

CONF- 960389 - -3

Hanford Double Shell Tank Corrosion Monitoring Instrument Tree Prototype

RECEIVED
JAN 30 1996
OSTI

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



Westinghouse
Hanford Company Richland, Washington

Management and Operations Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

Copyright License By acceptance of this article, the publisher and/or recipient acknowledges the
U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering this paper.

Approved for public release; distribution is unlimited

LEGAL DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, nor any of their contractors, subcontractors or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or any third party's use or the results of such use of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof or its contractors or subcontractors. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

This report has been reproduced from the best available copy.

Printed in the United States of America

DISCLM-2.CHP (1-91)

Hanford Double Shell Tank Corrosion Monitoring Instrument Tree Prototype

**J. L. Nelson
G. L. Edgemon
P. C. Ohl**

**Date Published
November 1995**

**To Be Presented at
CORROSION/96
March 24-29, 1996
Denver, Colorado**

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



**Westinghouse
Hanford Company**

P.O Box 1970
Richland, Washington

Management and Operations Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

Copyright License By acceptance of this article, the publisher and/or recipient acknowledges the
U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering this paper.

Approved for public release; distribution is unlimited

HANFORD DOUBLE SHELL TANK
CORROSION MONITORING INSTRUMENT TREE PROTOTYPE

J. L. Nelson, G. L Edgemon and P. C. Ohl
Westinghouse Hanford Company
P. O. Box 1970
Richland, WA 99352

ABSTRACT⁽¹⁾

High-level nuclear wastes at the Hanford site are stored underground in carbon steel double-shell and single-shell tanks (DSTs and SSTs). The installation of a prototype corrosion monitoring instrument tree into DST 241-AZ-101 was completed in December, 1995. The instrument tree has the ability to detect and discriminate between uniform corrosion, pitting, and stress corrosion cracking (SCC) through the use of electrochemical noise measurements and a unique stressed element, three-electrode probe. The tree itself is constructed of AISI 304L stainless steel (UNS S30403), with probes in the vapor space, vapor/liquid interface and liquid. Successful development of these trees will allow their application to single shell tanks and the transfer of technology to other U.S. Department of Energy (DOE) sites.

Keywords: Hanford, radioactive waste, high-level waste tanks, electrochemical noise, probes, double-shell tanks, single-shell tanks, corrosion

INTRODUCTION

The DOE Hanford Site nuclear reservation, located in southeastern Washington State, maintains 61 million gallons (231 million liters) of radioactive waste stored in 177 large underground storage tanks. Both DSTs and SSTs are constructed of reinforced concrete with carbon steel liners. They primarily store caustic

¹ This report has been publicly released under the same title as Westinghouse Hanford Company Report # WHC-SA-3010-FP.

wastes from defense reprocessing of spent nuclear fuels. Some of the SSTs have leaked radioactive liquid waste to the soil. These leaks are possibly caused by nitrate-induced SCC. Efforts to avoid nitrate-induced SCC in newer DST designs appear successful. These efforts include a post weld stress relief at the time of construction and more rigorous control of hydroxide and nitrite waste chemistry. A potential for pitting and crevice corrosion in the carbon steel liners exists, particularly at the vapor/liquid interface and in the vapor zone. There is no evidence of significant uniform corrosion of the carbon steel liners.

In spite of careful control of waste chemistry, one DST has operated with low hydroxide levels for several years, and four additional DSTs have recently been corrected from below normal hydroxide levels. It appears that the highly complicated waste chemistries contained in the tanks may be consuming hydroxide at a slow rate. Such problems in controlling waste chemistry have accentuated the need for in-tank corrosion monitoring.

