Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

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Performance of Vapor Corrosion Inhibitors for Localized Corrosion Mitigation of Double Shell Storage Tanks at Hanford

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ABSTRACT

Hanford stores millions of gallons of high-level waste storage site in 27 carbon-steel double shell, underground tanks (DSTs). A secondary shell surrounds the primary shell, where the bottom plate of the secondary shell rests on a grooved concrete pad. There have been instances of metal loss on the secondary shell bottom plates in contact with the concrete basemat where accumulation of the groundwater solution in the pads' grooves may have caused corrosion. In addition, uneven contact between the basemat and shell could create occluded areas where localized corrosion in form of crevice corrosion is possible. In previous studies, several commercially available vapor corrosion inhibitors (VCIs) were tested for their ability to mitigate concrete-basemat side corrosion of the secondary liner bottom. The previous studies involved use of disk coupons either immersed or placed in vapor space of the groundwater solution dosed with VCIs. Even though, VCIs have been found to be effective in mitigating corrosion on the disk coupons, it is not clear if VCIs can be sufficiently effective in mitigating the crevice corrosion. A study was conducted with disk coupons where each coupon's disk surface was partially occluded using a crevice former. Several sets of the disk coupons were exposed to the groundwater solution for several months in three separate corrosion tests. Approximately half of the coupons were extracted after several months of exposure and analyzed for metal loss. The remaining sets of coupons were exposed for additional time to the commercially available VCIs by adding the VCIs to the ground water solutions in the tests. The remaining coupons were extracted after completion of the tests and analyzed for metal loss. Corrosion rate data of the pre- and post-VCI exposed coupons were compared to evaluate effectiveness of the VCIs for mitigating the crevice corrosion. The paper will present experimental data and results on effectiveness of VCIs in mitigating crevice corrosion on the bottom plates of the secondary shells.

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

INTRODUCTION

High-level radioactive waste generated during reprocessing of spent nuclear fuel at Hanford has been stored in several single- and 27double shell tanks (DSTs). Each DST consists of a primary shell (inner) surrounded by secondary (outer) liner. The secondary liner rests on a concrete basemat. The schematic diagram, which shows the concrete basemat and drain slots, is presented in Figure 1.

