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Analyses Methodologies for In-Situ Corrosion Monitoring of Tank Bottom Plate Corrosion Using Electrical Resistance Probes

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ABSTRACT

Electrical Resistance (ER) probes are routinely used to monitor above ground storage tank bottom plate corrosion. However, surface and material conditions between the tank plate and ER probes can differ significantly. In addition, a tank plate may have undergone weathering in various conditions that may not be applicable for the ER probe elements. In such situations, ER probes' accumulative corrosion could be different compared to that of the tank bottom plate. The corrosion could further change when vapor corrosion inhibitor treatments are applied to the tank pad. Experiments were conducted using coupons and ER probes. Coupons were weathered by initially exposing them to the corrosive conditions, and then three different VCI treatments were applied. The ER probe data were continuously collected. The coupons' and ER-probe corrosion rates were compared before and after VCI treatments using a ratio method. This paper presents various comparison methodologies that can be used to bound the bottom plate corrosion conditions and corresponding corrosion rates. The paper also presents recommendations for using various element-type ER probes for the tank bottom plates' in-situ corrosion monitoring.

Key words: Vapor Corrosion Inhibitors, Hanford, Double Shell Tanks, Bottom Plate, Pitting Corrosion.

INTRODUCTION

Electrical resistance (ER) based corrosion monitoring probes are sometimes used to monitor soil-side corrosion of Aboveground Storage Tanks (ASTs). Lyublinski et al.¹ and Whited et al.² used the ER probe to report effectiveness of Vapor Corrosion Inhibitors (VCIs), however, a correlation between the actual corrosion rate at the tank bottom and ER probe data has not been established in the literature. There is an implicit assumption that ER probe data is representative of corrosion of tank bottoms with and without VCI application. In other words, ER-measured corrosion rates are equivalent to the corrosion rates of the tank bottom plate with or without VCI dosing. To test the assumption of the corrosion rate of tank bottom plate being close to that of the ER probes, laboratory scale experiments were conducted on a nuclear system described next.

High-level radioactive waste generated during reprocessing of spent nuclear fuel at Hanford has been stored in several single- and 27 double shell tanks (DSTs). Each DST consists of a primary shell (inner) surrounded by secondary (outer) liner. The secondary liner rests on a concrete foundation. Rainwater seeps in and accumulates in the drain slots and corrode the exterior of the secondary liner. Evidence of corrosion has been detected via ultrasonic inspections of the secondary shell floor. The inspection is confined to the annular space between the primary and secondary tank shells; there is a concern that corrosion is widespread on the underside of the bottom plate. Since the rainwater level can vary in the drain slots based on accumulation, corrosion could be caused by direct contact with the accumulated water; when the leak detection pit (LDP) water level is below the structural limit, vapor space corrosion (VSC) could also occur. Accumulated water is drained through the sumps in the LDP. The LDP water was analyzed for its constituents, and two simulants were developed considering the chemical composition range of the accumulated water. The simulants are identified as leak detection pit and ground water (GW); compositions are listed in Table 1. A previous study established that GW simulant is more corrosive than the leak detection pit, therefore GW was used in the VCI effectiveness study.³

Laboratory experiments were conducted to address the concerns of both immersed-phase corrosion and VSC of the tank steel exposed to GW simulant. VCIs were also tested to determine their efficacy in mitigating the corrosion on pre-corroded surfaces. VCIs' application for above ground storage tank bottom corrosion control is recent, and several studies have documented effectiveness of VCIs for tank bottom plate corrosion control.^{1,2,4,5,6} Electrical resistance (ER) probes were used to in-situ monitor efficacy of the VCIs. The coupon and ER probes were analyzed, and corrosion rates using the two were estimated. The corrosion rate data from the two were estimated.

Table 1
Composition of the Leak Detection Pit and Ground Water Simulants

Source chemical	Concentration (M)	
	Leak Detection Pit	Ground Water
Sodium bicarbonate	1.12×10^{-3}	1.75×10^{-3}
Calcium hydroxide	1.21×10^{-4}	1.50×10^{-3}
Potassium nitrate	6.75×10^{-5}	2.40×10^{-4}
Magnesium Nitrate, 6hydrate	1.52×10^{-5}	–
Strontium Nitrate	4.04×10^{-6}	2.87×10^{-6}
Sodium sulfate	1.83×10^{-6}	–
Ferric sulfate	–	6.25×10^{-4}
Sodium Metasilicate, 5hydrate	4.57×10^{-5}	6.00×10^{-4}
Ferric chloride	2.67×10^{-6}	7.67×10^{-5}
Manganese Chloride	–	3.100E-04
Acetic Acid	3.00×10^{-4}	3.000E-04
Adjusted pH	7.6	7.6

