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Corrosion of Low-Carbon Steel Under Environmental Conditions at Hanford: Two-Year Soil Corrosion Test Results

Prepared for the U.S. Department of Energy Office of Environmental Restoration and Waste Management



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

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Date Published November 1995

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1. INTRODUCTION

At the Hanford Site, located in southeastern Washington state, nuclear production reactors were operated from 1944 to 1970. The handling and processing of radioactive nuclear fuels produced a large volume of low-level nuclear wastes, chemical wastes, and a combination of the two (mixed wastes). These materials have historically been packaged in U.S. Department of Transportation (DOT) approved drums made from low-carbon steel, then handled in one of three ways:

- A. Before 1970, the drums were buried in the dry desert soil. It was assumed that chemical and radionuclide mobility would be low and that the isolated, government-owned site would provide sufficient protection for employees and the public.
- B. After 1970, the drums containing long-lived transuranic radionuclides were protected from premature failure by stacking them in an ordered array on an asphalt concrete pad in the bottom of a burial trench. The array was then covered with a large, 0.28-mm- (011-in.-) thick polyethylene tarp and the trench was backfilled with 1.3 m (4 ft) of soil cover. This burial method is referred to as soil-shielded burial. Other configurations were also employed but the soil-shielded burial method contains most of the transuranic drums.
- C. Since 1987, US Department of Energy sites have complied with the *Resource Conservation and Recovery Act of* 1976 (RCRA) regulations. These regulations require mixed waste drums to be stored in RCRA compliant large metal sheds with provisions for monitoring. These sheds are provided with forced ventilation but are not heated or cooled.

The current drums meet DOT Type 17H or 17C specifications. The most recent drums also meet the new DOT performance-based packaging standards. The 208-L (55-gal) drums are liquid-tight through use of gaskets and sealants and are painted with a chemically resistant epoxy on the outer surfaces. A bolted ring securely attaches the lids of these drums.

To determine how long waste materials might reasonably be contained and how much corrosion might be present during planned retrieval efforts for certain waste classes, the corrosion rate of low-carbon steel in the environments associated with the three types of drum storage outlined above must be determined.

Westinghouse Hanford Company (WHC), in collaboration with the Pacific Northwest Laboratory (PNL), initiated corrosion experiments on drum material coupons a) exposed to the atmosphere in large metal storage sheds and b) buried in Hanford soil, simulating the environmental conditions expected during storage as described in storage methods A to C above (Bunnell et al., 1994). This report

focuses only on the experiments performed simulating the burial of drums deep in the Hanford soil, as described in the storage method A above.

The soil corrosion experiments were initiated in 1993 with exposure times ranging from 0.5 yr to 16 yrs. The experiments included specimens of bare carbon steel, painted carbon steel, painted and damaged carbon steel (to simulate the corrosion of both non-damaged and damaged drums), and alternative materials, viz., galvanized carbon steel and AISI Type 304L stainless steel (304L). Corrosion coupons exposed for 0.5 yr (actual time of exposure was 0.75 yr) and 1 yr were retrieved from the soil in 1994 and evaluated for corrosion (Duncan and Bunnell, 1995). The present report describes the results obtained on corrosion coupons exposed to Hanford soil for 2 yrs.

2. EXPERIMENTAL METHOD

A total of six shafts (five peripheral shafts for specimens and a central shaft for instrumentation) were drilled in 1993 to accommodate burial times of up to 16 yrs. The original plan was to drill six circumferential shafts surrounding the central shaft in a hexagonal pattern. Figure 1 outlines the planned. retrieval times, differing by factors of two, and the shaft layout. As indicated in the figure, the sixth circumferential shaft was not drilled. Instead, after the 1-yr specimens were removed from the 1-yr shaft in July 1994, 16-yr specimens were placed in the same shaft to avoid the expense of decommissioning the shaft and drilling a new shaft. Details of instrumentation in the central shaft were described by Bunnell et al. (1994) previously.