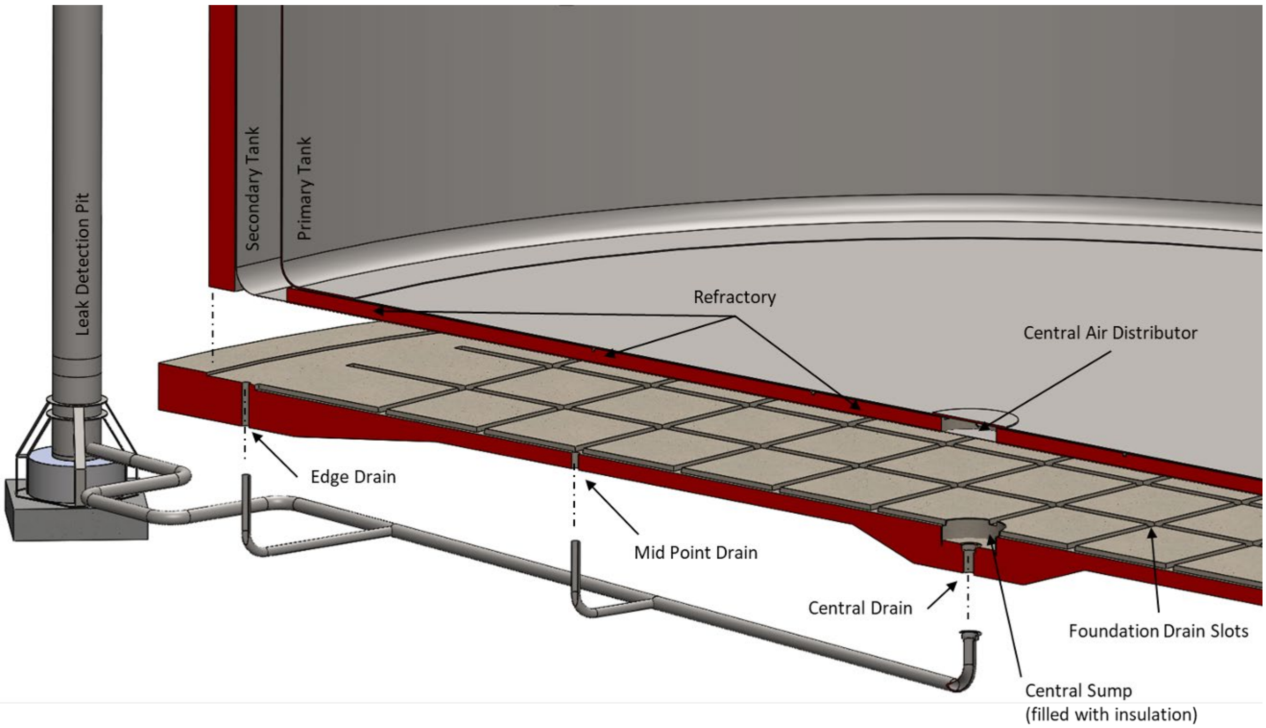


Figure 1: Schematic of a double shell tank depicting primary and secondary tank shells, concrete foundation, and drain slots.

It is postulated that groundwater seeps in and accumulates in the drain slots, and corrodes 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 water 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. The pH of both simulants was adjusted using sodium carbonate and acetic acid to 7.6 after preparation. A previous study established that GW simulant is more corrosive than the leak detection pit, therefore GW was used in the VCI effectiveness study.[1]

Laboratory experiments were conducted to address the concerns of both immersed-phase corrosion and VSC of the tank steel exposed to GW simulant. Vapor corrosion inhibitors (VCIs) were also tested to determine their efficacy in mitigating the corrosion on pre-corroded surfaces. VCIs have been used for corrosion mitigation in numerous applications for decades. However, 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.[2 3 4 5]

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}7E-05
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 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 clear epoxy solution (EpoKwick® from Buehler) so that one face of the coupon was exposed to the test electrolyte. An image showing a test coupon is presented in Figure 2. The coupons' exposed surfaces were modified to simulate crevice corrosion. A crevice former was tightly attached to each coupon surface using tape and wire. The crevice formers partially covered the coupons' surfaces, which created conditions for localized corrosion under the crevice formers.

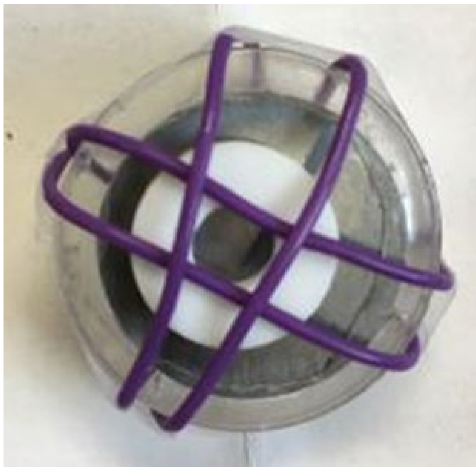
Table 2
Chemical Composition of AAR TC 128 Steel (wt.%)

	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

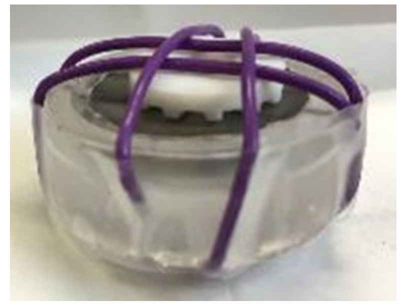
A glass vessel of dimensions 1.0 m (3.3 ft) tall and 14 cm (5.5 inch) diameter was used for each experiment. Approximately 1.25 L of GW simulant was added to the bottom of the vessel for each experiment. Each vessel has a water jacket around the simulant holding area which was used to circulate warm 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 two vessels used is presented in Figure 3(a). Coupons were exposed to the electrolyte and vapors of the electrolyte in each experiment by suspending them using a rod shown in Figure 3(b). 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.

