

Laboratory Studies Related to External Corrosion of Hanford's Double-Shell Tanks in Contact with Concrete Liner

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ABSTRACT

Radioactive and chemically hazardous wastes are stored at the Hanford site in underground double-shell tanks (DSTs) constructed of carbon steel. The corrosion management of these tanks has mainly focused on the complex waste chemistries that are contained in the primary tank. More recently, attention has been given to the corrosion that has been found on the exterior of the secondary liners of the DSTs. The cause of the external corrosion could be related to the intrusion and accumulation of water in the tertiary leak detection systems, which exist under the concrete foundation of the DSTs. However, site investigations have discovered significant reductions in wall-thickness (up to 70%) for DSTs that do not have a history of water intrusion. Furthermore, site measurements have indicated that wall-thinning may not be as severe at locations where the tank exterior is in contact with the slotted concrete foundation.

Laboratory tests involving periodic wetting and condensing moisture on steel specimens were conducted to better understand this external corrosion threat. A simulated groundwater solution of near-neutral pH served as the wetting or moisture source in the tests. The laboratory tests consisted of weight-loss evaluations along with more involved studies using a multi-electrode array (MEA). The effect of concrete on the corrosion of steel was also examined by incorporating areas of metal-to-concrete contact on the weight-loss specimens and MEA. The results of this testing revealed that corrosion rates, calculated from weight-loss, can underestimate wall-thinning due to the severe localized attack that was found. Also, the inhibition of corrosion due to concrete-contact may relate more to the obstruction of condensation on the steel rather than an influence on the local pH.

Keywords: Hanford, carbon steel, vapor phase corrosion, pitting, multi-electrode array, concrete.

INTRODUCTION

The Hanford site contains approximately 55 million gallons (2.08×10^8 liters) of radioactive and chemically hazardous wastes arising from weapons production beginning with World War II and continuing through the Cold War era. The wastes are stored in 177 carbon steel underground storage tanks, of which 149 are single-shell tanks (SSTs) and the remaining are double-shell tanks (DSTs). Historically, tank failures have been associated with the SSTs. However, in 2012, a failure was encountered for the first time in

one of the DSTs: Tank AY-102. The failure of AY-102 occurred from corrosion of the primary liner¹, which allowed the waste to leak into the annular space between the primary liner and the secondary liner. The waste never reached the surrounding soils due to the containment provided by the secondary liner.

Although the failure of AY-102 was related to the primary liner of the tank, one of the outcomes or recommendations from the failure investigation was to perform ultrasonic testing (UT) on the secondary liners as part of the core inspection practices for the DSTs. The need for routine inspection is further underscored by the fact that there is a reported history of water ingress and accumulation in the leak detection pit (LDP) systems, which reside on the external side of the secondary liners.² The LDPs are tertiary containment systems that are designed to collect any liquid waste draining from beneath the tanks in the event of a secondary liner breach. The LDP designs vary depending on the tank farm but share the common feature of having drain slots that channel liquid into a leak detection well or pit. The drain slots for the LDPs are provided by the DST foundations, which are slotted concrete pads.

The water levels in the LDP systems have been tracked since the time when the DSTs were placed in service. Many of the LDPs have exhibited a history of water intrusion, which has required pumping to reduce the water level below specified limits. For a few tanks, the LDP water level managed to exceed the structural limit, which means that the LDP drain line and foundation slots were filled and water was in contact with the bottom of the secondary liner structure.² For one of the tanks (SY-103), the water level remained above the structural limit for approximately 4 years. At this time, there is not an obvious connection between the severity of water accumulation in the LDPs and the extent of wall thinning (as determined by UT inspections). For example, Tank AP-102 has shown the greatest wall loss to date (~70% wall loss) even though it has experienced low levels of water intrusion based on its LDP records.³ Furthermore, some of the tanks that have experienced regular accumulations in the LDP have not yet received UT scans of their secondary liner. According to a 2019 document⁴, UT scans of the secondary liner have been performed on 15 of the 27 DSTs that are active. Even though these scans have been limited to the annulus floor of the DSTs, 13 have shown reportable wall loss (loss > 10% of nominal thickness). The reportable wall loss of the secondary liners is localized in nature.³ For Tank AP-102, there was at least one location of severe wall thinning that showed evidence of pitting. It has also been found that the wall thinning tends to align with the slots of the concrete foundation.³ Although it is known that concrete provides an alkaline environment for protecting embedded steel (e.g., reinforcing steel), it is not certain if the tank areas that are resting on the concrete foundation should be less susceptible to corrosion due to local alkalinity effects. It is also not clear why the wall thinning has not developed more uniformly given the non-passivating type of environment.

