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UNDERGROUND CORROSION OF ACTIVATED METALS IN AN ARID VADOSE ZONE ENVIRONMENT

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ABSTRACT

The subsurface radioactive disposal site located at the Idaho National Engineering and Environmental Laboratory contains neutron-activated metals from nonfuel nuclear-reactor-core components. A long-term corrosion test is being conducted to obtain site-specific corrosion rates to support efforts to more accurately estimate the transfer of activated elements in an arid vadose zone environment. The tests use nonradioactive metal coupons representing the prominent neutron-activated material buried at the disposal location, namely, Type 304L stainless steel, Type 316L stainless steel, nickel-chromium alloy (UNS NO7718), beryllium, aluminum 6061-T6, and a zirconium alloy. (UNS R60804). In addition, carbon steel (the material presently used in the cask disposal liners and other disposal containers) and a duplex stainless steel (UNS S32550) (the proposed material for the high-integrity disposal containers) are also included in the test program. This paper briefly describes the test program and presents the early corrosion rate results after 1 year and 3 years of underground exposure.

Keywords: beryllium, stainless steel, aluminum, zirconium alloys, vadose zone, neutron-activated metals, nuclear reactor components, microbiologically induced corrosion, soil analysis, underground corrosion, soil gas.

INTRODUCTION

The long-term corrosion test is designed to assist in the determination of site-specific corrosion rates of neutron-irradiated metals buried in an arid vadose zone environment at the Idaho National

Engineering and Environmental Laboratory. Corrosion rates are based on mass loss from metal coupons exposed to underground site conditions. The corrosion rates, once determined, reduce the uncertainty of the site-specific transfer of radioactive isotopes to the environment (radiological release rates). Of interest are the metals used to fabricate nuclear reactor components that, when exposed to high neutron fluxes in a reactor environment, become activated with long-lived radioactive isotopes. After disposal, corrosion processes can cause these radioactive isotopes to be released from the irradiated metallic waste to the environment.

The long-term corrosion testing includes direct corrosion testing (i.e., burying metal coupons in the soil); soil characterization, sampling, and analysis for physical, chemical, hydraulic, and microbiological properties; and monitoring of field conditions, including precipitation, soil moisture, soil pore water, and soil-gas composition. The direct corrosion testing provides corrosion rate data, while the soil characterization and field monitoring aid in the evaluation and comparison of the corrosion results to other studies and conditions at the disposal site.

The long-term corrosion test is situated in an arid vadose zone environment adjacent to the subsurface radioactive disposal site where the neutron-activated metals are buried. Results so far consist of corrosion rates after 1 year and 3 years of exposure. The use of nonradioactive metal coupons assumes that activation does not significantly affect corrosion characteristics or corrosion mechanisms.

EXPERIMENTAL PROCEDURE

Direct corrosion testing using buried coupons is the most widely used and simplest method of underground corrosion testing.[1,2,3] Dimensions and mass are measured for metal coupons of known compositions before burial. The direct testing uses nonradioactive coupons of various metals and alloys selected to generally represent the irradiated metals buried at the disposal site. The materials included in the direct testing are Type 304L stainless steel, Type 316L stainless steel, welded Type 316L stainless steel, nickel-chromium alloy (UNS NO7718), beryllium, aluminum 6061, and zirconium alloy(UNS R60804). In addition, low-carbon steel (the material presently used in the disposal liners of the 55-ton scrap casks and other disposal liners and containers) and duplex stainless steel (a proposed material for construction of high-integrity disposal containers) are included as part of the test. The corrosion coupons are 3 × 3 × 1/8 in. (7.62 × 7.62 × 0.32 cm) with a 0.56 in. (1.42 cm) diameter hole in the center. In general, the coupon surface finish is 120 grit (see Figure 1); however, the beryllium coupons, with a 125 RMS finish (see Figure 2), have the same surface finish as the beryllium waste disposed of at the disposal site. Twelve sets of 36 coupons were prepared and slated for testing. One complete set of coupons is stored and maintained as an archived set for comparison with the timed tests.