(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.
(3) American Association of Railroads, 425 3rd Street SW, Washington, DC 20024
(4) ASTM International, 100 Barr Harbor Dr., West Conshohocken, PA 19428-2959

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 different heights with respect to the solution using the rods. The coupons' positions, with respect to solution 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:



(a) Top view



(b) Side View

Figure 2: Image of a coupon used in the study. (a) Top, and (b) side views of the partially covered coupon. The coupon's surface is partially covered with a white crevice former, which is held in place using purple wire and tape.

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 shown in Figure 3(b). 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 afterward has infrequent or no contact with the water. However, this region is exposed to the humidified air.

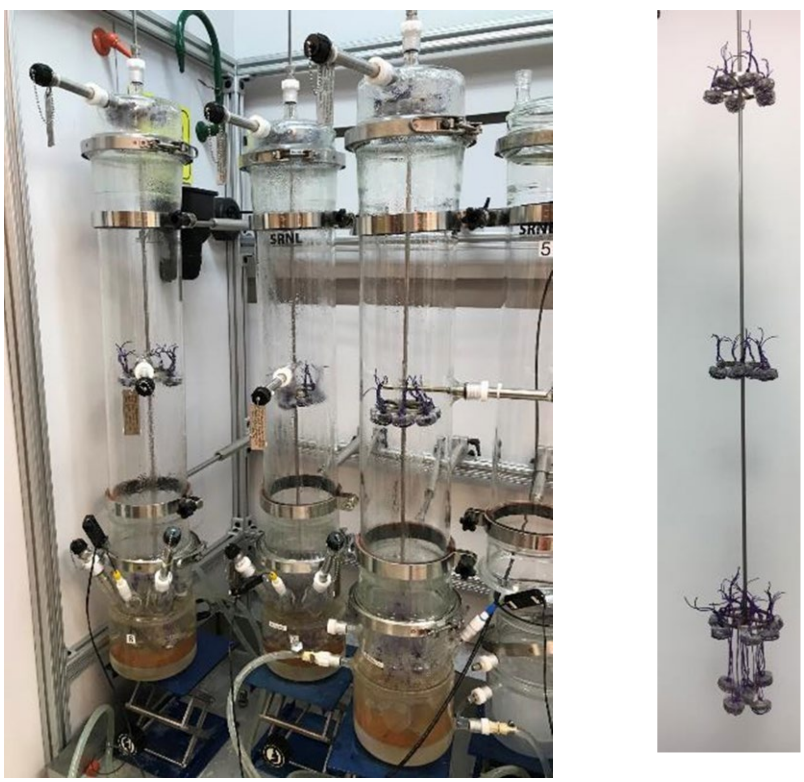
Level 3: Top or high level. This set of coupons was not exposed to the solution prior to testing. The coupons were suspended approximately 91.4 cm (36 inch) above the simulant. This level is representative of the secondary liner bottom plate region that is only exposed to the humidified air and any volatile species from the solution.

Description of the vessels for each solution 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 six months of exposure, cleaned with Clarke's solution[6] to remove corrosion products, and measured for weight losses.

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. A representative image of an exposed coupon and its profiled surface are presented in Figure 4.

