (28) tanks have a double-shell and are constructed of welded ASTM A537-Class 1 (UNS K02400), ASTM A515-Grade 70 (UNS K03101), or ASTM A516-Grade 60 (UNS K02100) material. The inner tanks of the double-shell tanks were stress relieved following fabrication. One hundred and forty-nine (149) tanks have a single-shell and were 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 constituents resulting from plutonium separation processes.[1-4]

Leaks began to appear in the single-shell tanks shortly after the introduction of untreated nitrate-based wastes in the 1950s. Leaks are now suspected to be present in a significant number of single-shell tanks.[1] The probable modes of corrosion failures are reported as nitrate stress corrosion cracking (SCC) and pitting.[2]

Previous efforts to monitor internal corrosion of waste tank systems have included linear polarization resistance (LPR) and electrical resistance techniques.[5-6] These techniques are most effective for monitoring uniform corrosion, but are not well-suited for the detection of localized corrosion (pitting and SCC). The characterization of electrochemical noise (EN) for monitoring waste tank corrosion was investigated by the Savannah River Site, but the tests were not conclusive.[7]

For many years, EN has been observed during corrosion and other electrochemical reactions, and the phenomenon is well established.[8-19] Typically, EN consists of low frequency (< 1 Hz) and small amplitude signals that are spontaneously generated by electrochemical reactions occurring at corroding or other surfaces.[20] Laboratory studies and recent reports on field applications have reported that EN analysis is well suited for monitoring and identifying the onset of localized corrosion, and for measuring uniform corrosion rates.[14-27] A two year laboratory study was undertaken to provide a technical basis for using EN in nuclear waste systems.[28]

A typical EN based corrosion-monitoring system measures instantaneous fluctuations in corrosion current and potential between nominally identical electrodes of the material of interest in the test environment of interest. Time-dependent fluctuations in corrosion current are described by electrochemical current noise, and time-dependent fluctuations of corrosion potential are described by electrochemical potential noise. It has been shown that each type of corrosion phenomenon presents a unique relationship between corrosion current and potential transients in the temporal data.[14-28] Other methods of analysis involve transforming temporal data to the frequency domain for analysis.[29]

A prototype corrosion-monitoring system using EN as a monitoring technique was developed and deployed into double shell tank 241-AZ-101 at the Hanford Site in August, 1996. The system is designed to detect the onset of localized corrosion phenomena if tank conditions should change to allow these phenomena to occur. The system also facilitates the use of the Tafel and LPR techniques to generate uniform corrosion rate information. This paper summarizes the design, development and preliminary data collected from the prototype probe during the first 1300 hours of operation.

SYSTEM DESIGN AND CONFIGURATION

The following design goals were established for the prototype probe system:

- Provide at least two years of service
- Fit through 10 cm diameter riser
- Facilitate remote data acquisition and system control
- Facilitate decontamination by minimizing liquid retention

The probe system was designed to operate under the following conditions:

- Temperature ranges up to 100°C
- Liquid phase pH ranges from 7 to 14
- Radiation levels up to 1000 R/hr
- Liquid phase flow rates up to 1.0 m/s

The prototype probe is installed in double-shell tank 241-AZ-101, located in the AZ tank farm of the 200 East area of the Hanford Site. The primary liner of the tank is constructed of ASTM A515-Grade 60 carbon steel (UNS K02401). This tank went into service in 1976. It was constructed to store neutralized current acid waste, a high-level waste produced from the reprocessing of irradiated nuclear fuels in the Plutonium Uranium Extraction plant (PUREX). The tank first received water in late in 1976. During the next seven years, the tank received a mixture of evaporator feed, double-shell slurry feed, complexed, noncomplexed, and dilute noncomplexed waste from PUREX and other miscellaneous streams.[30] From 1984 through 1986, the tank received aging waste from the PUREX plant exclusively. The remainder of the fill history of tank 242-AZ-101 consisted of a series of small additions of water and dilute noncomplexed wastes from other double-shell tanks.[30]

As of August 1996, 241-AZ-101 contained a total of 3.2 million liters of liquid supernate waste and sludge. This volume fills the tank to an approximate depth of 782 cm. This fill level comprises approximately 33 cm of solid sludge at the bottom of the tank, with the remainder as liquid supernate. Waste chemistry and operating conditions are shown in Tables 1 and 2.