EXPERIMENTAL

Three experiments were set up using GW simulants as electrolytes. VCIs were added to each experiment mid-course. Two VCIs were used and are identified as VCI-A⁽¹⁾ and VCI-B⁽²⁾. The experiments included (i) initially GW simulant, and then 100% of the recommended dosage of VCI-A after 2 months, (ii) initially GW simulant, and then

⁽¹⁾ VCI-A was VpCI-337 manufactured by Cortec Corporation.

⁽²⁾ VCI-B was a mixture of VpCI-649 MF and VpCI-609, both manufactured by Cortec Corporation.

100% of the recommended dosage of VCI-B after 2 months, and (ii) initially GW simulant, and then 50% of the recommended dosage of VCI-B after 2 months.

Disk coupons, machined from a legacy carbon steel plate, were used in the experiments. The legacy carbon steel is based on specifications of Association of American Railroads⁽³⁾ Tank Car (AAR TC 128) steel, and its chemistry and microstructure are similar to the vintage steel from which the tanks were fabricated UNS K02401 (i.e., American Society for Testing and Materials (ASTM)⁽⁴⁾ A515 Grade 60 carbon steel). The chemical composition of the legacy carbon steel is listed in Table 2. All elemental compositions except for Mn and Si meet the ASTM A515 Grade 60 specification. The coupons were 25 mm (1 inch) diameter with a thickness of 3 mm (0.125 inch) and polished to a 600-grit finish. The coupons were potted in a mold prepared with a two-part transparent epoxy mold such that that one face of the coupon was exposed to the test electrolyte. The coupons' exposed surfaces were modified to simulate crevice corrosion. A crevice former was attached to each coupon surface using tape and wire. The crevice formers partially covered coupons' surfaces, which created conditions for localized corrosion under the crevice formers. Additional experimental details are in Shukla et al.⁷

Table 2
Chemical Composition of AAR TC 128 Steel (wt.%)

	C	Mn	P	S	Si	Fe
Specification	0.24 (max.)	0.9 (max.)	0.035 (max.)	0.04 (max.)	0.13 to 0.33	Balance
Measured	0.212	1.029	0.012	0.013	0.061	Balance

A glass vessel of dimensions 1.0 m (3.3 ft) tall and 14 cm (5.5 inch) diameter was used for each experiment. Approximately 1.25 L of GW simulant was added to a vessel for each experiment. Each vessel has a water jacket around the simulant holding area which was used to circulate warm water to maintain the simulant temperature at 45 ± 2 °C. Each vessel also has several ports, which were used to insert thermocouples and electrical resistance (ER) probes. An image showing the three vessels used is presented in Figure. Coupons were exposed to the electrolyte and vapors of the electrolyte in each experiment by suspending them using rods. The coupons were suspended such that the exposed surfaces of the vapor space coupons were facing the electrolyte. The rods holding the coupons were placed inside the vessels. Several coupons were immersed in each vessel, and coupons were also placed in vapor space of each vessel. The vapor space coupons were placed at several height levels with respect to the electrolyte using the rods. The coupons' positions, with respect to electrolyte in each vessel, simulated different vapor space conditions and water levels in the drain slots. These levels, representing the drain slot characteristics and its position with respect to the bottom, are described:

Level 1: Bottom or low level. Coupons were dipped in the simulant for five minutes prior to testing. The coupons were hung at the bottom fixed ring of the rod. These coupons were suspended approximately 25 mm (1 inch) above the liquid level of the simulant. Every two weeks, the coupons were lowered into the simulant for 5 minutes. This level is representative of the situation when the secondary liner bottom plate experienced periodic wetting/drying.

Level 2: Intermediate or middle level. Coupons were dipped in the simulant for five minutes prior to testing. The coupons were hung at the middle-fixed ring approximately 46 cm (18 inch) above the liquid simulant in each vessel. This level is representative of a vapor space region of the secondary liner bottom that at one time was exposed to water, but has infrequent or no contact with the water. However, this region is exposed to the humidified air.