BACKGROUND

Hanford DST liners are fabricated from ASTM A515-Grade 60 (UNS K02401), A516-Grade 65 (UNS K02403) and A537-Class 1 (UNS K02400) carbon steels. The stored wastes are principally aqueous solutions of nitrates, nitrites, hydroxides, and aluminates. Lesser quantities of fluorides, chlorides, and organic chelating agents are also present. Radiation levels in the solutions can be as high as 1,000 Rad/hr, with some tanks containing sufficient quantities of heat producing radioisotopes that the wastes would boil spontaneously if not cooled. Chemistry controls for the DSTs specify that pH is to be maintained at greater than 10. Numerous laboratory studies of waste tank corrosion have demonstrated that carbon steel passivates under oxidizing conditions with pH between 8 and 14¹. Uniform corrosion under these conditions can be controlled to less than 0.5 mil per year².

The presence of significant quantities of nitrates in the waste raises the concern of nitrate induced SCC. It is this mechanism that is believed to be the cause of many of the SST failures at Hanford. Stress corrosion cracking in DSTs is controlled chemically by the use of hydroxide³ and nitrite as inhibitors, but maintaining adequate hydroxide concentrations in the waste is difficult. The slow consumption of hydroxide by the waste and the occasional addition of low hydroxide (uninhibited) water to the tanks tend to lower hydroxide levels to undesirable ranges. Moreover, the vapor above the waste is low in hydroxide and tends to condense on the tank walls. This condensate runs down into the waste, lowering the hydroxide concentration at the vapor/liquid interface near the tank wall. This relatively dilute, uninhibited waste in the vapor space and at the vapor/liquid interface is a serious concern for inducing SCC and pitting in these regions. It is these corrosion mechanisms being monitored by the instrument tree.

INSTRUMENT TREE

The Hanford corrosion monitoring instrument tree prototype has been constructed of 1.5 inch diameter (3.8 cm) AISI 304L stainless steel (UNS 30403) pipe for extended service in the waste tank environment. It is designed to fit through a nominal 4 inch (10.2 cm) tank riser and extends 33.2 feet (10.1 meters) into the tank. The tree is fitted with three probe arrays at the 13, 29 and 33 foot depths (4.0, 8.8 and 10.0 meters). The upper probe is positioned to detect vapor phase corrosion, while the lower two probes will be immersed in the liquid waste. Each probe array consists of three electrodes of ASTM A516-Grade 55 steel (UNS K01800), representing the tank wall material. One of the three electrodes in each array is stressed to 30,000 psi (207 MPa) as described below. An overall view of the instrument tree orientation in the tank is shown in Figure 1. A diagram of the complete tree assembly is shown as Figure 2.

The system has been designed to be capable of operation under the following service conditions:

- Temperature range from 10°C to 100°C
- Liquid phase pH range from 7 to 14
- Vapor phase relative humidity to 100%

- Liquid phase radiation levels to 1,000 Rad/hr
- Vapor phase radiation levels to 200 Rad/hr
- Tank liquid levels fluctuating from 6 in to 36 ft (15 cm to 11 m)
- Liquid phase flow rates of up to 3 ft/s (1 m/s)
- Vapor phase displacements of up to 495 standard cubic feet per minute (14 standard cubic meters per minute)

The following design goals were established for the instrument tree:

- Design service life shall be a minimum of two years. Data cables to be isolated from the service environment.
- Instrument assembly will fit through a maximum size circular tank dome opening of 4 in (10 cm).
- Data acquisition and instrument control will require a minimum of "on location" operation in order to reduce operator exposure to hazardous conditions. Remote data acquisition and control via modem, hard-wire, or radio frequency link is required.
- Minimum services provided at the tank opening are 110 volt a.c. (single phase, 20 amp, 60 hertz) power, phone (data) connection, and weather tight service box. Additional service requirements such as purge gas, environmental (heating/cooling) control, or water supply are not planned.
- Instrumentation will be designed to facilitate ease of decontamination by minimizing features where liquids could be retained.

A 10 conductor shielded data cable extends through the interior of the sealed pipe. At each probe array, three of the conductors are removed from the shielding and attached one each to the electrodes. A commercially available "glass-to-metal seal" is used to penetrate the pipe wall while providing electrical isolation for the signal from the electrode. The glass portion of the glass-to-metal seal provides electrical isolation between the data cable and the body of the instrument tree. Threads on the interior of the electrode are isolated from the tank waste through the use of an ethylene propylene (EPDM) gasket suitable for use in radiation environments. The configuration of the electrode, seal and wiring is shown in Figure 3. The cable terminates at the top of the assembly in a weather tight box. Approximately 40 feet (12.2 meters) of above ground cable runs from the top of the probe to a weather tight enclosure where the commercial electrochemical noise monitoring instrument is maintained within the radiation zone. This instrument is controlled by modem link to the corrosion engineers office.