A schematic diagram of the corrosion monitoring system is shown in Figure 1. The prototype corrosion probe was constructed from 3.8 cm diameter AISI 304L stainless steel (UNS 30403) pipe for extended service in the waste tank environment. It was designed to fit through a nominal 10.2 cm diameter tank riser and extends approximately 10 meters into the tank.

The probe was designed with electrode arrays at the 4.0 m, 8.8 m, and 10.1 m depths. The upper two electrode arrays (Arrays A and B) were positioned to monitor vapor phase corrosion, while the lower most array (Array C) was immersed approximately 38 cm into the liquid waste. The probe tree assembly is shown in Figure 2.

Each electrode array consisted of three nominally identical C-ring (ASTM G-38) electrodes constructed of archived ASTM A537-Class 1 (UNS K02400) tank steel to best represent actual tank wall material (Figure 3). The center electrode in each array was pre-cracked by cyclic fatigue and strained beyond the proportional limit just prior to immersion in the waste. The other two electrodes were not strained.

A 10 conductor shielded data cable extends 10 m through the interior of the sealed pipe. At each electrode array, three conductors are removed from the shielding and attached to the three electrodes, one per electrode. A commercially available glass to metal seal is used to penetrate the probe tree pipe wall while maintaining electrical isolation between the electrodes and the pipe wall. Electrodes are isolated from the glass to metal seal through the use of an ethylene propylene (EPDM) gasket suitable for use in radiation environments. The shielded cable terminates at the top to the probe tree assembly in a weather tight box. Approximately 24 meters of above ground cable runs from the top of the probe to the corrosion monitoring instrumentation. The above ground cabling is shielded from electrical interference by way of driven Faraday shields emanating from the corrosion monitoring instrumentation.

The corrosion monitoring instrumentation utilizes a Gamry Instruments CMS100 electrochemical measurement system with an eight channel multiplexer and associated software.(1) Operating software scripts for this system were customized and optimized for EN data collection. Data were collected sequentially on the electrode arrays using three channels of the multiplexer. Potential and current measurements were recorded once per second on the active array. Data was typically

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(1) Gamry and CMS100 are trademarks of Gamry Instruments, Inc., Willow Grove, Pennsylvania.

collected for 500 - 600 seconds per channel. Post-collection data analysis was performed using internally developed statistical analysis software and Microsoft Excel.(2)

PRELIMINARY RESULTS AND DISCUSSION

The prototype probe was installed on August 1, 1996. Data acquisition began after electronics package installation on August 8, 1996.

Liquid Array Data

Statistical summary data for the first 1300 hours after probe installation are shown in Figures 4 through 6. Mean current (Fig. 4) ranged from 1 to 3 micro amperes (μA) and generally decreased over time.

The effect of raw water additions during three thermocouple tree removal operations can be clearly seen at approximately 325, 980, and 1225 hours. The effect of a Tafel scan can be seen at approximately 1040 hours.

Mean current decrease during the water additions is thought to be due to waste oxygenation and mixing causing a temporary increase in passive film thickness (that is, water injection from the top of the tank forces high hydroxide waste from deeper within the tank to mix with lower hydroxide waste near surface).

The initial potential difference between the stressed and unstressed C-ring electrodes was approximately 10 millivolts (mV), with the stressed electrode anodic with respect to the unstressed electrode. In general, mean potential difference (Figure 5) decreased steadily over time. The effect of the raw water additions and Tafel scan are also evident.