The long-term corrosion testing began in 1997 when a berm was constructed near the underground disposal site to test corrosion rates at two distinct depths: 4 ft (1.22 m) below surface and 10 ft (3.05 m) below surface. A 6-ft (1.83-m) diameter auger (see Figure 3) was used to drill holes in the berm for placement of the coupons. The coupons were installed as arrays, each consisting of a set of 36 coupons (four samples of each metal type) assembled on polypropylene rods with polytetrafluoroethylene (PTFE) tubing as spacers between coupons. Polypropylene and PTFE were selected because these materials are chemically inert and will electrically isolate the coupons from each other. Each end of the polypropylene rod has engraved PTFE identification markers and is secured with a threaded nylon nut. A coupon array (see Figure 4) was placed at each depth at four locations.

After 1-year exposure to underground corrosion conditions, coupons were removed and examined in 1998.[4] Coupons exposed to 3 years of underground corrosion conditions were removed and examined in 2000.[5] The coupons were recovered by reopening the hole manually and with the auger drill. The coupons were extracted carefully from the hole, with care not to lose the adhering soil around them (see Figure 5). The excavated coupons were double-bagged (see Figure 6) and transported to the appropriate laboratory and disassembled, and the corrosion products, including the potential presence of microorganisms, were sampled.

The vadose zone hydrology, physical soil parameters, soil gas, and soil resistivity were monitored before and during the corrosion testing. The timing and extent of soil characterization work depended on funding availability and research needs.

The coupon cleaning process is designed to remove all corrosion products from the coupons. The mass of the coupon after corrosion and cleaning is compared to the original mass, and the difference represents the loss of metal to corrosion. All coupons were cleaned with a washing/brushing process according to the requirements of ASTM G 1,[6] using deionized water and a nonmetallic soft bristle brush. The duplex stainless steel, 304L stainless steel, 316L stainless steel, nickel-chromium alloy, and zirconium alloy coupons required no further cleaning. The carbon steel, aluminum, and beryllium coupons were chemically cleaned according to the appropriate method defined in Table A1 of ASTM G 1 (in addition to the wash/brush process) as follows: carbon steel—C.3.5, aluminum—C.1.1, and beryllium —C.5.2 (as recommended by the material vendor).

After the coupons were cleaned, they were weighed on a precision balance. The mass was subtracted from the original mass of the coupon (before exposure) to calculate the mass loss due to corrosion, and the corresponding corrosion rate was calculated. The coupons were also examined with a stereomicroscope for localized corrosion and pitting.

RESULTS

Two coupon arrays were retrieved in the fall of 2000 and two in the fall of 1998. The results of the corrosion evaluations are presented here. Corrosion rates and the potential for microbiologically induced corrosion were examined for each coupon. In all, 72 three-year coupons and 72 one-year coupons were recovered, cleaned, and weighed. The average corrosion rates for each metal type are presented in Table 1 and Table 2 for coupons buried at 4-ft (1.22-m) and 10-ft (3.05-m) depths, respectively. A notation of "Not reportable" indicates that no significant mass loss was measured. The reported corrosion rates consider general corrosion of an assumed mathematically flat surface, so as to adequately describe the amount of metal loss from the samples.

The aluminum, beryllium, and carbon steel coupons all experienced pitting to some degree. Figures 7 and 8 are photographs of the front and back of a carbon steel coupon before cleaning (from the 10-ft (3.05-m) depth), and Figures 9 and 10 show the same specimen, front and back, after cleaning. Likewise, Figures 11 and 12 are photographs of the front and back of a beryllium coupon before cleaning (from the 10-ft [3.05-m] depth). Figures 13 and 14 show the same specimen, front and back, after cleaning. Pitting was evident in three metal types: aluminum, beryllium, and carbon steel. Table 3 details the pit depth measurements from six coupons with visibly deep-pitted surfaces.