Level 3: Top or high level. This set of coupons was not exposed to the solution prior to testing. The coupons were suspended approximately 92 cm (36 inch) above the simulant. This level is representative of the secondary liner bottom plate region that is only exposed to the humidified air and any volatile species from the solution.

Description of the vessels for each electrolyte is provided in Table 3. Electrical Resistance (ER) probes were placed in Vessels 1, 2, and 3; placement positions are detailed in Table 3. ER probe data were collected periodically. Coupons were removed after several months of exposure, cleaned with Clarke's solution⁸ to remove corrosion products, and weighed to determine the mass loss of each coupon.

⁽³⁾ American Association of Railroads, 425 3rd Street SW, Washington, DC 20024

⁽⁴⁾ ASTM International, 100 Barr Harbor Dr., West Conshohocken, PA 19428-2959

Pitting and patch-like corrosion occurred on all the coupons. Coupon surfaces were profiled and the deepest pit in each coupon was measured from the surface profile data. In addition, each coupon's mass change was also recorded.

Table 3
Electrolyte Description, Vessel Identification, and Coupons and ER Probe Information

Electrolyte	Corrosion Cell	Notes
Initially GW simulant, and then 100% of the recommended dosage of VCI-A after 2 months	Vessel 1	<ul style="list-style-type: none"> 6 coupons each in immersed, Level 1, Level 2, and Level 3 positions, total 24 coupons. ER probes at each level. Cylindrical element probes at immersed, Levels 1 and 3, and wire element probe at Level 3.
Initially GW simulant, and then 100% of the recommended dosage of VCI-B after 2 months	Vessel 2	<ul style="list-style-type: none"> 6 coupons each in immersed, Level 1, Level 2, and Level 3 positions, total 24 coupons. ER probes at each level. Cylindrical element probes at immersed, Levels 1 and 3, and wire element probe at Level 3
Initially GW simulant, and then 100% of the recommended dosage of VCI-B after 2 months	Vessel 3	<ul style="list-style-type: none"> 6 coupons each in immersed, Level 1, Level 2, and Level 3 positions, total 24 coupons. Cylindrical element probe at Level 2 and wire element probe at Level 3



Figure 1: Image of the experimental setup

EXPERIMENTAL DATA AND RESULTS

The corrosion rate data for the coupons in Vessels 1, 2, and 3 are listed in Table 4. The data is for the coupons exposed to GW for the first two months and then GW + 100% VCI-A for additional four months. The tables have averages and standard deviations of surface average corrosion rate data. The coupons exposed for two and six months are referred as 2-month and 6-month coupons, respectively, hereafter. The 2-month coupons were exposed to GW simulant whereas 6-month coupons were exposed to GW for the first two months and then GW plus VCI dosage for an additional four months. The detailed corrosion rate data for the coupons can be found in a companion paper by Shukla et al.⁷

Table 4
Coupon and Electrical Resistance Probe Corrosion Rates

Vessel	Level	Coupon Corrosion Rates ($\mu\text{m}/\text{yr}$)***		ER Probe Corrosion Rates ($\mu\text{m}/\text{yr}$)			
				3-period rolling average		5-period rolling average	
		2-month*	6-month**	2-month*	6-month*	2-month*	6-month*
1	Immersed	149 \pm 14	61 \pm 0	224	0	224	0
	Level 1	86 \pm 51	45 \pm 12	–	0	–	0
	Level 2	71 \pm 22	36 \pm 6	66	0	66	0.3
	Level 3	60 \pm 24	36 \pm 9	197	81	197	67
2	Immersed	125 \pm 9	82 \pm 5	207	22	207	7.5
	Level 1	94 \pm 13	42 \pm 8	15	0	15	0
	Level 2	97 \pm 18	49 \pm 8	60	5	60	0
	Level 3	59 \pm 24	36 \pm 2	147	61	147	53
3	Immersed	116 \pm 15	51 \pm 5	–	–	–	–
	Level 1	58 \pm 15	22 \pm 4	–	–	–	–
	Level 2	60 \pm 8	47 \pm 26	37	0	37	0
	Level 3	36 \pm 12	47 \pm 36	7	0	7	0
*2-month coupons were exposed to GW only **6-month coupons were exposed to GW for the first two months and then to GW plus VCI for additional four months ***Corrosion rate data is estimated using three coupons per exposure level, 25 $\mu\text{m}/\text{yr}$ = 1 mil/yr = 1 mpy *Corrosion rates are for the duration of the VCI treatment							

