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Twelve Year Study of Underground Corrosion of Activated Metals

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M. Kay Adler Flitton
Timothy S. Yoder

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M. Kay Adler Flitton
CH2M-WG Idaho, LLC
Idaho National Laboratory
P.O. Box 2010
Idaho Falls, ID 83403-2010
United States of America

Timothy S. Yoder
Battelle Energy Alliance
Idaho National Laboratory
P.O. Box 1625
Idaho Falls, ID 83415-2095
United States of America

ABSTRACT

The subsurface radioactive disposal facility located at the U.S. Department of Energy's Idaho site contains neutron-activated metals from non-fuel nuclear-reactor-core components. A long-term corrosion study 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 study uses non-radioactive metal coupons representing the prominent neutron-activated material buried at the disposal location, namely, two types of stainless steels, welded stainless steel, welded nickel-chromium steel alloy, zirconium alloy, beryllium, and aluminum. Additionally, carbon steel (the material used in cask disposal liners and other disposal containers) and duplex stainless steel (high-integrity containers) are also included in the study. This paper briefly describes the test program and presents the corrosion rate results through twelve years of underground exposure.

Key words: beryllium, aluminum, zirconium alloy, duplex stainless steel, stainless steel, nickel-chromium alloy, neutron-activated metals, nuclear reactor components, underground corrosion, microbiologically induced corrosion, arid vadose zone.

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 radioactive disposal facility at Idaho National Laboratory. Corrosion rates are based on total mass loss from nonradioactive 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 study includes parameters known to influence underground metal corrosion. As well as direct corrosion testing (i.e., burying metal coupons in the soil) and real-time monitoring (i.e., using electrical resistance corrosion probes), 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 are part of the test program. The direct corrosion testing and real-time monitoring provide corrosion rate data, while the soil characterization and field monitoring aid in the evaluation and comparison of the corrosion results to other underground corrosion studies and conditions at the disposal facility. The results, presented here, consist of corrosion rates through twelve years of testing for the direct corrosion testing.

EXPERIMENTAL PROCEDURE

Direct corrosion testing using buried coupons is the most widely used and simplest method of underground corrosion testing.[1-3] The direct testing uses nonradioactive coupons of various metals and alloys selected to generally represent the irradiated metals buried at the Idaho National Laboratory site disposal facility. The materials included in the direct testing are Type 304L stainless steel (UNS[a] S30403), Type 316L stainless steel (UNS S31603), welded Type 316L stainless steel, nickel-chromium alloy (UNS NO7718), beryllium, aluminum 6061-T6 (UNS A96061), and zirconium alloy (UNS R60804). In addition, low-carbon steel (UNS G10180 similar material commonly used in disposal liners and containers) and duplex stainless steel (UNS S32550, a high-integrity disposal container material) are included as part of the test. Table 1 has the material properties of each metal type used in the direct testing. The corrosion coupons are 3 x 3 x 1/8 in. (7.62 x 7.62 x 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 (averaging RMS 4.3 µm); however, the beryllium coupons, with a 125 RMS finish (averaging 1.3 µm), have the same surface finish as the beryllium waste disposed of at the disposal site. Twelve sets of 36 coupons (4 of each metal type) 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. After carefully measuring and recording the dimensions and mass of each coupon, the coupon sets were placed in drill holes made in the berm. The 10 ft (3.05 m) set was placed first, the hole was backfilled with soil to the next test depth at 4 ft (1.22 m), the second set was placed, the hole was then backfilled with soil to the surface. This process was completed at 4 test 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] Coupons exposed to 6 years of underground corrosion conditions were removed and examined in 2003.[6] The most recent coupons to be recovered, in 2009, were exposed 12 years to underground corrosion conditions.[7]

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.[8] The duplex stainless steel, 304L stainless steel, 316L stainless steel, nickel-chromium alloy, and zirconium alloy coupons required no further cleaning. Figure 1 shows a typical Type 316L stainless steel coupon after 12-years of exposure and Figure 2 shows the same coupon after 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 (as recommended by the material vendor)[9] - C.5.1 (for the coupons exposed 1 year) and C.1.1 (for the coupons exposed 3, 6, and 12 years).

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[a] Unified Numbering System for Metals and Alloys (UNS). UNS numbers are listed in Metals & Alloys in the Unified Numbering System, 10th ed. (Warrendale, PA: SAE International and West Conshohocken, PA: ASTM International, 2004).

Table 1
Material Chemical Properties

Aluminum (UNS A96061) %
Al: Balance, C: 0.227, Fe: 0.480, Mg: 0.955, Mn: 0.089, Ni: 0.007, Si: 0.644, Ti: 0.021, Zn: 0.048
Low-Carbon Steel (UNS G10180) %
Al: 0.054, C: 0.163, Cr: 0.018, Fe: Balance, S: 0.009, Mo: 0.004, N: 0.999, Ni: 0.008, Ni: 0.008,
P: 0.010, Mn: 0.787, Si: 0.010, Ti: 0.001, V: 0.002
Nickel-chromium Alloy (UNS NO7718) %
Al: 0.620, B: 0.004, C: 0.040, Co: 0.240, Cr: 18.410, Cu: 0.220, Fe: Balance, Mn: 0.120, Mo: 3.150,
Nb: 5.400, Ni: 52.700, P: 0.011, S: 0.002, Si: 0.110, Ta: 0.030, Ti: 1.120
Stainless Steel (UNS S31603) %
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
Stainless Steel (UNS S30403) %
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
Zirconium Alloy (UNS R60804) ppm
Al: 38, B: 0.25, C: 146, Ca: 10, Cd: <0.25, Cl: 5, Co: <1, Cr: 1190, Cu: 25, Fe: 2210, H: 7, Hf: 64,
Mg: 10, Mn: 25, Mo: 10, N: 32, Na: 5, Nb: 50, Ni: 35, O: 1300, P: 8, Pb: 25, Si: 96, Sn: 15400,
Ta: 100, Ti: 25, U: 1, V: 25, W: 50
Duplex Stainless Steel (UNS S32550) %
C: 0.010, Cr: 25.200, Cu: 1.940, Fe: Balance, 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

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 and vertical scanning interferometry for localized corrosion and pitting.