An examination of the coupons and the adhering soil was performed to determine microbial presence. Microorganisms were found on the surface of all examined coupons as well as in the surrounding soil. From the 1-year coupons, the presence of heterotrophic microorganisms, some of which are organic acid producers, was identified. There was no indication of denitrifying bacteria or the mineral acid producing *Thiobacillus thiooxidans (T. thio.).* Although sulfate-reducing bacteria were found in the soil, none were detected on the 1-year coupons. The 3-year coupons showed similar results, with the exception of sulfate-reducing bacteria colonizing some but not all of the carbon steel, aluminum, and beryllium coupons.

Environmental factors known to influence underground corrosion have been included in the test. Results from the evaluation of some of these factors follow. Soil resistivity, as measured according to ASTM G 57, is reported as 8,500 to 10,000 ohm-cm.[7] This soil resistivity results in a mild soil corrosivity rating for carbon steel according to the work of Palmer.[8] The soil type is mildly alkaline (pH 8.1-8.3). Soil moisture is dependent on depth and location, and at the test location, it varies seasonally at the shallow depth (4 ft [1.22 m]) between 15 and 25%. At the greater depth (10 ft [3.05 m]) the soil moisture is approximately 25%. Other testing has further characterized the soluble ions and exchangeable cations.[7] The temperature at depth (10 ft [3.05 m]) has been measured at around 40°F (11°C). Soil gas sampling is useful in classification of microorganisms, and samples can be taken at the test location in proximity to the underground coupon locations.

CONCLUSIONS

Of the various metals subjected to long-term corrosion testing and evaluated after one year and three years of underground exposure, carbon steel and beryllium exhibited the highest corrosion rates, with higher corrosion rates on coupons at greater depth (10 ft [3.05 m]). Corrosion rates for coupons composed of aluminum, austenitic stainless steel (Type 304L and Type 316L), nickel-chromium alloy, and duplex stainless steel were low but detectable after 3 years. Corrosion rates for the zirconium alloy coupons were very low, below detection limits in most cases.

Of interest is the underground corrosion behavior of beryllium, a metal for which the authors are unaware of any available underground corrosion data. The potential release rate of specific long-lived radionuclides from buried, activated beryllium directly influences the management of the radioactive burial site and its subsequent closure and remediation.

Pitting caused by corrosion was evident on the carbon steel, beryllium, and aluminum coupons. The contributions of pitting rather than uniform corrosion, particularly with beryllium, are significant. In instances where pitting occurs, the coupon evaluations must include pit characterization (i.e., pit geometry) for the results to be meaningful. There are several possible methods for pit characterization, including surface profiling using vertical scanning interferometry and metallography.

Further investigation is needed to understand the differences between the corrosion conditions at the test location and those at the actual disposal location. Such investigations will necessitate continuing monitoring of the arid vadose zone environment, especially for those factors that influence corrosion. Additional soil characterization, soil moisture monitoring, and other monitoring at both the test location and at the disposal site need to be completed for comparison and data correlation purposes.

Indications of microorganism presence were found on the surface of all the examined coupons. Results indicate an increasing occurrence of organic acid producing microbial species colonizing the coupon surfaces along with sulfate-reducing bacteria colonization of carbon steel, aluminum, and in particular beryllium. The environment is suitable for the promotion of microbiologically induced corrosion. By inference, microbiologically induced corrosion should be expected at the disposal site and might have significant impact on the calculated rate of activated metal corrosion.