The work presented in this paper consists of laboratory tests aimed at understanding the localized wall-thinning that has been observed in some of the secondary liners. Electrochemical measurements along with weight-loss evaluations were used to assess the corrosion rates of carbon steel under vapor phase and periodic immersion conditions. Most of the testing incorporated some form of concrete in contact with the steel specimens/electrodes to evaluate whether concrete could help protect the steel from corrosion. The pH of concrete and the effect of groundwater pH on steel passivity were also examined.

EXPERIMENTAL

Steel Compositions and Test Specimens

American Association of Railways Tank Car (AAR TC) 128 Grade B steel⁽¹⁾ (TC-128) was used in the experimental work to simulate the legacy steel used in the construction of the double-shell tanks. TC-128 steel is similar in composition to the UNS K02401 Grade 60 steel used in the Hanford AY-101 and AY-102 tanks. The chemical composition of TC-128 steel, as well as the specified limits for UNS K02401 steel, are shown in Table 1. Cylindrical test specimens (25.4 mm length x 4 mm diameter) and bullet-

⁽¹⁾ This steel does not have a corresponding UNS number.

shaped specimens (38 mm length x 4.8 mm diameter) were fabricated from plates of the TC-128 steel. The cylindrical test specimens were used for weight-loss studies in the vapor phase corrosion test while the bullet-shaped specimens were used for cyclic potentiodynamic polarization (CPP) tests. Both specimen types were polished to a 600-grit finish and degreased in acetone prior to testing.

Due to the unavailability of TC-128 steel as an insulated wire product, a generic carbon steel was used for fabricating the multi-electrode array (MEA). The wire consisted of an Alloy 1010 core and a thin sheathing of polyimide polymer, which provided electrical isolation of the wires. The diameter of the steel core in each wire was 0.5 mm. The total diameter of the wire, including sheathing, was 0.56 mm. The chemical composition of the annealed, Alloy 1010 wire is shown in Table 2.

Table 1: Chemical composition of AAR TC 128 Grade B steel (TC-128 steel) and specified limits for legacy steel (UNS K02401).

Steel	Element, weight %									
	C	Mn	P	S	Si	Cu	Cr	Ni	V	Fe
TC-128	0.21	1.03	0.01	0.01	0.06	0.04	0.05	0.02	0.07	Bal.
UNS K02401*	0.24	0.9	0.04	0.04	0.15 - 0.40					

*Max. allowable, unless otherwise noted

Table 2: Chemical composition of Alloy 1010 carbon steel wire.

Element, weight%					
C	Mn	P	S	Si	Fe
0.08	0.31	0.04	0.042	0.061	Bal.