The ER probes metal loss data for Vessel 1 is presented in Figure 2(a); ER probes were at immersed, and Levels 1-3. The first 18-days data were collected using Model-A datalogger, at which time the Model A datalogger malfunctioned. Therefore, the Model B datalogger was used for about 40 days. The Model A datalogger was repaired and data collection began on day 77 of the experiments. The data collected using the two dataloggers are marked in Figure 2(a). The Model B data were found to be erratic and inconsistent, and therefore, were not used to estimate corrosion rates. The Model A data were used to estimate 2-month corrosion rates. The Model A data were also used to estimate the probes' corrosion rates during VCI treatment. The ER probe data during the VCI treatment were continuously collected. The probes' data were found to fluctuate between measurements. Considering this, daily, 3-period, and 5-period rolling averages of the ER probe data were calculated. For Vessel 1, the ER probe data collected using Model A datalogger during the VCI treatment is presented in Figure 3(a), Figure 3(b), Figure 3(c), and Figure 3(d) for immersed, Level 1, Level 2, and Level 3 probes, respectively; the data in each of the four figures also include corrosion rates estimated using the 5-period rolling average of the ER probes'

data. The 2-month and 6-month (for VCI treatment only) ER-probe-derived corrosion rates are listed in Table 4. A parallel treatment was applied to Vessels 2 and 3 ER probe data, and the coupons' and ER probes' corrosion rates are listed in Table 5. Vessels 2 and 4 ER probe raw data are presented in Figures 2(b) and 2(c), respectively. Vessel 2 ER probes' processed data and corresponding 5-period-rolling-average-derived corrosion rates are presented in Figure 4(a), Figure 4(b), Figure 4(c), and Figure 4(d) for immersed, Level 1, Level 2, and Level 3 probes, respectively. Similarly, Vessel 3 ER probes' processed data and corresponding 5-period-rolling-average-derived corrosion rates are presented in Figure 5(a) and Figure 5(b) for Level 2 and Level 3 probes, respectively.

The coupon and ER probe data were analyzed for cross consistency and to evaluate the effectiveness of VCIs in mitigating corrosion. All coupons were exposed to GW for the first two months, and only half of the coupons were extracted when VCIs were added in mid-course. The remaining coupons were exposed to GW + VCI for an additional four months. The corrosion rates of the GW + VCI exposed coupons, i.e., 6-month coupons, are expected to be at least one third of the GW only exposed coupons, i.e., 2-month coupons. Ratios of the 2-month to 6-month coupons' corrosion rates were calculated; for each VCI treatment and ER probe location, two ratios were calculated using the following equation:

$$\text{Ratio 1} = \frac{(\text{Corrosion Rate} - \text{Std})_{2\text{-month}}}{(\text{Corrosion Rate} + \text{Std})_{6\text{-month}}} \quad \text{Ratio 2} = \frac{(\text{Corrosion Rate} + \text{Std})_{2\text{-month}}}{(\text{Corrosion Rate} - \text{Std})_{6\text{-month}}} \quad (1)$$

where *Ratio 1* and *Ratio 2* represent lower and upper bounds of the ratio range. The calculated ratio range, 6-month ER probe corrosion rates, and corrosion notes are listed in Table 5. If the ratio range includes a number 3 or the upper bound is close to number 3, or the ER probe corrosion rates being zero or close to zero is consistent with the corrosion rate data.