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TABLE 1
MATERIAL CHEMISTRY

Metal		Chemical Composition, Weight %		
Aluminum alloy	Al: BALANCE	C: 0.227	Cr: 0.145	Fe: 0.480
6061-T6	Mg: 0.955	Mn: 0.089	Ni: 0.007	Si: 0.644
	Ti: 0.021	Zn: 0.048		
Carbon steel	Al: 0.054	C: 0.163	Cr: 0.018	Fe: BALANCE
1018	Mn: 0.787	Mo: 0.004	N: 0.999	Ni: 0.008
	P: 0.010	S: 0.009	Si: 0.010	Ti: 0.001
	V: 0.002			
Nickel-chromium alloy	Al: 0.620	B: 0.004	C: 0.040	Co: 0.240
UNS NO7718	Cr: 18.410	Cu: 0.220	Fe: BALANCE	Mn: 0.120
	Mo: 3.150	Nb: 5.400	Ni: 52.7000	P: 0.011
	S: 0.002	Si: 0.110	Ta: 0.030	Ti: 1.120
316L stainless steel	C: 0.010	Co: 0.140	Cr: 16.490	Cu: 0.290
	Fe: BALANCE	Mn: 1.790	Mo: 2.060	N: 0.034
	Ni: 10.170	P: 0.030	S: 0.013	Si: 0.380
Zirconium alloy	Al: 38 PPM	B: 0.25 PPM	C: 146 PPM	Ca: 10 PPM
UNS R60804	Cd: <0.25 PPM	Cl: 5 PPM	Co: <1 PPM	Cr: 1190 PPM
	Cu: 25 PPM	Fe: 2210 PPM	H: 7 PPM	Hf: 64 PPM
	Mg: 10 PPM	Mn: 25 PPM	Mo: 10 PPM	N: 32 PPM
	Na: 5 PPM	Nb: 50 PPM	Ni: 35 PPM	O: 1300 PPM
	P: 8 PPM	Pb: 25 PPM	Si: 96 PPM	Sn: 15400 PPM
	Ta: 100 PPM	Ti: 25 PPM	U: 1 PPM	V: 25 PPM
	W: 50 PPM			
304L stainless steel	C: 0.020	Co: 0.100	Cr: 18.230	Cu: 0.390
	Fe: BALANCE	Mn: 1.760	Mo: 0.400	N: 0.086
	Ni: 8.250	P: 0.030	S: 0.016	Si: 0.410
Duplex stainless steel	C: 0.010	Cr: 25.200	Cu: 1.940	Fe: BALANCE
UNS S32550	Mn: 1.040	Mo: 3.100	N: 0.210	Ni: 5.880
	P: 0.018	S: 0.002	Si: 0.400	
Beryllium	Al: 0.030	Be: 99.000	C: 0.050	Fe: 0.100
	Mg: <0.010	Si: 0.020		

TABLE 2
CORROSION TEST DATA—4 FOOT (1.22 m) BELOW SURFACE

Composition	1-YEAR AVERAGES Mass Loss (g)	1-YEAR AVERAGES Corrosion Rate (mm/y)	3-YEAR AVERAGES Mass Loss (g)	3-YEAR AVERAGES Corrosion Rate (mm/y)
Aluminum 6061	0.0013	3.895×10^{-5}	0.0218	9.969×10^{-5}
Beryllium	0.0470	2.007×10^{-3}	0.0919	4.635×10^{-4}
Carbon Steel	0.3121	3.175×10^{-3}	0.6893	3.087×10^{-3}
Duplex stainless steel	Not reportable	Not reportable	0.0022	7.620×10^{-6}
Nickel-chromium alloy	Not reportable	Not reportable	0.0025	8.255×10^{-6}
304 L Stainless Steel	Not reportable	Not reportable	0.0018	6.350×10^{-6}
316L Stainless Steel	Not reportable	Not reportable	0.0027	8.890×10^{-6}
316L SS Welded	Not reportable	Not reportable	0.0014	5.080×10^{-6}
Zirconium alloy	Not reportable	Not reportable	Not reportable	Not reportable