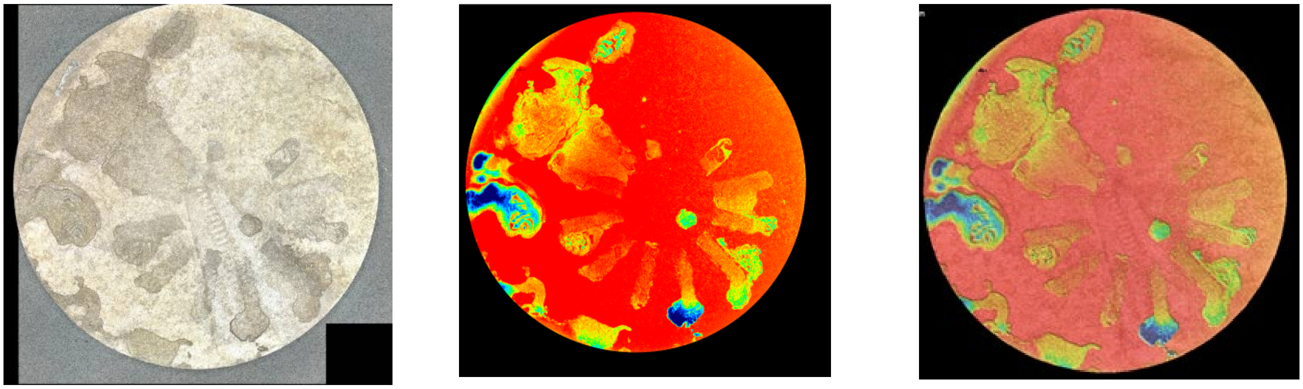
Table 3
Solution Description, Vessel Identification, and Coupons and ER Probe Information

Solution	Corrosion Cell	Notes
Initially GW simulant, and then 100% of the recommended dosage of VCI-A after 2 months	Vessel 1	• 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 2, 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	• 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 2, 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	• 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



(a) (b)

Figure 3: Images of the (a) experimental configuration, and (b) steel rod to suspend the coupons inside the vessel containing electrolyte.



(a) (b) (c)

Figure 4: (a) Image of a representative coupon, (b) false color map, and (c) digital profile of the coupon surface

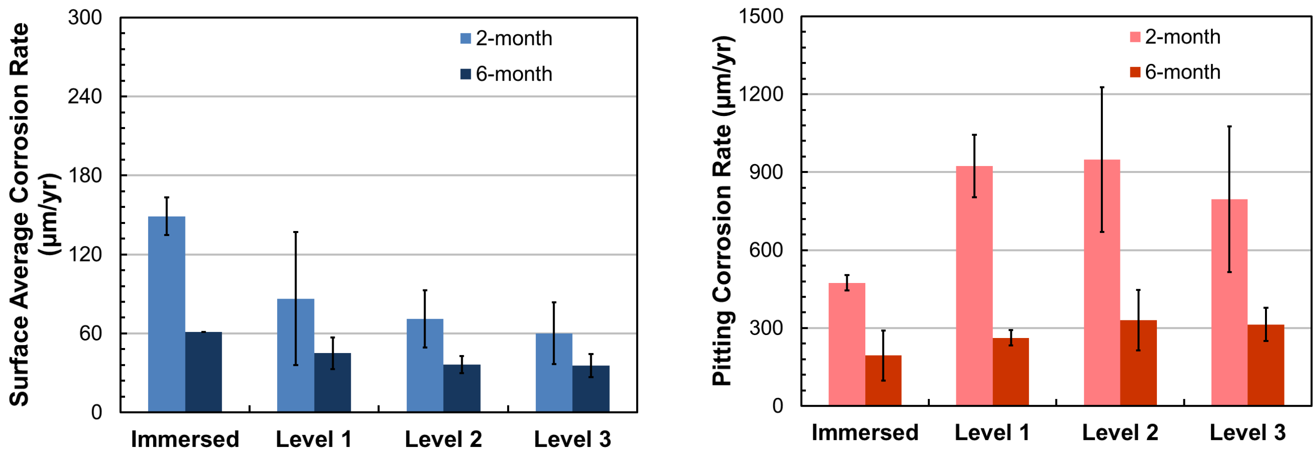
EXPERIMENTAL DATA AND RESULTS

The corrosion rate data for the coupons in Vessel 1, i.e., GW for the first two months and then GW + 100% VCI-A, are listed in Table 4. The table has corrosion rate data, including surface average and pitting corrosion rates for the coupons exposed to GW and then GW plus 100% recommended dosage of VCI-A. The data are for coupons exposed for two and six months; 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 100% VCI-A dosage for additional four months. Table 4 data also include averages of surface average and pitting corrosion rates and corresponding standard deviations; the corrosion rates and standard deviations are presented in Figure 5.