INSTRUMENTATION

Previous efforts to monitor internal corrosion of waste tank systems have included linear polarization resistance and electrical resistance techniques⁴⁻⁵. These techniques are effective in monitoring uniform corrosion, but have not been effective for detection of localized corrosion in nuclear waste tank systems.

For many years, electrochemical noise has been observed during corrosion and other electrochemical reactions and the phenomenon is well-established⁶⁻¹⁷. It has also been reported that electrochemical noise evaluation is well suited for monitoring and identifying localized corrosion, and for the measurement of uniform corrosion rates¹²⁻¹⁸. Typically, electrochemical noise consists of low frequency (< 1 Hz) and amplitude (0.1 μ V-10 mV and 10 nA/cm² to 10 μ A/cm²) signals which are spontaneously generated by electrochemical reactions occurring at corroding surfaces¹⁸. The use of electrochemical noise for the monitoring and detection of localized corrosion processes, in particular, has received considerable attention and plant application in recent years¹⁹⁻²⁵.

The electrochemical noise evaluation approach involves the monitoring of instantaneous fluctuations in corrosion current and corrosion potential between nominally identical electrodes of the material of interest in the environment of interest. Time-dependent fluctuations in corrosion current are described by electrochemical current noise (ECN), while time-dependent fluctuations of corrosion potential are described by electrochemical potential noise (EPN). The most straight forward method involves simple statistical examination of the temporal

records with detailed investigation of identified regions of interest. Other methods of analysis involve transformation of temporal data to the frequency domain for analysis²⁶. Each type of localized corrosion phenomenon presents a unique relationship between ECN and EPN transients, thereby allowing distinction between different types of localized corrosion (e.g. pitting, stress corrosion cracking, intergranular attack, etc.). Representations of EPN and ECN for uniform corrosion, pitting, and stress corrosion cracking are shown in Figures 4 and 5 respectively. These data were produced during laboratory studies of tank steels in simple waste simulants.

In the prototype tree assembly, potential and current are measured between three electrodes of the same materials the tank liner. One electrode serves as a reference electrode. To detect stress corrosion cracking, one electrode will be stressed²⁷.

Electrodes

Electrodes for the instrument tree have been specially manufactured of representative tank steel alloy by Cortest Instruments⁽²⁾. Outwardly, all electrodes are 0.25 inch diameter (0.64 cm) by 1 inch long (2.54 cm) cylinders, tapped and threaded at one end to fit the industry standard glass-to-metal probe connector. A stressed electrode has been introduced by machining one electrode per set of three to accept an oversized internal pin. An interference fit is achieved by cooling the pin to -70° F (-57° C) and pressing it into the electrode body. The circumferential stress at the electrode surface for a 0.001 inch (0.0025 cm) interference fit is given by equation 1:

$$\sigma \approx E[(\pi(D_2 + 0.001) - \pi D_2)/(\pi D_2)] \quad (1)$$

D_2 = electrode outside diameter

σ = stress

E = Young's modulus of elasticity

A stress of 30,000 psi (207 MPa) has been applied to the electrode. The maximum stress in a tank wall is estimated to be 40,000 psi (276 MPa).

Electrochemical Noise Equipment

The initial electrochemical instrumentation consists of a multi-technique integrated electrochemical corrosion monitoring system manufactured by Gamry Instruments⁽³⁾ with the associated software used for data collection and processing. It is expected that the capabilities of this instrument will be augmented through the use of a Capcis March Ltd⁽⁴⁾ instrument with similar capabilities.

SUMMARY

A prototype corrosion monitoring probe system has been installed in a Hanford DST. The probe system utilizes electrochemical noise for evaluation of tank corrosion conditions. Through the use of a unique stressed-element, three-electrode probe, it is believed that local corrosion phenomena such as pitting and stress corrosion cracking can be observed as they occur.