TABLE 3
CORROSION TEST DATA—10 FOOT (3.05 m) BELOW SURFACE

Composition	1-YEAR AVERAGES Mass Loss (g)	1-YEAR AVERAGES Corrosion Rate (mm/y)	3-YEAR AVERAGES Mass Loss (g)	3-YEAR AVERAGES Corrosion Rate (mm/y)
Aluminum	Not reportable	Not reportable	0.0051	4.53×10^{-5}
Beryllium	0.1098	4.540×10^{-3}	0.5072	7.248×10^{-3}
Carbon Steel	0.6453	6.350×10^{-3}	3.3061	1.131×10^{-2}
Duplex stainless steel	0.0011	1.101×10^{-5}	0.0018	5.715×10^{-6}
Nickel-chromium alloy	Not reportable	Not reportable	0.0036	1.079×10^{-5}
304 L Stainless Steel	Not reportable	Not reportable	0.0025	8.255×10^{-6}
316L Stainless Steel	Not reportable	Not reportable	0.0036	1.206×10^{-5}
316L SS Welded	Not reportable	Not reportable	0.0022	7.620×10^{-6}
Zirconium alloy	Not reportable	Not reportable	0.0008	5.080×10^{-6}

TABLE 4
CORROSION TEST DATA—PITTING MEASUREMENTS

Composition	Exposure Time (year)	Test Depth (ft)	(m)	Pit Depth (µm)
Carbon Steel	1	4	1.22	141
Beryllium	1	4	1.22	153
Aluminum	3	4	1.22	209
Aluminum	3	10	3.05	188
Carbon Steel	3	4	1.22	121
Carbon Steel	3	10	3.05	167

FIGURE 1. 304L stainless steel coupon.

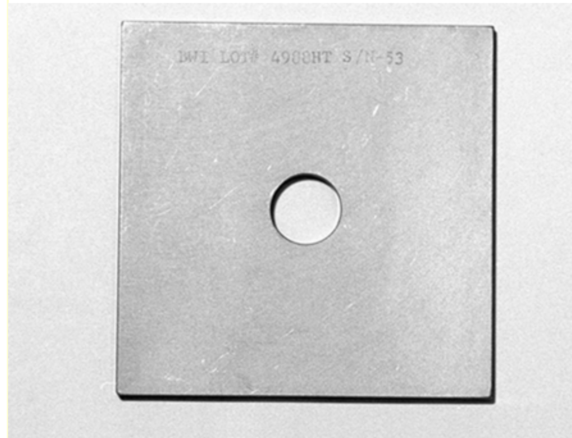

FIGURE 2. Beryllium coupon.

FIGURE 3. 6-ft diameter auger drill.

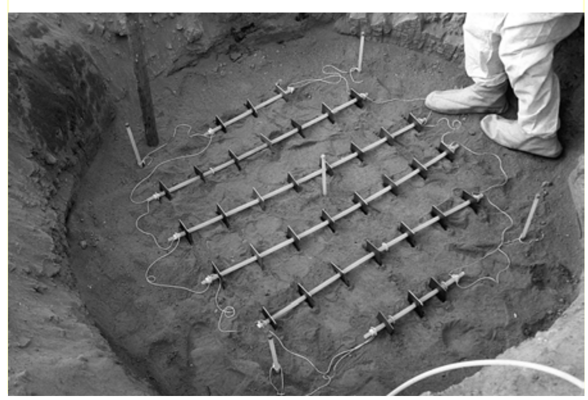

FIGURE 4. Coupon array.

FIGURE 5. Carbon steel coupn on rod.

FIGURE 6. Double bagging coupons.