Table 4
Vessel 1 (100% VCI-A After Two Months) Coupon Corrosion Rate Data

Corrosion Type	Corrosion Rate (µm/yr)*							
	Immersed		Level 1		Level 2		Level 3	
	2-month	6-month	2-month	6-month	2-month	6-month	2-month	
Surface Average Corrosion	145	61	58	41	91	43	74	41
	165	61	56	36	74	30	74	41
	137	61	145	58	48	36	33	25
Average** ± std***	149 ± 14.4	61 ± 0	86.4 ± 50.6	44.9 ± 12	71.1 ± 21.7	36.4 ± 6.4	60.1 ± 23.5	35.6 ± 8.8
Pitting Corrosion	457	127	965	279	1168	356	1092	305
	457	152	787	229	1041	432	762	381
	508	305	1016	279	635	203	533	254
Average** ± std***	474 ±29	195 ± 96	923 ± 120	263 ± 29	948 ± 279	330 ± 116	796 ± 281	313 ± 64

*25 µm/yr = 1 mil/yr = 1 mpy
**Average values are calculated for 3 coupons
***std denotes standard deviation of the corrosion rate data used to calculate the average



(a) (b)

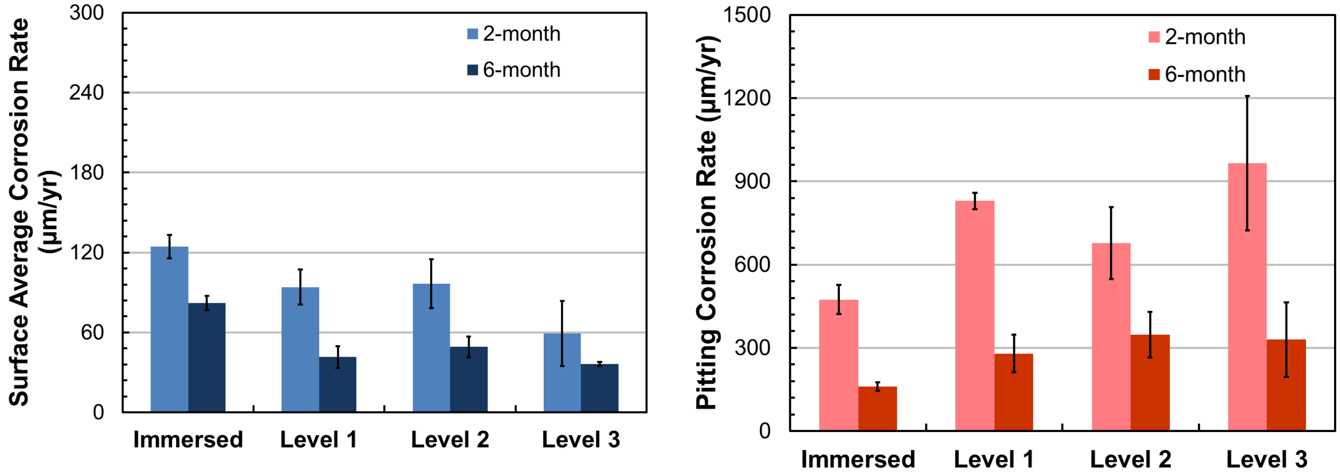
Figure 5: Average of (a) surface average, and (b) pitting corrosion rates for coupons in Vessel 1 with 100% VCI-A after two months. The black line in each bar represents the standard deviation.