Concrete Material

A vendor that specializes in concrete research and testing was commissioned to prepare the concrete test blocks in conformance with the general specifications of the structural concrete used in the DST foundations. Based on the available information for the original concrete formulation, test blocks were prepared to the following specifications:

- Constituents
 - Type II Portland Cement – Meeting the ASTM⁽²⁾ C150⁵ specification
 - Gravel Coarse Aggregate, 1.5 in. Maximum Size – Meeting the ASTM C33⁶ specification
 - Natural Sand – Meeting the ASTM C33 Specification
- Preparation
 - Air entraining admixture meeting the ASTM C260⁷ Specification
 - Concrete prepared in accordance with ASTM C192⁸ Specification
 - Air Entrained with an Air Void Content of 3 to 6 %
 - Slump of 2 to 4 inches.
- Compressive strength of 4500 psi (minimum) at 28 days (the foundation concrete used in the 241-AP tank farm required a minimum 28-day compressive strength of 4,500 psi.⁹)

Figure 1 shows photographs of “wafers” (102 mm diameter, 25.4 mm thick) machined from the cast concrete cylinders. The square markings on the wafers indicate where concrete test blocks were extracted for corrosion testing use.

⁽²⁾ ASTM International (ASTM), 100 Barr Harbor Dr., West Conshohocken, PA 19428-2959.

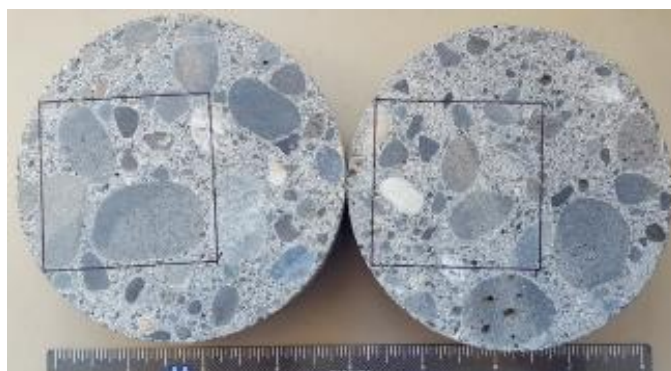


Figure 1: Photograph of concrete wafers cut from cast concrete cylinders.

Simulated Groundwater Chemistries

Analyses of groundwater samples that were collected at the Hanford site indicate that the groundwater contains low concentrations of ionic species and has a slightly alkaline pH (7.9 to 8.4).¹⁰ According to the analyses, bicarbonate is the dominate species, but other prominent ions include calcium, sodium, sulfate, chloride, and nitrate. None of the analyses of groundwater samples from the site indicated the presence of nitrite, which acts as a corrosion inhibitor toward carbon steel.

Table 3 contains the chemical composition of the groundwater simulants that were used for corrosion testing. The standard groundwater simulant shown in the table (Column 2) was based on the chemistry of the groundwater samples that were collected at the Hanford site. Accordingly, it has a pH near neutral and was used for the vapor phase corrosion tests involving weight-loss specimens and the MEA. The concrete-contacted groundwater (CCGW) simulants shown in the table (Column 3 and Column 4) were higher pH versions of the standard groundwater simulant. They were intended to simulate groundwater conditions in which the pH was elevated due to interaction with concrete. The CCGW simulants were only used for CPP testing of carbon steel specimens.

Table 3: Chemical compositions of groundwater simulants.

Simulant ID	Standard Groundwater	CCGW (pH 10)	CCGW (pH 11)
Source Chemical	Concentration (mol/L)		
NaHCO ₃	1.75E-03	1.75E-03	1.75E-03
Ca(OH) ₂	1.50E-03	1.50E-03	1.50E-03
KNO ₃	2.40E-04	2.40E-04	2.40E-04
Fe ₂ (SO ₄) ₃	6.25E-04	6.25E-04	6.25E-04
FeCl ₃	7.67E-05	7.67E-05	7.67E-05
Sr(NO ₃) ₂	2.87E-06	2.87E-06	2.87E-06
Na ₂ SiO ₃ ·9H ₂ O	6.00E-04	6.00E-04	6.00E-04
MgCl ₂ ·6H ₂ O	3.10E-04	3.10E-04	3.10E-04
C ₂ H ₄ O ₂	3.00E-04	3.00E-04	3.00E-04
NaOH	--	3.08E-03	8.84E-03
Measured pH at 45°C	8.3	10.3	11.0