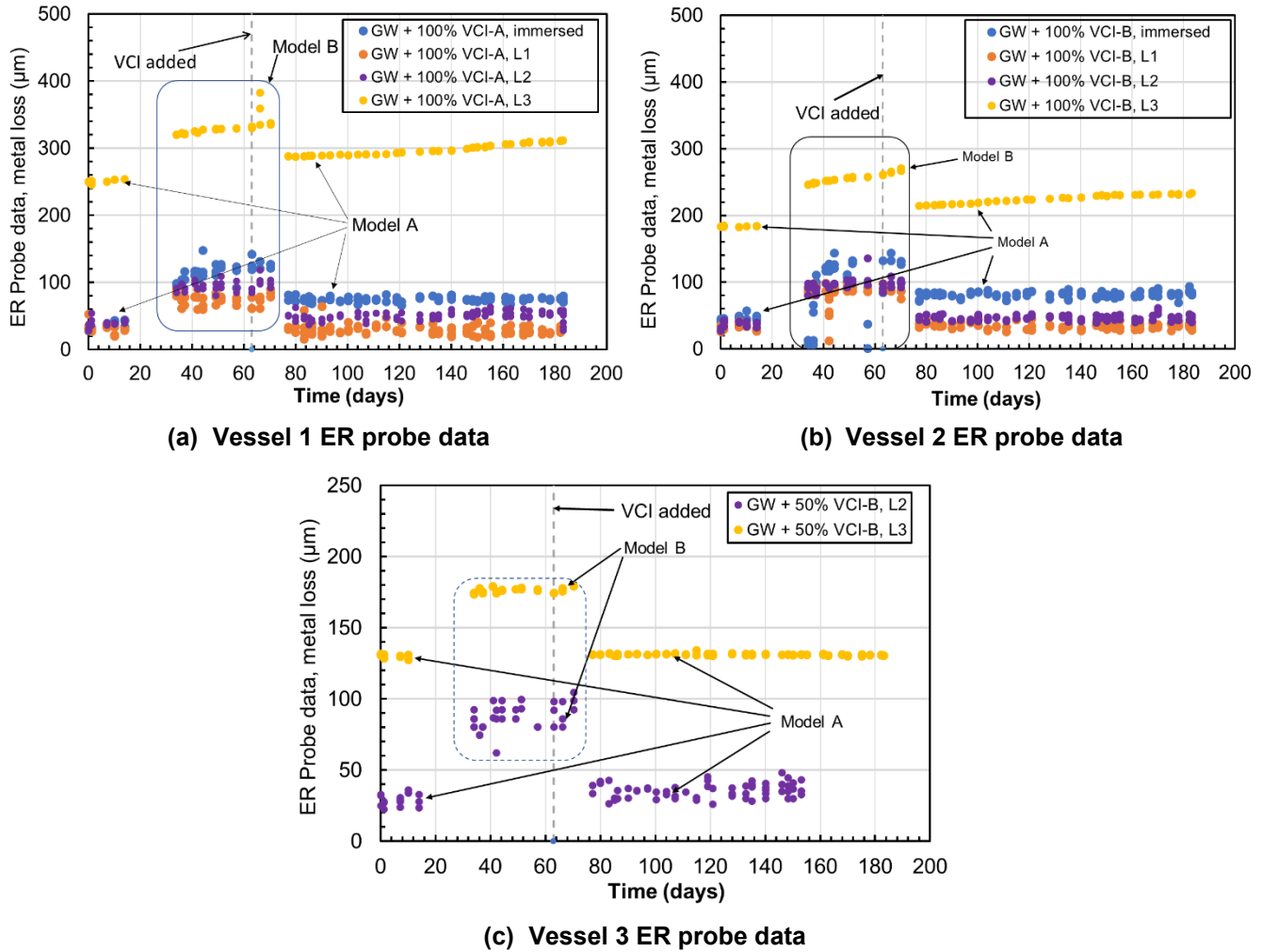
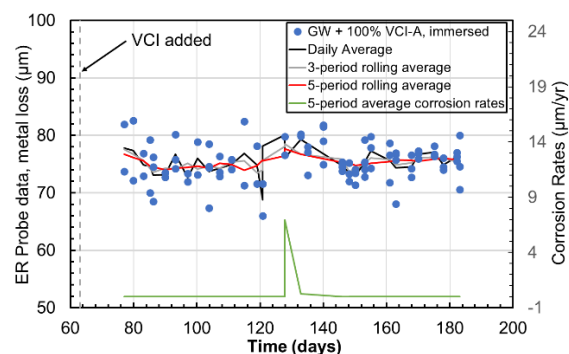
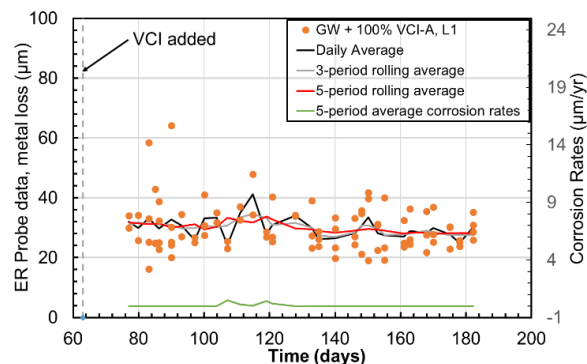