Resistance noise (R_n) is defined as the ratio of the standard deviations of potential and current between two working electrodes and has been reported to be equivalent to polarization resistance (R_p).[31] Trends in R_n over the first 1300 hours are shown in Figure 6 along with an average value of R_p calculated from eight LPR tests conducted periodically during operation. The magnitude of R_n and fluctuations in R_n decreased steadily over time. Average R_n values were approximately 15,000 ohm-cm^2. The effect of the raw water additions and Tafel scan are also evident.

Values of R_p from LPR tests were approximately four times greater (56,000 ohm-cm^2) than the measured R_n values. Tafel slopes measured in the tank waste were $b_c = 46$ and $b_a = 214$ mV/decade which corresponds to a general corrosion rate of less than 0.2 mils per year. These data were consistent with previous laboratory results in simulated waste solutions.[32]

Raw EN data files from approximately 200, 350, 950 and 1100 hours (before and after the first two raw water additions), are shown in Figures 7 through 10. Current and potential data were

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characteristic of low rates of general corrosion and showed no characteristics of localized corrosion.

Vapor Space Array Data

Data from the vapor space arrays were inconclusive with respect to both EN and LPR due to the low relative humidity. There appears to be insufficient electrolyte to support corrosion monitoring using the installed equipment. Current and potential fluctuated randomly near the limit of detection for the equipment. Measured R_p values were greater than 65,000,000 ohm-cm^2. Redesign of the vapor space electrodes or further software optimization may permit collection of useful corrosion information from the tank vapor space.

SUMMARY AND CONCLUSIONS

A prototype corrosion monitoring instrument tree was installed into a double-shell waste tank at the Hanford Site. The instrument tree monitors transients in corrosion current and corrosion potential occurring on three electrode arrays immersed in the waste liquid and in the vapor space above the waste. By monitoring these transients, changes in the corrosive characteristics of the waste have been rapidly detected and correlated with operational changes in the tank.

EN data collected during the first 1300 hours indicated that uniform corrosion was the dominant active corrosion mechanism. No characteristic current and voltage transients indicative of localized corrosion were observed. The effects of raw water additions during thermocouple tree removal operations were also observed.

Tafel slopes measured in the waste solutions were $b_c = 46$ and $b_a = 214$ mV/decade. Polarization resistance, R_p, values were on the order of 56,000 ohms-cm^2 and were approximately four times higher than resistance noise, R_n, values. These results were consistent with previous laboratory studies.

General corrosion rates calculated from LPR data were less than 0.2 mpy.

Data collected from the Hanford Site prototype corrosion probe indicates that an EN/LPR based corrosion monitoring system can be successfully applied to nuclear waste tank environments

ACKNOWLEDGMENTS

This work represents the cooperative efforts of many people and organizations from diverse backgrounds and ranges of interest. The tests were conducted over the course of two years by personnel from the Materials and Corrosion Engineering group within Westinghouse Hanford Company, the Materials Development group of the Structural Materials Research Section at Pacific Northwest National Laboratory. The technical support of Dr. Mike Danielson of Pacific Northwest National Laboratory, Mr. Ivar Husa of Westinghouse Hanford Company and Mr. Matt Rhodes of Gamry Instruments Incorporated is also gratefully acknowledged.

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Table 1: Supernate Composition of 241-AZ-101 Waste

Component	**1995 Supernate Sample**
Cations	moles per liter
Al^{+3}	0.34
Na^{+}	4.14
Anions	
Cl^{-}	0.0049
F^{-}	0.083
OH^{-}	0.67
NO_3^{-}	1.06
NO_2^{-}	1.20
PO_4^{-3}	0.014
SO_4^{-2}	0.160
Carbon	
Total inorganic carbon	0.48
Total organic carbon	0.086
Radionuclides	microcuries per milliliter
^{137}Cs	1590
$^{89/90}Sr$	1.20