The corrosion rate data for the coupons in Vessels 2 and 3 are listed in Tables 5 and 6, respectively. Table 5 has corrosion rate data, including surface average and pitting corrosion rate, for the GW (2-month) and then GW plus 100% recommended dosage of VCI-B (6-month), and Table 6 has the data for the GW (2-month) and then GW plus 50% recommended dosage of VCI-B (6-month). The data are for the 2-month and 6-month coupons. The 2-month coupons were exposed to GW simulant whereas 6-month coupons were exposed to GW for the first two months followed by GW plus VCI-B for additional four months. The table data also include average of surface average and pitting corrosion rates corresponding standard deviation. These average values and standard deviations of the averages are presented in Figures 6 and 7.

Table 5
Vessel 2 (100% VCI-B After Two Months) Coupon Corrosion Rate Data

Corrosion Type	Corrosion Rate (μm/yr)*							
	Immersed		Level 1		Level 2		Level 3	
	2-month	6-month	2-month	6-month	2-month	6-month	2-month	6-month
Surface Average Corrosion	114	84	79	51	112	41	33	38
	130	76	102	36	76	51	64	36
	130	86	102	38	102	56	81	36
Average** ± std***	125 ± 9	82 ± 5	94 ± 13	42 ± 8	97 ± 18	49 ± 8	59 ± 24	36 ± 2
Pitting Corrosion	432	178	864	330	787	254	686	483
	533	152	813	305	711	406	1092	229
	457	152	813	203	533	381	1118	279
Average** ± std***	474 ±53	161 ±15	830 ± 29	279 ± 67	677 ± 130	347 ± 82	965 ± 242	330 ± 134

*25 μm/yr = 1 mil/yr = 1 mpy
**Average values are calculated for 3 coupons
***std denotes standard deviation of the corrosion rate data used to calculate the average



(a) **(b)**

Figure 6: Average of (a) surface average, and (b) pitting corrosion rates for coupons in Vessel 2 with 100% VCI-B after two months. The black line in each bar represents the standard deviation.

A statistical analysis was conducted to determine significance of the decrease in corrosion rate due to addition of 100% recommended dosages of VCI-A and VCI-B, and 50% dosage of VCI-B. The statistical method used was Student's t-test, which is based on the hypothesis that there is no statistically significant difference between the corrosion rates for the two electrolyte parameters used in the t-test—that is, that they are essentially identical to each other in terms of the coupon corrosion rates. The statistical result calculated by the test, P value, is the probability that the hypothesis is true. The higher the P-value, the greater the chance that the two sets of corrosion rates for the 2- and 6-month coupons are statistically similar. If the P-value is equal to or less than 0.05, it indicates that there is a less than 5% chance that the two sets of coupons have similar corrosion rates—that is, it means, with 95% confidence, that there is a statistically significant difference between the two 2- and 6month coupons. The P-values are listed in Table 7.

Table 6
Vessel 3 (50% VCI-B After Two Months) Coupon Corrosion Rate Data

Corrosion Type	Corrosion Rate (µm/yr)*							
	Immersed		Level 1		Level 2		Level 3	
	2-month	6-month	2-month	6-month	2-month	6-month	2-month	6-month
Surface Average Corrosion	99	51	58	25	66	18	48	8
	122	56	74	18	64	66	25	53
	127	46	43	23	51	58	33	79
Average** ± std***	116 ± 15	51 ± 5	58 ± 15	22 ± 4	60 ± 8	47 ± 26	36 ± 12	47 ± 36
Pitting Corrosion	914	305	1168	305	1092	229	1143	254
	1245	686	914	305	584	203	1092	254
	1118	635	813	381	610	203	483	305
Average** ± std***	1092 ± 167	542 ± 207	965 ± 183	330 ± 44	762 ± 286	212 ± 15	906 ± 368	271 ± 29

*25 µm/yr = 1 mil/yr = 1 mpy
**Average values are calculated for 3 coupons
***std denotes standard deviation of the corrosion rate data used to calculate the average