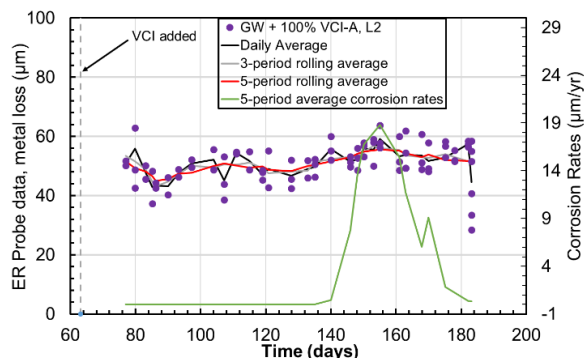
Figure 2: ER probes' metal loss data for the Vessels 1, 2, and 3



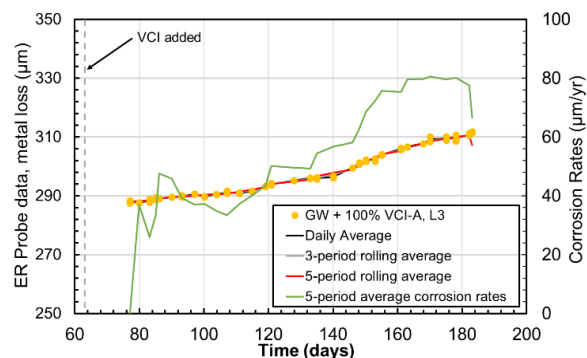
(a) Immersed level ER probe data and corrosion rates



(b) Level 1 ER probe data and corrosion rates



(c) Level 2 ER probe data and corrosion rates



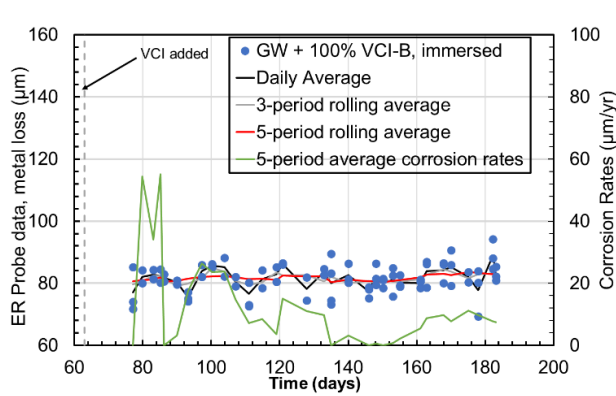
(d) Level 3 ER probe data and corrosion rates

Figure 3: Vessel 1 ER probe data and corresponding corrosion rates

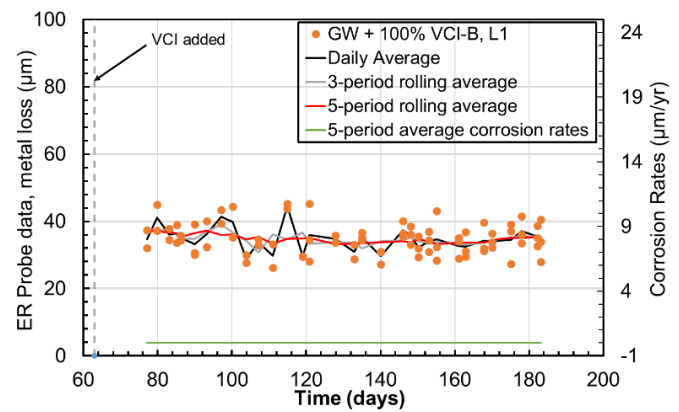
Table 5
Ratio Analysis of Coupon Corrosion Rates and Comparison with Electrical Resistance
Probe Corrosion Rates

Vessel	Level	Ratio of 2-month to 6-month coupon corrosion rates	6-month ER probe corrosion rates during VCI treatment* (μm/yr)	Notes
Vessel 1 (GW for first two months, and GW+100% VCI-A for additional four months)	Immersed	2.2 to 2.7	0	Ratio range upper limit is close to 3
	Level 1	0.6 to 4.2	0	Ratio range includes 3
	Level 2	1.1 to 3	0.3	Ibid
	Level 3	0.8 to 3	67	Ratio range include 3, but ER probe corrosion rates were high
Vessel 2 (GW for first two months, and GW+100% VCI-B for additional four months)	Immersed	1.3 to 1.8	7.5	Ratio range does not include 3
	Level 1	1.6 to 3.1	0	Ratio range includes 3
	Level 2	1.4 to 2.8	0	Ratio range upper limit is close to 3
	Level 3	0.9 to 2.5	53	Ratio range upper limit is close to 3
Vessel 3 (GW for first two months, and GW+50% VCI-B for additional four months)	Level 2	0.7 to 3.2	0	Ratio range includes 3
	Level 3	0.3 to 4.4	0	Ibid