Table 2: Operating Conditions in 241-AZ-101

Parameter	**Supernate**	**Vapor**
Temperature, °C	60 avg - 72 max	15-30
Humidity, %RH	--	60-100
Radiation dose rate, R/hr	900	250-400
pH	13.6	--
Density, g/ml	1.2	--

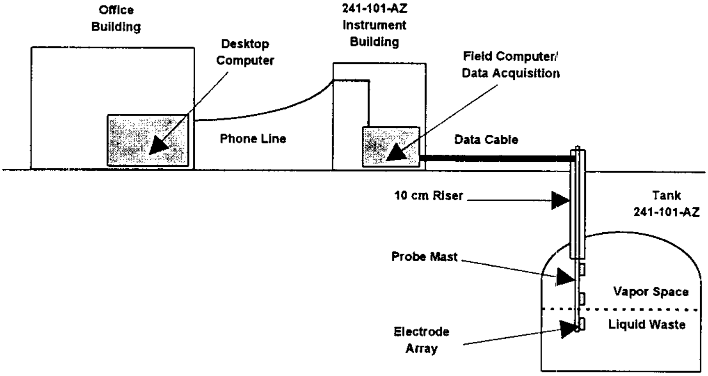

Figure 1: Schematic of Hanford Site prototype corrosion monitoring system with remote operation capability

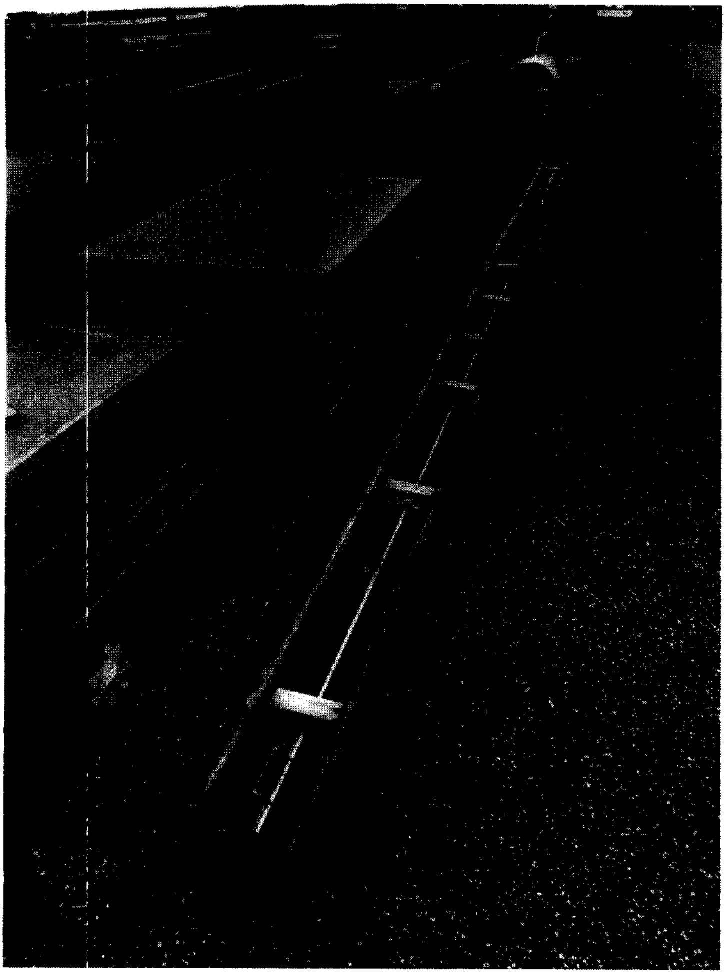

Figure 2: Prototype probe in lay-up yard prior to installation in double shell tank 241-AZ-101