Specimens measuring 2.5-cm by 6-cm (1-in. by 4-in.) of bare carbon steel, painted steel, damaged painted steel, 304L, and galvanized steel were emplaced at known depths to 9.1 m (30 ft). The specimens were attached with nylon screws to hexagonal polyethylene blocks to avoid galvanic corrosion. One specimen of each type at each of four depths, for six planned retrieval times for a total of 24 duplicate specimens were emplaced. Figure 2a shows a hexagonal block with specimens installed. To protect specimens from damage during retrieval drilling, these specimen blocks were inserted into 0.6-m (2-ft) lengths of slotted polyvinyl chloride (PVC) casing (see Figure 2b). During specimen emplacement, soil removed from the intended burial depth was carefully packed around the hexagonal block inside the casing, and the assemblies were lowered into shafts drilled to the desired depths. Ground vibrations associated with extraction of drill casings are assumed to have packed the surrounding soil to a density approaching undisturbed soil.

Specimens were retrieved by using a core-barrel drilling rig, in which a heavy-walled steel pipe was driven into the soil and then pulled to the surface with the soil column it contained. The pipe was oversized relative to the original shaft diameter of 30 cm (12 in.); the original shafts were drilled within 5 cm (2 in.) of plumb over their length. Thus, the retrieval technique was to simply redrill the original shaft at a slightly larger diameter to ensure the specimens would be captured.

3. RESULTS AND DISCUSSION

To summarize previous retrievals, the nominal 6-mo retrieval was accomplished in March 1994, after an actual exposure time of 9 months. Specimens located 1.5 and 3 m (5 and 10 ft) below grade were retrieved easily. Those at the 6.1-m (20-ft) level were more difficult to retrieve; one specimen was lost and others were damaged. The soil around the specimens at the 9.1-m (30-ft) level was very dry, which made it difficult to pull to the surface. Smaller-bore core barrels were tried, but the specimens and their plastic carrier were destroyed. Retrieval of the 1-yr specimens was accomplished in July 1994, but once again one of the specimen sets (buried 1.5-m deep) was destroyed during drilling.

The retrieval of the 2-yr specimens was performed on September 20 and 21, 1995. All specimens were retrieved. The major variation that occurred was that no specimens were found at 1.5-m (5-ft). Instead, specimens were found at 3-m, 4.6-m, 6.1-m, and 9.1-m (10-ft, 15-ft, 20-ft, and 30-ft). No indication has been found in the records as to why this placement of coupons occurred.

During retrieval, it was observed that the soil at about 0.6 to 1-m was extremely wet. Below that the soil was much drier with the driest soil between about 3- and 6-m. Below that it had a noticeable moisture content but not to the degree noted earlier. These observations are considered consistent with the cooler, wetter summer experienced in 1995. The soil from 0' to 14' contained medium size cobbles, gravel and brown sand. The soil from 14'-15' depth consisted of brown sand and a small amount of silt. The soil retrieved from 14'-30' depth again contained a small amount of silt with lighter brown sand.

After retrieval, specimens were photographed mounted on their polyethylene blocks, then removed and examined. Specimens of bare low-carbon steel and 304L were cleaned of corrosion products by using inhibited hydrochloric acid. Specimens were dried and then weighed to determine the amount of metal lost to corrosion.

Table 1 presents the 2-yr exposure results and Table 2 shows the summary of results to date. Based on the 2-yr data, weight-loss measurements indicated that bare low-carbon steel was corroded by contact with the soil at rates from 15.2 to 35.6 microns(μ m)/yr (0.6 to 1.4 mil/yr [mpy]). These values lie between the extremes observed at 9-mo and slightly higher than the values observed at 1-yr. As mentioned earlier, the 2-yr specimens were retrieved from 10', 15', 20' and 30' depths. If we assume that the 9-month and 1-yr specimens were also located at the same depths (instead of 5', 10', 20' and 30' depths as reported previously), the agreement in corrosion rates between the 3 sets of measurements for bare carbon steel is much better.

Weight-loss measurements also indicated that 304L was not corroded to a measurable extent in the 2-yr exposure.

The bare low-carbon steel displayed a rough surface typical of underground corrosion after 2-yr of exposure (see Figure 3). The specimen located at 3-m had one spot on an edge that might be indicative of pitting but the location on the

edge of the specimen prevented any determination of depth. From the appearance of the uncleaned specimens, this was the only carbon steel specimen that may have actually pitted.