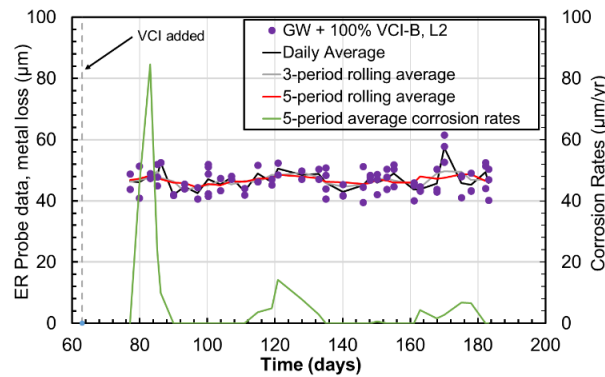
*VCI treatment only corrosion rates based on 5-period rolling average



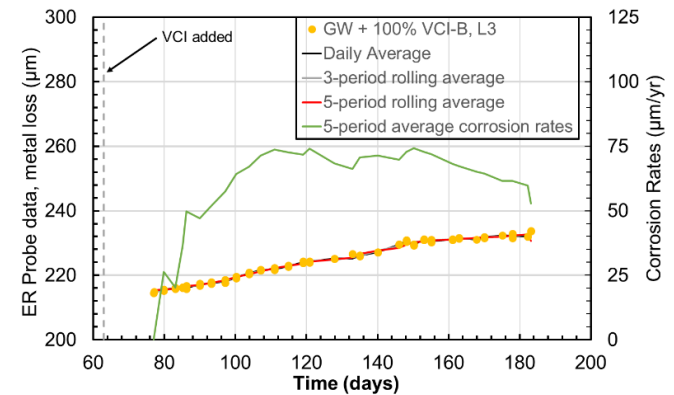
(a) Immersed level ER probe data and corrosion rates



(b) Level 1 ER probe data and corrosion rates

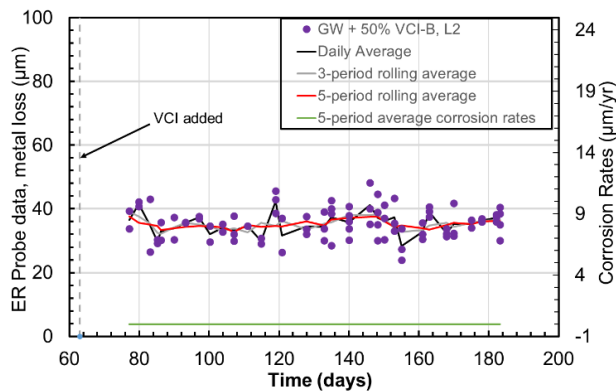


(c) Level 2 ER probe data and corrosion rates

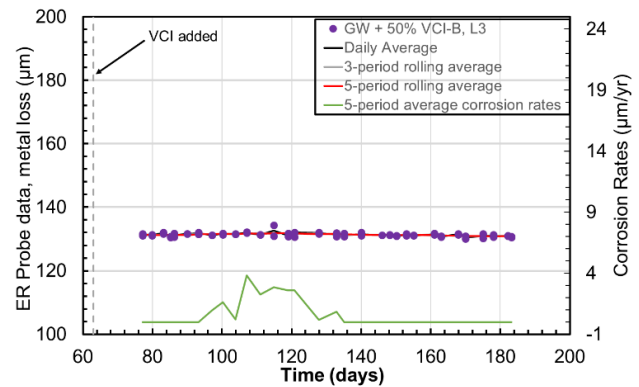


(d) Level 3 ER probe data and corrosion rates

Figure 4: Vessel 2 ER probe data and corresponding corrosion rates



(a) Level 2 ER probe data and corrosion rates



(b) Level 3 ER probe data and corrosion rates

Figure 5: Vessel 2 ER probe data and corresponding corrosion rates

The Vessel 1 ER-probe-derived corrosion rates are consistent with coupons' corrosion rates in immersed, Level 1, and Level 2, as listed in Table 5. However, Level 3 ER-probe-derived corrosion rate was found to be not consistent with the coupons' corrosion rate. The 3-period-rolling-average and 5-period-rolling-average-ER-probe-derived corrosion rates are 81 and 67 $\mu\text{m}/\text{yr}$, respectively, but needed to be close to zero to be consistent with the coupons' corrosion rates. This discrepancy in the ER-probe-derived corrosion rate may be due to the wire element probe that was used at Level 3. The thinness and surface area of the wire probe element was much smaller compared to the cylindrical probes that were used in immersed, Level 1, and Level 2.