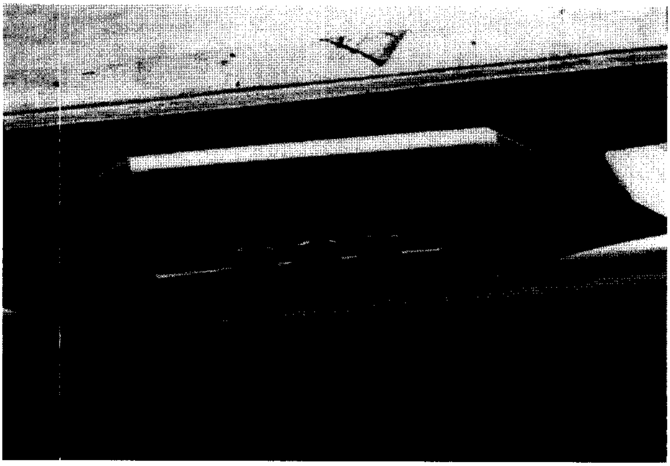

Figure 3: Close-up of C-ring electrode array on prototype probe. Center electrode is pre-cracked by cyclic fatigue then strained beyond the proportional limit (near yield) prior to installation.

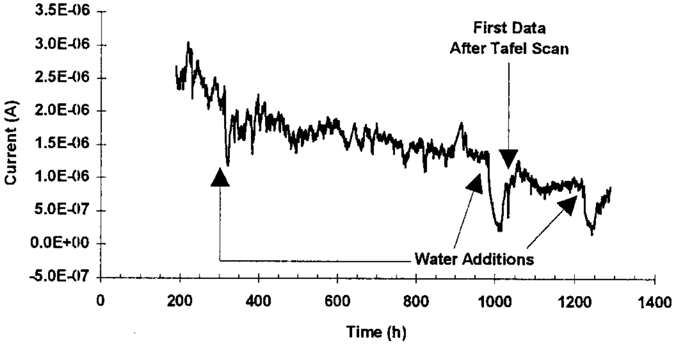

Figure 4: Mean current during first 1300 hours of operation. First ~200 hours unrecorded due to delay in data acquisition system installation following probe installation.

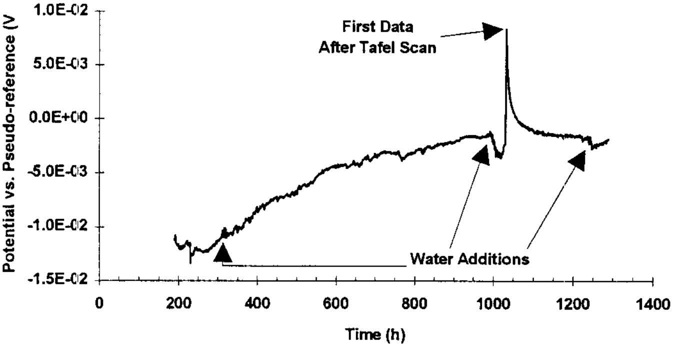

Figure 5: Mean potential during first 1300 hours of operation. First ~200 hours unrecorded due to delay in data acquisition system installation following probe installation.

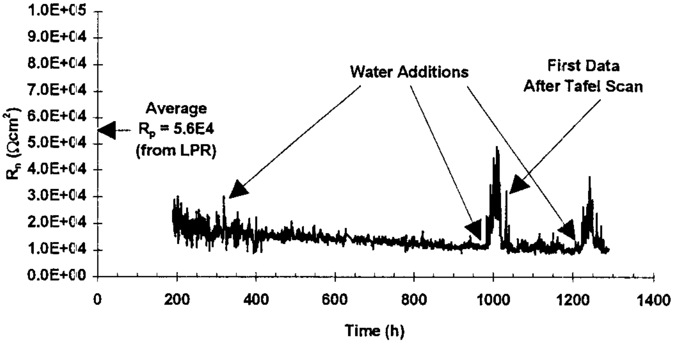

Figure 6: Comparison of R_n and R_p during first 1300 hours of operation. First ~200 hours unrecorded due to delay in data acquisition system installation following probe installation. Average R_p value is calculated from eight LPR tests conducted periodically during probe operation.