² Cortest Instrument Systems, Willoughby, Ohio.

³ Gamry Instruments, Inc., Willow Grove, Pennsylvania.

⁴ Capcis March Ltd., Inc., Kingwood, Texas.

REFERENCES

1. Metals Handbook, Ninth Edition, 1987, Vol. 13, Corrosion, American Society for Metals, Metals Park, Ohio.
2. Divine, J. R., W. M. Bowen, D. R. Mackey, D. J. Bates, and K. H. Pool, 1985, "Prediction Equations for Corrosion Rates of A-537 and A-516 Steels in Double Shell Slurry, Future Purex, and Hanford Facilities Wastes," PNL-5488, Pacific Northwest Laboratory, Richland, Washington.
3. Ondrejcin, R. S., 1984, "Prevention of Stress Corrosion Cracking in Nuclear Waste Storage Tanks," Corrosion/84, paper no. 258, (Houston, TX: NACE International 1984).
4. J. L. Nelson et al., "Hanford DST Corrosion Monitoring Instrument Tree," CORROSION/95, paper no. 440, (Houston, TX: NACE International, 1995).
5. R.K. Shukla, et al., "Corrosion Monitoring of High Level Waste Storage Tank 8-D2 at the West Valley Demonstration Project," CORROSION/94, paper no. 121, (Houston, TX: NACE International, 1994).
6. T. Haygard and J. R. Williams, Trans. Farad. Soc. 57, (1961): p. 2288-2298.
7. P. Bindra, et al., Discussions of Faraday Soc. 56, (1974): p. 189-209.
8. M. Fleischmann, et al., Surface Science 100-101, (1980): p. 583-601.
9. G. J. Bignold and M. Fleischmann, Electrochemical Acta 19, (1974): p. 363-374.
10. E. Budevski, et al., Electrochemical Acta 28, (1983): p. 925-931.
11. G. Blanc, et al., Electrochemical Acta 23, (1978): p. 337-340.
12. K. Hladky and J. L. Dawson, Corrosion Science 22, (1982): p. 231-237.
13. D. A. Eden, et al., International Patent Classification G01N17/00, 19 Nov 87.
14. K. Hladky, U.S. Patent No. 4,575,678, March 11, 1986.
15. U. Bertocci, "Electrochemical Noise Analysis and Its Application to Corrosion," CORROSION/89, paper no. 24, (Houston, TX: NACE International, 1989).
16. J. L. Dawson, et al., "Corrosion Monitoring Using Electrochemical Noise Measurements," CORROSION/89, paper no. 31, (Houston, TX: NACE International, 1989).
17. D. A. Eden, A. N. Rothwell, and J. L. Dawson, "Electrochemical Noise for Detection of Susceptibility to Stress Corrosion Cracking," CORROSION/91, paper no. 444, (Houston, TX: NACE International, 1991).
18. D. M. Farrell, Industrial Corrosion 9, (1991): p. 7-13.
19. A. N. Rothwell, T. G. Walsh and W. M. Cox, "On Line Corrosion Investigation and Surveillance - Chemical Plant Case Studies," CORROSION/91, paper no. 170, (Houston, TX: NACE International, 1991).