The 304L coupons showed no signs of pitting with the exception of the one at 9.1-m. It had small pits, 13- to $38-\mu m$ (0.5- to 1.5-mils) deep, at the bottom end of the coupon on the back. Therefore, it is likely that it was due to crevice corrosion.

The last column of Table 2 shows corrosion rates of low-carbon steel as indicated by electrical resistance probes placed at various soil depths in the central shaft (see Figure 1) located at about 6.1 m (20 ft) from the shaft from which the 2-yr specimens were extracted. These measurements indicate that corrosion rates varied with depth and ranged from about 25 to 50 μ m/yr (1 to 2 mpy). This is the same general range as that measured by weight-loss methods. However, the correlation at various depths varied, with the probes running typically 13 to 25 μ m/yr (0.5 to 1 mpy) higher than the coupons. Because the coupons showed a rough texture of the corroded surface, it is assumed the probes also underwent a similar variability in corrosion on each coupon. As a consequence, it is reasonable to assume that the 125 μm (5 mil) thick steel foil has been penetrated in some locations which would effectively raise the resistance of the probe and indicate a higher resistance. On this basis, in accordance with the original design, the probes should start to fail in the near future. Nevertheless, both sets of data showed a trend of decreasing corrosion rates with depth and do not give contradictory results.

Upon initial examination, some painted low-carbon steel specimens showed paint damage in small areas, but lack of corrosion on the steel surfaces indicated that the damage occurred by mechanical means during retrieval. There were several other small areas showing degradation, possibly indicating some damage despite care during installation. The epoxy paint used on the drum metal is strong but quite brittle and is subject to damage when contacted by hard objects such as rocks.

Even after 2-yr, the damaged painted steel specimens showed very little corrosion of the exposed steel, and paint lifting had not occurred at the paint-steel interface. Corrosion was apparently too localized to spread laterally or lift paint. These characteristics are likely to be observed in specimens exposed for longer periods.

The galvanized steel specimens at the two upper levels were black suggesting corrosion is in process. The two lower level specimens showed only slight and uniform dulling of the surface, suggesting that little zinc was lost to corrosion.

The 304L specimens showed no visible corrosion; pitting occurred only on the lowest specimen. It occurred only on the back of the coupon and appears to be more of a case of crevice corrosion even though very localized.

- 4. CONCLUSIONS
- 1. After two years of exposure to soil at the Hanford Site, bare low-carbon steel appears, by both weight-loss measurements and electrical resistance probes, to be corroding at about 15 to 35 μ m/yr (0.6 to 1.4 mpy). In general, the carbon steel corrosion rate decreases with increasing depth.
- 2. The 304L corrodes at a much lower rate than low carbon steel, 0.08 to 0.2 μ m/yr (0.003 to 0.008 mpy) in Hanford soil and shows some pitting, 13 μ m to 38 μ m (0.5 to 1.5 mil) deep after two years of exposure on the deepest specimen.
- 5. SUMMARY

Tests are presently under way to measure the corrosion of bare low-carbon steel, painted low-carbon steel, painted and damaged low-carbon steel, and possible alternative container materials. The tests focus on attacks by either the atmosphere or by soil at the Hanford Site. Alternative materials include galvanized steel and 304L. Both of these tests include sensors to characterize the environment surrounding the exposed specimens.

Soil corrosion is proceeding at a measurable and approximately constant rate on low-carbon steel. This rate ranges from 15 to 35 μ m/yr (0.6 to 1.4 mpy) and decreases with increasing depth. The 304L corrodes at less than 1 μ m/yr in the soil environment but appears to suffer some crevice attack on the deepest specimen.

In general, corrosion has occurred at a very low rate in both carbon steel and 304L, reflecting the relatively benign and dry environments at the Hanford Site. The corrosion rates are less than those suggested by previous work as summarized by Divine (1991). These results are applicable to other direct buried steels at the Hanford Site, viz., transfer lines, burial boxes etc.

6. REFERENCES

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Duncan, D. R., Bunnell, L. R., Measurements of the Corrosion of Low-Carbon Steel Drums Under Environmental Conditions at Hanford: One-Year Test Results, WHC EP 0859, May 1995.