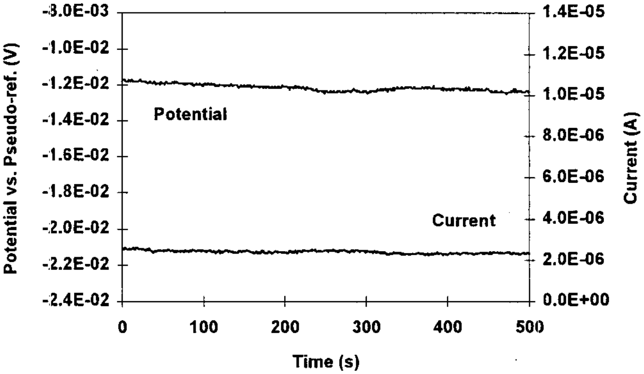

Figure 7: Current and potential data recorded ~200 hours after probe installation from electrode array in liquid waste. Data is characteristic of uniform corrosion.

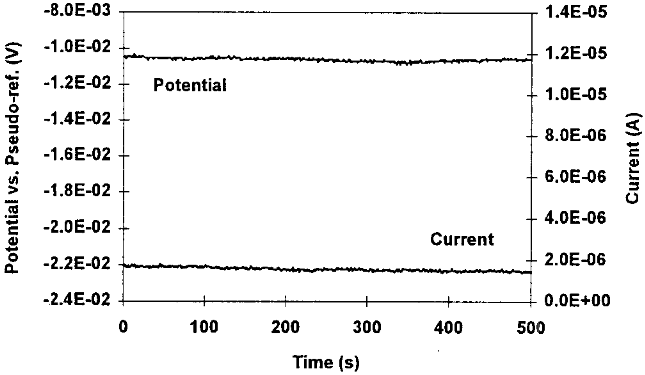

Figure 8: Current and potential data recorded ~350 hours after probe installation from electrode array in liquid waste. Data is characteristic of uniform corrosion.

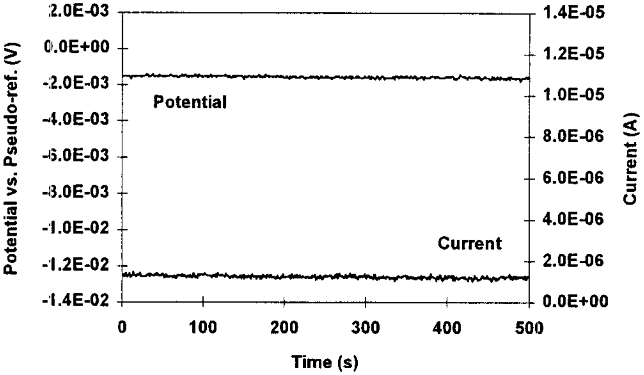

Figure 9: Current and potential data recorded ~950 hours after probe installation from electrode array in liquid waste. Data is characteristic of uniform corrosion.

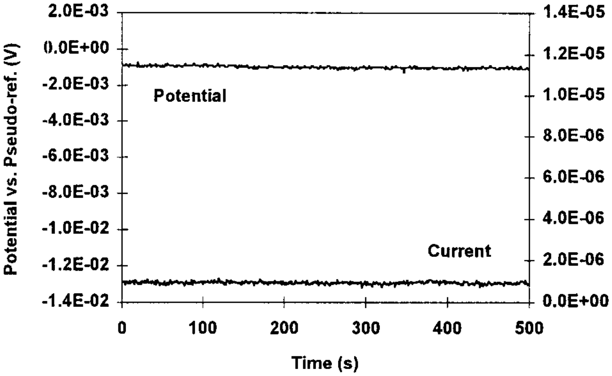

Figure 10: Current and potential data recorded ~1100 hours after probe installation from electrode array in liquid waste. Data is characteristic of uniform corrosion.