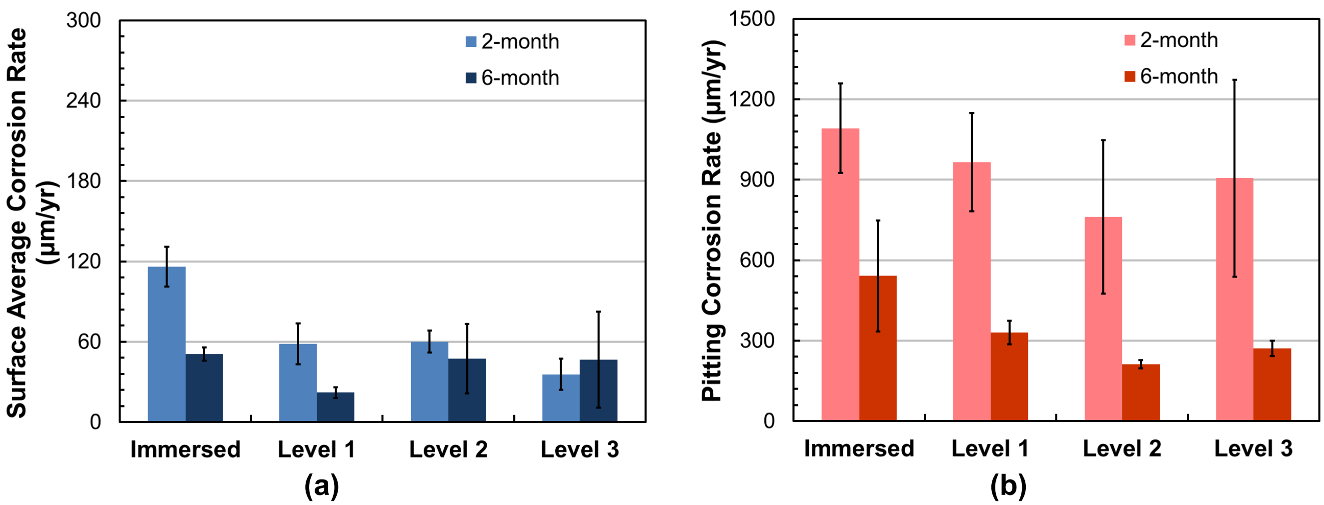


Figure 7: Average of (a) surface average, and (b) pitting corrosion rates for coupons in Vessel 3 with 50% VCI-B after two months. The black line in each bar represents the standard deviation.

Pitting corrosion of the secondary shell is the main hazard for a tank failure, leading to breach of the secondary containment. The P-values listed in Table 7, obtained by statistical analysis of the pitting corrosion rates, are discussed. The P-values for Vessel 1 (100% VCI-A after two months) surface average corrosion rates are higher than 0.05 at Levels 1, 2, and 3, indicating that surface average corrosion was not mitigated with 95% confidence. However, the pitting corrosion rates in Vessel 1 were mitigated in immersed, Levels 1 and 2 coupons with 95% confidence, but only with 91% confidence in Leven 3 coupons.

The P-values for Vessel 2 (100% VCI-B after two months) surface average corrosion rates are lower than 0.05 in immersed, and Levels 1 and 2, indicating that surface average corrosion rate was mitigated with 95% confidence. However, the P-value is 0.25 for the Level 3 coupons, indicating that the corrosion rate was mitigated with only 75% confidence. It is noted that the surface average corrosion rates of the 2-month Level 3 coupons fall in the range of 59 ± 24 µm/yr (2.3 ± 1 mpy) whereas the surface average corrosion rates of the 6-month Level 3 coupons are in the range of 36 ± 2 µm/yr (1.4 ± 0.07 mpy). The pitting corrosion rates in Vessel 2 were mitigated at all levels, i.e., immersed and Levels 1, 2 and 3, with 95% confidence as indicated by the P-values.