20. J. L. Dawson, et al., On-line Monitoring of Continuous Process Plants, D. Butcher, editor (Ellis Horwood, 1983).
21. D. M. Farrell, W. M. Cox, and D. Gearey, Multi-System Corrosion Monitoring in a Cyclic Reheat Test Facility; Phase 1, EPRI CS-5776 (Pleasant Hill, CA: Electric Power Research Institute, 1988).
22. D. M. Farrell, W. M. Cox, and D. Gearey, Multi-System Corrosion Monitoring in FGD Systems; Phase 2, EPRI CS-5734 (Pleasant Hill, CA: Electric Power Research Institute, 1988).
23. W. M. Cox and B. C. Syrett, "The Future of On-line Materials Surveillance for Operating Process Plants," Life Prediction of Corrodible Structures - Cambridge, UK, Sept., 1991, (Houston, TX: NACE International, 1991).
24. W. M. Cox and H. D. Spriggs, "On-line Surveillance Instrumentation for Failure Investigation, Materials Evaluation and Process Control in Combustion Equipment," Fifth Annual International Pittsburgh Coal Conference - Pittsburgh, PA, Sept., 1988, (Pittsburgh, PA, 1988).
25. P.E. Doherty, et al., "An Application of Advanced Electrochemical Monitoring of Corrosion of Heat Exchanger Tubing," Fourth International Symposium on Environmental Degradation of Materials in Nuclear Power Systems - Jekyll Island, GA, Aug., 1989, (Houston, TX: NACE International, 1989).
26. D.A. Eden and J.L. Dawson, "Transformation of Electrochemical Noise Data into Impedance Spectra," First International Symposium on Electrochemical Noise for Corrosion Applications - Montreal, Canada, May 16-17, 1994, (Philadelphia, PA: ASTM, 1994).
27. Report to the ASTM Subcommittee G01.11.04 and New Standard Guide for Electrochemical Noise Measurement.

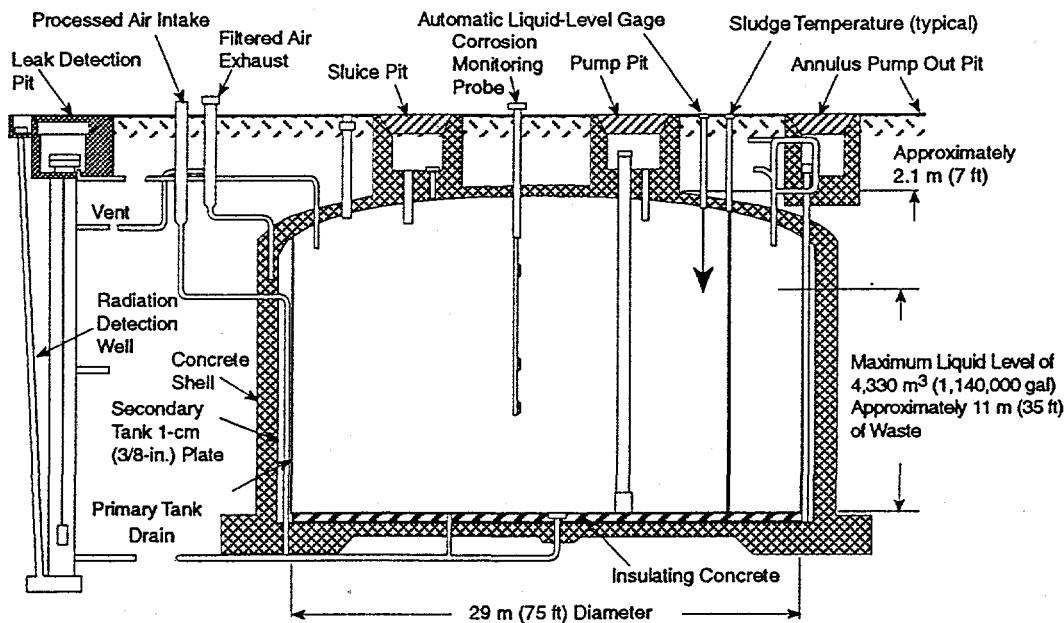


Figure 1 - Typical Double Shell Tank showing orientation of prototype corrosion monitoring instrument tree.