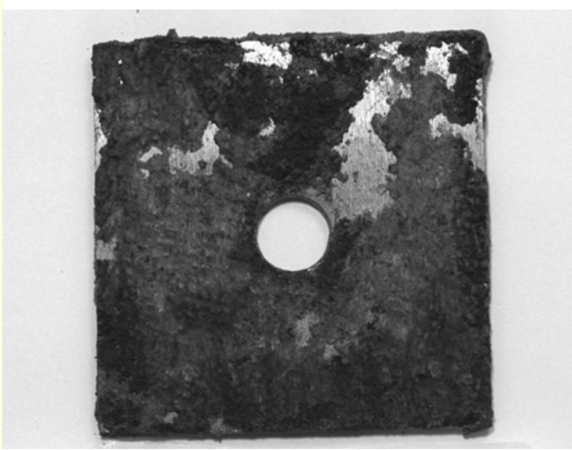

FIGURE 7. Carbon steel front - uncleaned.

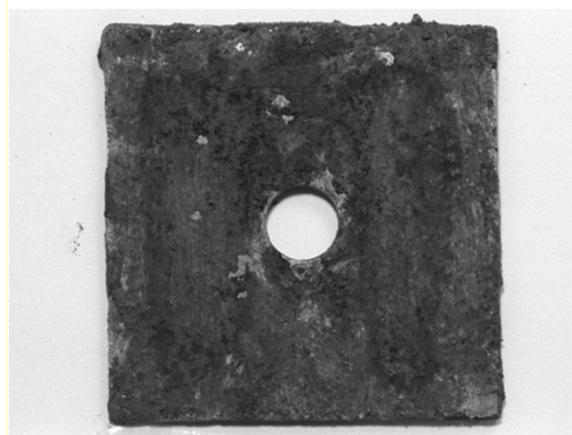

FIGURE 8. Carbon steel back - uncleaned.

FIGURE 9. Carbon steel front - cleaned.

FIGURE 10. Carbon steel back - cleaned.

FIGURE 11. Beryllium front - uncleaned.

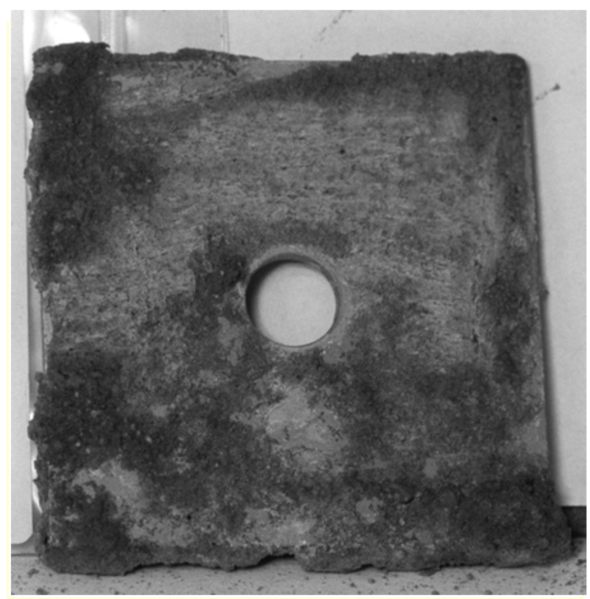

FIGURE 12. Beryllium back - uncleaned.

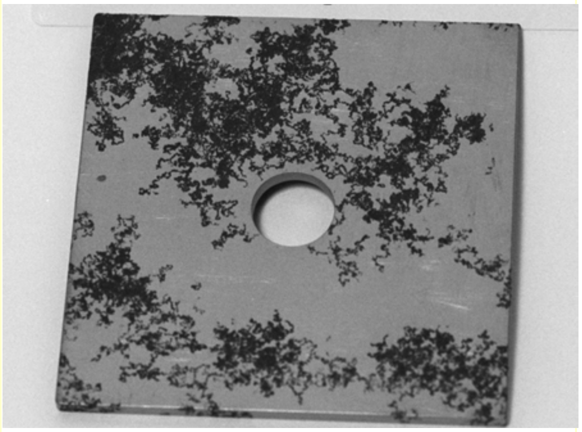

FIGURE 13. Beryllium front - cleaned.

FIGURE 14. Beryllium back - cleaned.