Table 7
Student's t-Test P-values* for Comparison Between Coupons Before and After VCI Treatment

Corrosion Cell	Corrosion Type							
	Surface Average Corrosion				Pitting Corrosion			
	Immersed	Level 1	Level 2	Level 3	Immersed	Level 1	Level 2	Level 3
Vessel 1 (100% VCI-A)	0.01	0.29	0.10	0.20	0.03	0.01	0.05	0.09
Vessel 2 (100% VCI-B)	0.00	0.01	0.03	0.25	0.01	0.00	0.03	0.03
Vessel 3 (50% VCI-B)	0.01	0.05	0.49	0.66	0.02	0.02	0.08	0.09
*P-values of 0.05 or less indicate statistically significant differences with 95% confidence								

The P-values for Vessel 3 (50% VCI-B after two months) surface average corrosion rates are lower than 0.05 in immersed, and Level 1, indicating that surface average corrosion rates were mitigated with 95% confidence. However, the P-values are 0.49 and 0.66 for the Levels 2 and 3 coupons, respectively, indicating that the corrosion rates were not mitigated at Levels 2 and 3. It is noted that the surface average corrosion rates of the 2-month Level 2 coupons fall in the range of 60 ± 8 µm/yr (2.4 ± 0.3 mpy) whereas the surface average corrosion rates of the 6-month Level 3 coupons are in the range of 47 ± 26 μm/yr (1.9 ± 1.0 mpy). Similarly, the surface average corrosion rates of the 2-month Level 3 coupons fall in the range of 36 ± 12 μm/yr (1.4 ± 0.5 mpy) whereas the surface average corrosion rates of the 6-month Level 3 coupons are in the range of 47 ± 36 μm/yr (1.9 ± 1.4 mpy). The pitting corrosion rates in Vessel 3 were mitigated in immersed and Level 1 coupons with 95% confidence as indicated by the P-values. However, the P-values are 0.08 and 0.09 for Levels 2 and 3 coupons, respectively; these two P-values indicate that pitting corrosion is mitigated with 90% confidence.

The ER probe data and probe-derived corrosion rates are presented in a companion paper.[7] The probe-derived corrosion rates were either zero or minimal at all levels after addition of VCIs. This further indicated that VCIs were effective in mitigating corrosion on the coupons.

CONCLUSION

A study was conducted to determine effectiveness of two commercially available vapor corrosion inhibitors in mitigating corrosion on bottom side of the secondary liner of the double shell tanks. VCIs were added during mid-course of experiments, i.e., after coupons have experienced corrosion in the untreated simulant. Three tests were conducted using VCI-A and VCI-B. The first two tests were conducted using 100% recommended dosages of VCI-A and VCI-B, and the third test with 50% of the recommended dosage of VCI-B. Following conclusions are made from the experimental data and results:

- The corrosion rates obtained from the coupons exposed to GW only are consistent with the past data.[5]
- Both VCIs were effective in mitigating the pitting corrosion rate in immersed, Levels 1 and 2 coupons at the 100% recommended dosages. VCI-B was effective in mitigating the corrosion rate even at Level 3. VCI-A also mitigated the pitting corrosion rate in the Level 3 coupons, but statistical analysis of the corrosion rate data indicated that the corrosion rate reduction at best has a confidence of 91%, slightly below the 95% level.
- 50% VCI-B was also effective in mitigating the pitting corrosion rate in immersed and Level 1 coupons with 95% confidence. The pitting corrosion rate was also mitigated at Levels 2 and 3 coupons, but the level of confidence was 92 and 91%, respectively, slightly below the 95% level.
- 100% VCI-B was also effective in mitigating the surface average corrosion rate in the immersed, Level 1, and Level 2 coupons with 95% confidence, where as VCI-A was only effective in the immersed coupons with 95% confidence. 50% VCI-B was also effective in mitigating the surface average corrosion rate in the immersed and Level 1 coupons with 95% confidence.

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