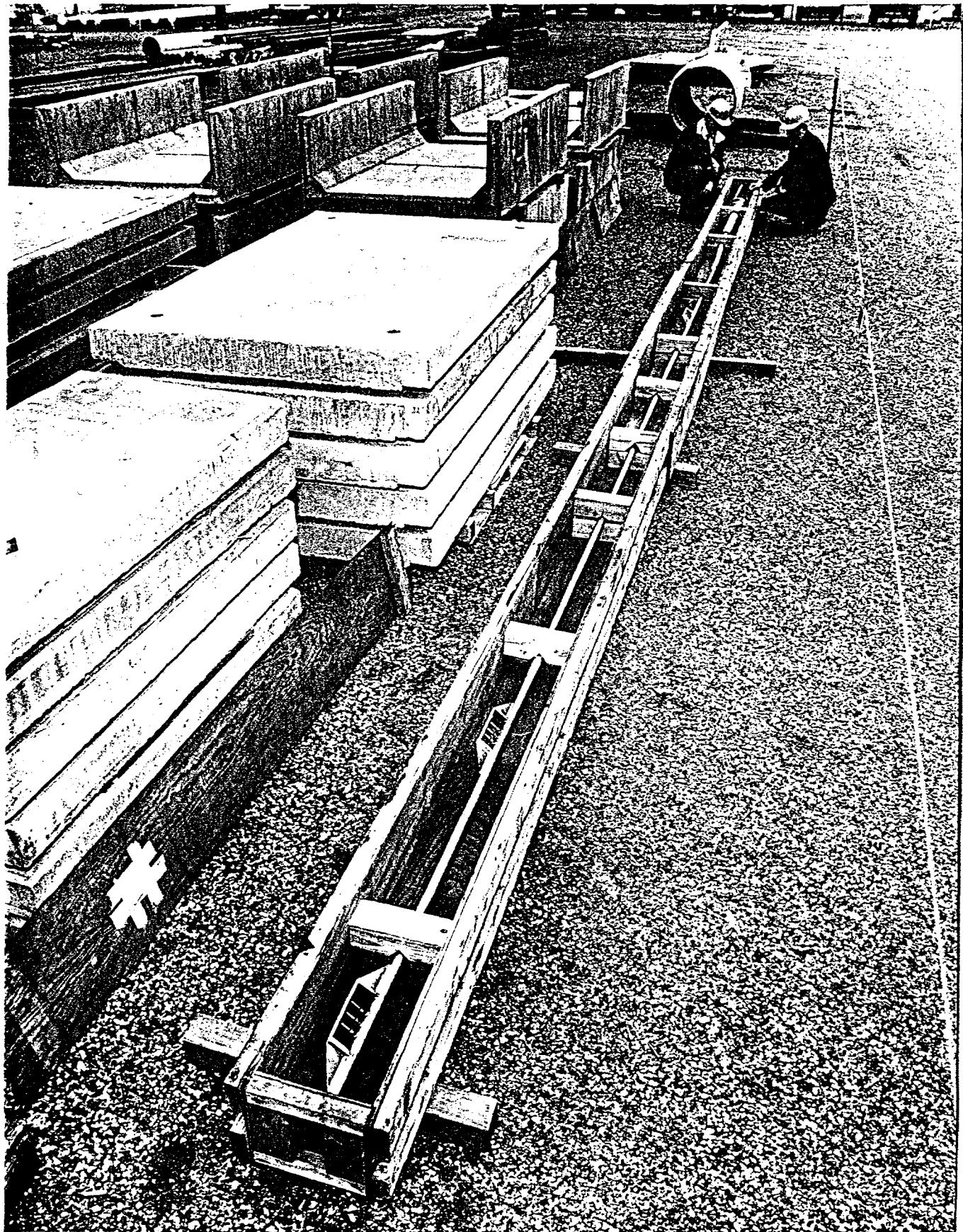


Figure 2 - Overall view of instrument tree assembly.