The Vessel 2 ER-probe-derived corrosion rates are consistent with coupons' corrosion rates at Level 1 and Level 2, as listed in Table 5. The corrosion rate ratio for the immersed coupons range between 1.3 to 1.8, and the 3-period-rolling-average and 5-period-rolling-average-ER-probe-derived corrosion rates are 22 and 7.5 $\mu\text{m}/\text{yr}$, respectively; the ER probe corrosion rate needed to be at least 33 $\mu\text{m}/\text{yr}$ to be consistent with the coupons' corrosion rate. Similarly, the ER-probe-derived corrosion rate at Level 3 were not consistent with the coupons' corrosion rates; the ER-probe rate was 53 $\mu\text{m}/\text{yr}$, but needed to be approximately 15 $\mu\text{m}/\text{yr}$ or lower to be consistent with coupons' corrosion rates. The use of wire-element probe may have skewed the data at Level 3.

The Vessel 3 ER-probe-derived corrosion rates are consistent with coupons' corrosion rates at Level 2 and Level 3, as listed in Table 5. The corrosion rates' ratio ranges for both Levels 2 and 3 included 3, and ER-probe derived corrosion rates were zero.

Overall, the cylindrical probe elements were used at six locations, and of those, five of them reported the corrosion rates that were consistent with the coupons' corrosion rates. Whereas, the wire-element probes were used at three locations, and only one location's ER probe data were consistent with the coupons' data.

CONCLUSION

Electrical resistance (ER) probes were used to in-situ monitor effectiveness of the vapor corrosion inhibitors. Cylindrical element ER probes were used at six locations and wire element ER probes were at three locations. The coupons were initially exposed to ground water conditions for two months. Several coupons were extracted after two months, and VCIs were added simultaneously. The remaining coupons were then exposed for additional four months. ER probe' were continuously monitored. The ER probe data were collected using a Model A datalogger that malfunctioned after 18 days. Another datalogger, identified as Model B, was used for few weeks. Model A datalogger was reused starting day 77. The ER probe data fluctuated from measurement to measurement. Therefor, 3- and 5-period rolling averages were used to estimate the ER-probes' corrosion rates. A consistency check was carried out between the coupons' and ER-probe derived corrosion rates. The following points are noted:

- The Vessel 1 coupons' corrosion rates were consistent with ER-probe derived corrosion rates in immersed, Level 1 and Level 2. The Vessel 2 coupons' corrosion rates were consistent with ER-probe derived corrosion rates at Level 1 and Level 2. Similarly, the Vessel 3 coupons' corrosion rates were consistent with ER-probe derived corrosion rates at Level 2 and Level 3.
- Vessel 1 Level 3, Vessel 2 immersed, and Vessel 3 Level 3 ER-probe derived corrosion rate were not consistent with the coupons' corrosion rates. Wire-element probes were used at Level 3 of Vessel 1 and Vessel 2. Small surface of the wire elements may have distorted the ER probe data, and corresponding corrosion rates. A cylindrical element ER probe was used in Vessel 2 immersed level; it is not clear why the ER-probe-derived corrosion rates were not consistent with the coupons' corrosion rates.
- ER probe data fluctuated from measurement to measurement. A rolling average method was used to estimate the probes' corrosion rates. It was determined that the 5-period rolling average corrosion rates were closest to the coupons' corrosion rates.
- Six cylindrical-element ER probes were used between the three vessels. Of those, the corrosion rates of the five were found to be consistent with the corresponding coupons' corrosion rates.
- Three wire-element ER probes were used between the three vessels. Of those, only one element's corrosion rate was found to be consistent with the corresponding coupons' corrosion rates.
- The study indicated that the cylindrical element ER probes with larger surface area compared to the wire-element ER probes provide more accurate representation of the bottom plate corrosion rates, and hence, are better suited to in-situ monitor effectiveness of VCIs in the tank bottom application.

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