Experimental Procedure for Vapor Phase Corrosion Testing

A schematic of the experimental setup that was used for vapor phase corrosion testing is shown in Figure 2. The test setup consisted of a solution reservoir and a peristaltic pump, which allowed the solution level

to be raised inside the primary vessel when the pump was active. The specimens were positioned at three levels or “zones” within the primary vessel. Each zone was associated with a specific type of exposure condition: the specimens in Zone 1 were exposed to alternate-immersion conditions (1-hr soak per day); the specimens in Zone 2 were briefly pre-soaked in the simulant and then restricted to vapor phase exposure; the specimens in Zone 3 were exposed to the vapor phase without pre-soaking. The pump was programed to cycle on for 1 hour of each day. When the pump was operating, the solution level increased above the weight-loss specimens in Zone 1 and the multi-electrode array (MEA), both of which were positioned about 2 inches above the stagnant solution level. Under stagnant conditions (pump-off), the solution volume inside the primary vessel was around 2 liters while the reservoir volume was approximately 4 liters. The solution inside the primary vessel and reservoir were both controlled at 45°C using heating bands, thermocouples, and temperature controllers. The test temperature of 45°C was selected based on the reported temperature range (30 to 45°C) beneath the waste tanks at the Hanford site.

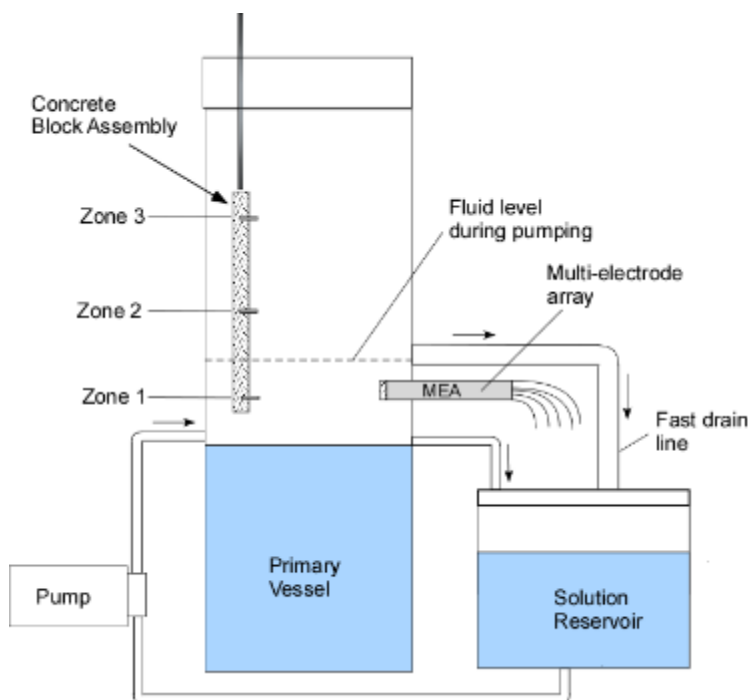


Figure 2: Schematic of experimental setup used for vapor phase corrosion testing.

Testing of weight-loss specimens

Cylindrical-shaped specimens made of TC-128 steel were used for weight-loss determinations in the vapor phase corrosion test. The weight-loss specimens were partially embedded in concrete blocks to evaluate whether corrosion would be inhibited in the specimen areas that contacted the concrete. Figure 3 is a schematic of the positioning of each specimen in the concrete blocks. Nine weight-loss specimens were embedded in the concrete blocks by drilling holes that were approximately the same diameter as the specimens. The holes were drilled to a depth of around 13 mm to allow half the length of each specimen to be embedded in concrete while the other half was boldly exposed. The embedded half of the specimens were lightly pressed into the holes to create relatively tight contact between the steel and concrete. Triplicate weight-loss specimens were aligned at three different levels in the concrete to create the separate exposure zones. All the specimens were exposed for a period of 4 weeks. After the test, the specimens were carefully removed from the concrete blocks and then cleaned with an inhibited acid