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TABLE 1

WHC-EP-0891

Corrosion Rate Calculations for Two Year Specimens Recovered in September 1995	9/20/95 10 ft & 15 ft 9/21/95 20 ft & 30 ft	Corrosion Rate, mpy	ty = 7.86 g/cc ^A (Area exposed to soil = 4 in ²)	1.4	1.0	0.4	0.6	d to soil = 7.5 in^2)	2.9E-03	3.6E-03	7.5E-03	4.9E-03
		Exposure Time, days		824	824	825	825		824	824	825	825
		Area [*] , cm ²		25.8	25.8	25.8	25.8		48.4	48.4	48.4	48.4
	Recovered: 9/ 9/	Wt Loss, g		1.6492	1.2025	0.4416	0.6872	ty = 7.94 g/cc^ (Area exposed to soil	0.0065	0.0080	0.0165	0.0108
		Final Wt#, g		24.7675	25.0055	26.2389	26.0053		55.3379	55.4863	53.5453	54.0547
		Initial Wt*, g Carbon steel - densi	26.4167	26.2080	26.6805	26.6925	Stainless steel – densi	55.3444	55.4943	53.5618	54.0655	
	6/18/93	Coupon No.†	Ca	13	6	ω	m	Staiı	19	16	21	10
	Buried:	Depth, ft		10	15	20	30		10	15	20	30

t based on notches observed on retrieved specimens

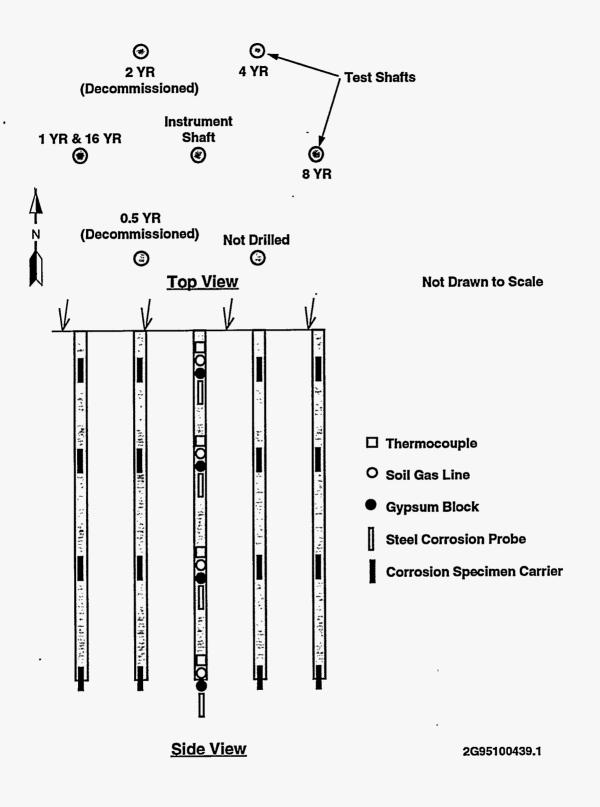
from Corrosion testing Lab report, adjusted for wt loss from the cleaning process

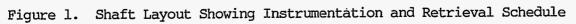
^A from NACE Corrosion Engineer's Reference Book, RS Treseder, Ed, NACE, Houston, TX, 1980

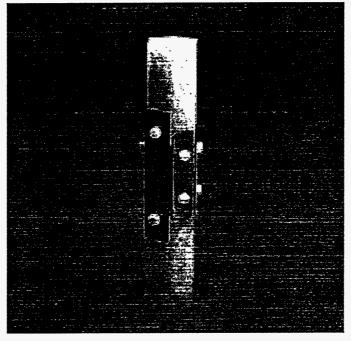
Note: Area used neglects clipped corners and notches. To be consistent with previous work the area of the mounting holes is neglected - $\approx 4\%$ for carbon steel and $\approx 2\%$ for the stainless steel.