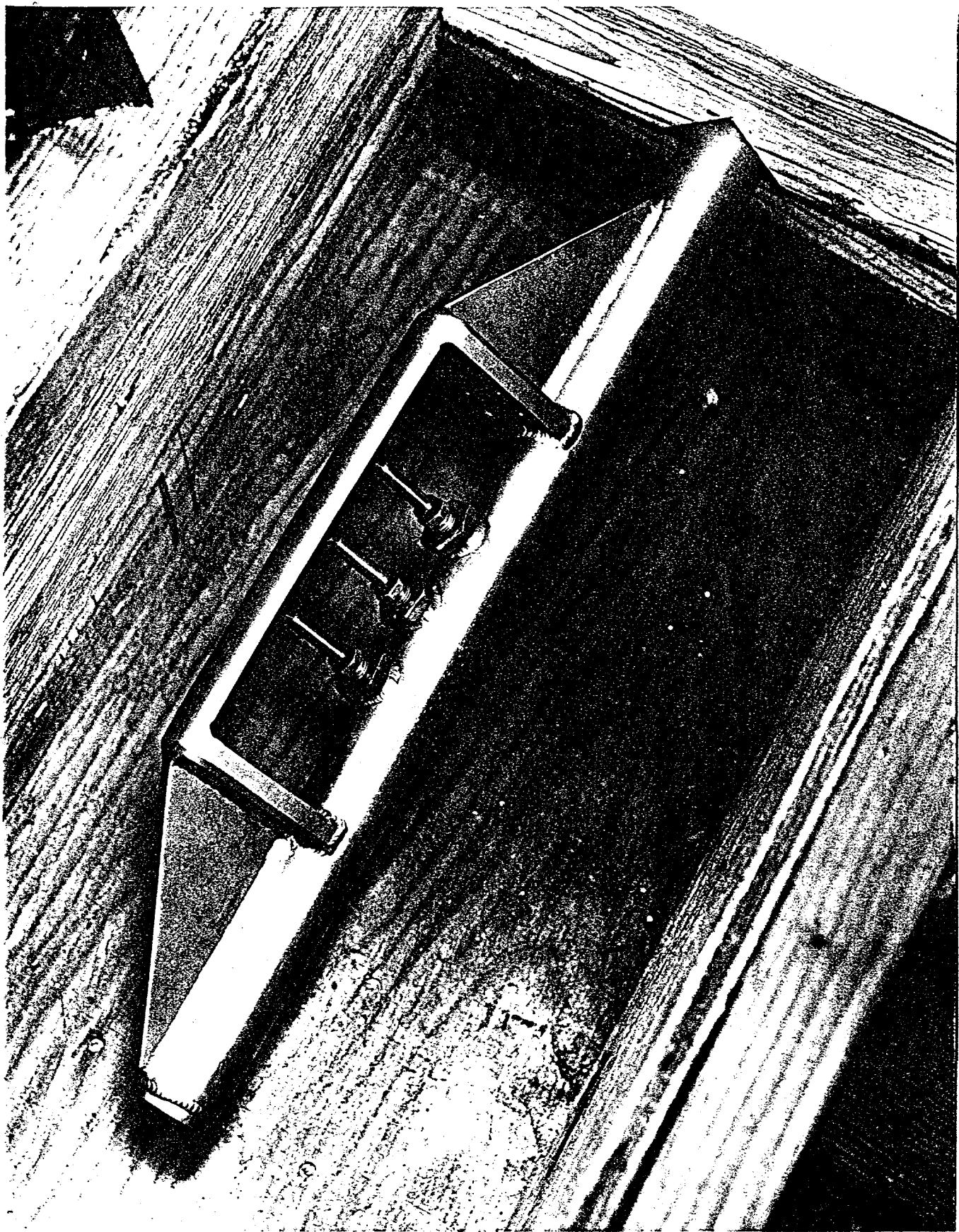
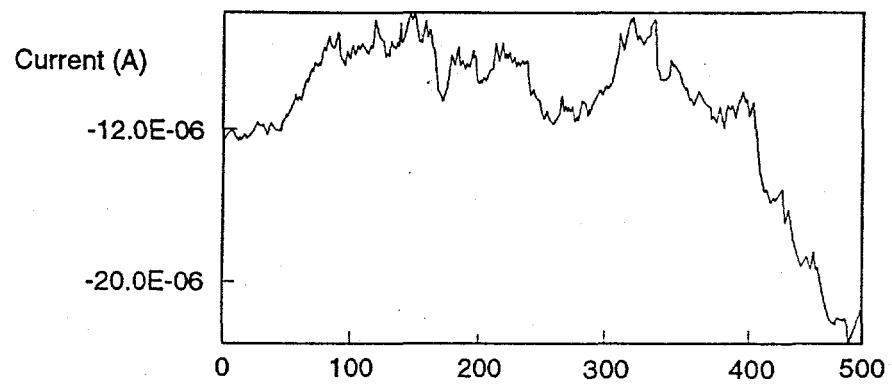
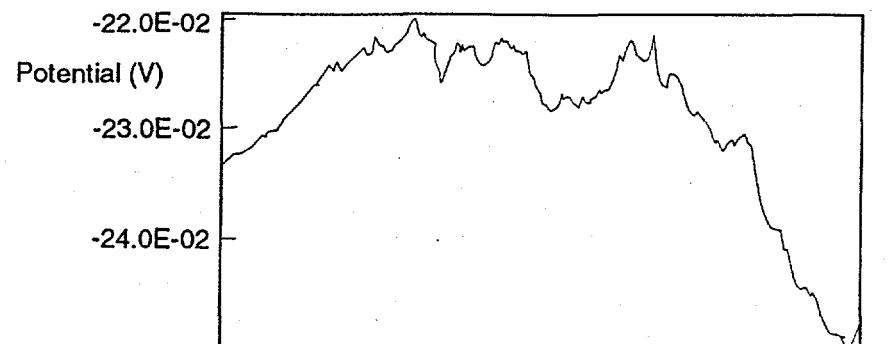
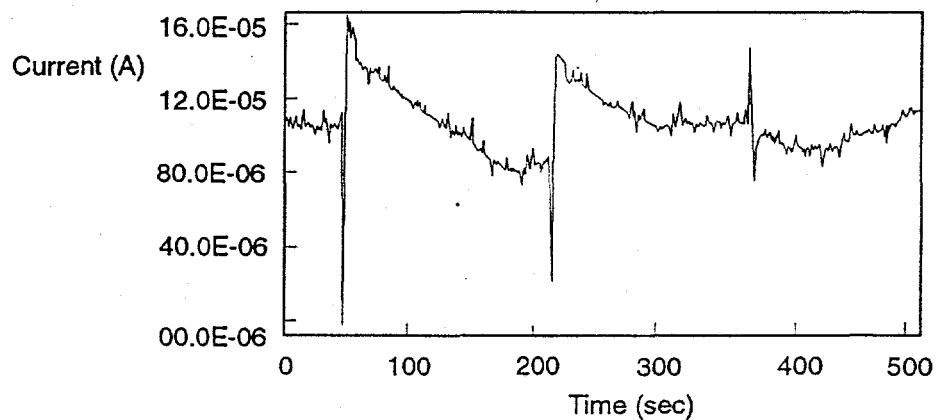
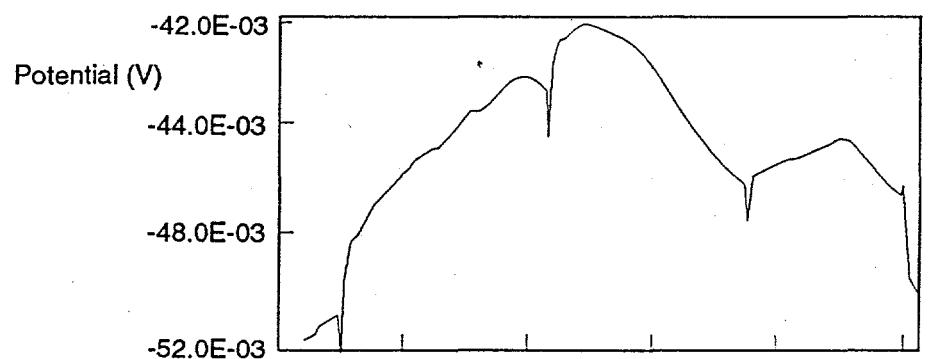


Figure 3 - Electrode array showing position of electrodes, glass-to-metal seals, gaskets, and electrode shield.



2G95110397.2

Figure 4 - Characteristic EPN and ECN transients for uniform corrosion.



2G95110397.3

Figure 5 - Characteristic EPN and ECN transients for pitting and stress corrosion cracking.