TABLE 2

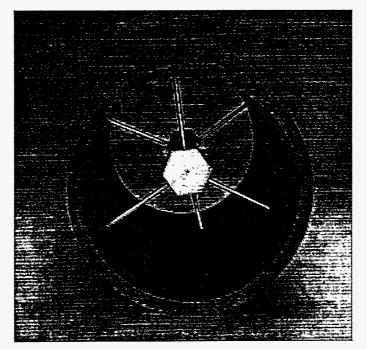
Comparison of 2-yr Data with 9-mo and 1-yr Data							
Depth (ft)	Corrosion Rate (mpy)						
	9-mo	1-yr	2-yr	Average	Probes		
Carbon Steel	······································	· · · · · · · · · ·	· · · · · · · · · · · · · · · · · · ·				
· 5	1.7			1.7	2.1		
10	0.9	1.0	1.4	1.1	2.1		
15			1.0	1.0			
20	0.3	0.6	0.4	0.4	1.5		
30		0.2	0.6	0.4	0.9		
Stainless Steel							
5	0.0065			0.0065			
10	0.0096	0.0210	0.0029	0.0112			
15			0.0036	0.0036			
20		0.0180	0.0075	0.0127			
30		0.0190	0.0049	0.0119			







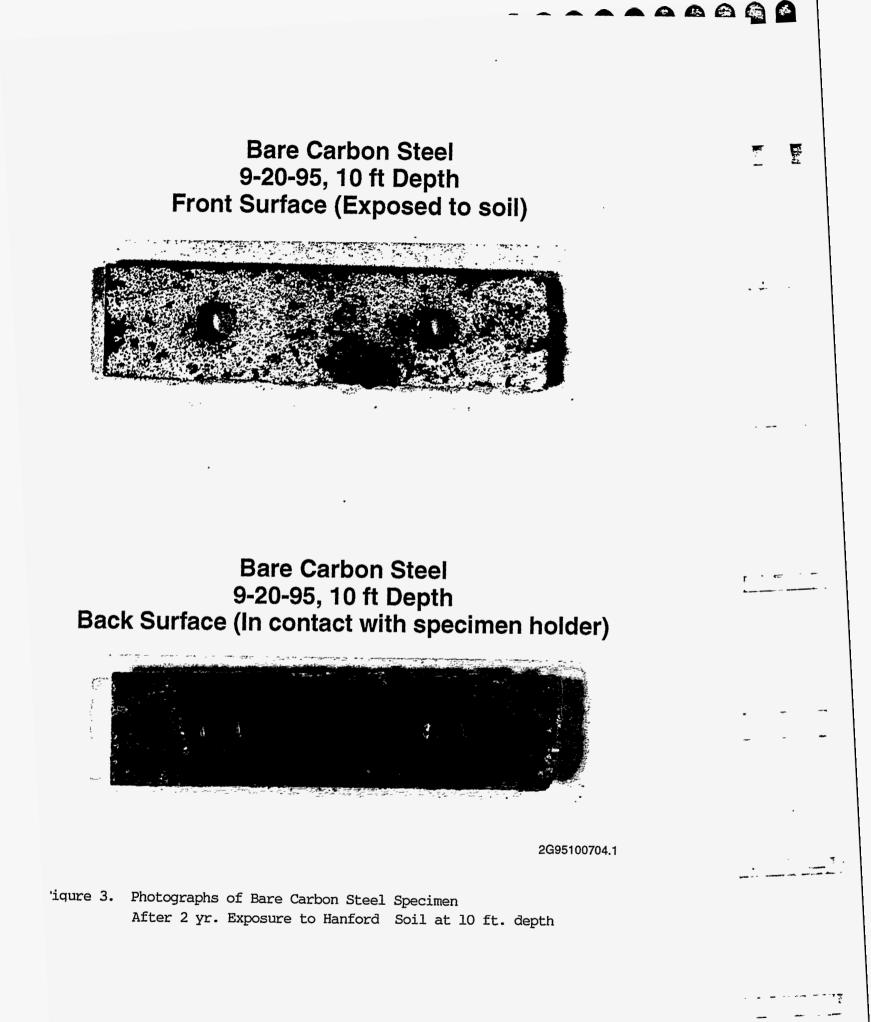
a. Specimens Attached to Polyethylene Block



b. Polyethylene Block (with Specimens) Mounted in PVC Pipe, Ready for Burial

2G95110079.1

Figure 2. Specimen Assembly Prior to Burial in Soil



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