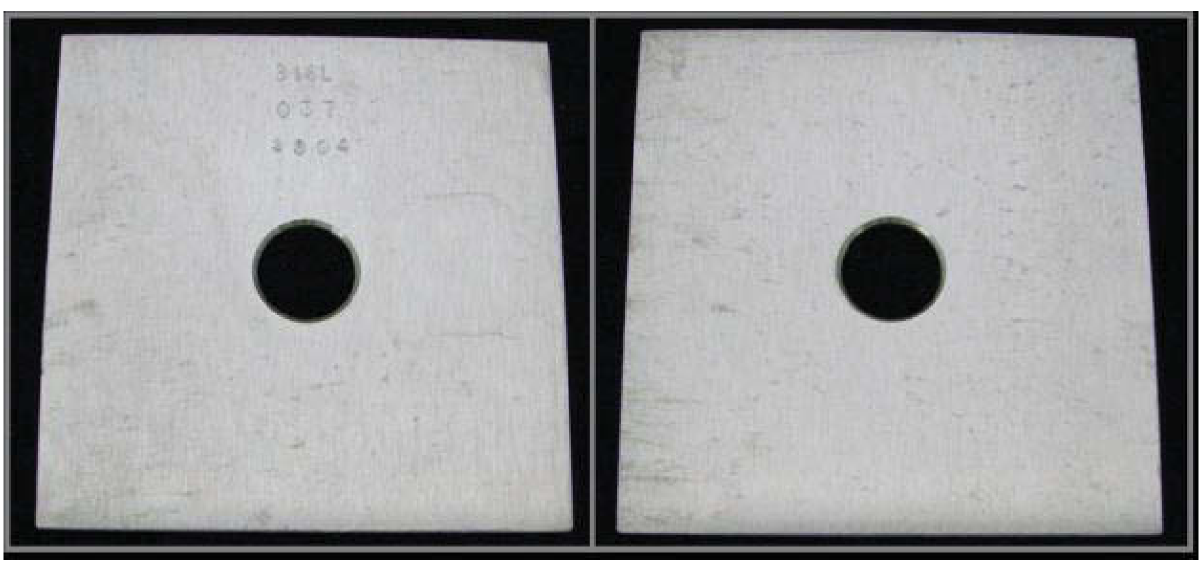

Figure 1: Type 316L stainless steel before cleaning, front and back views.

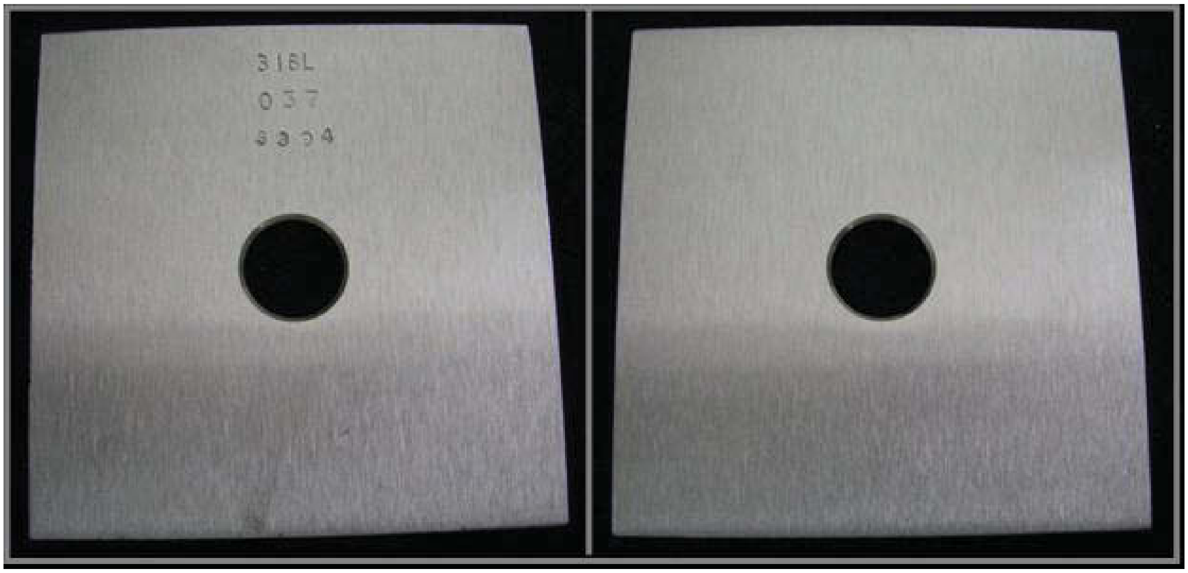

Figure 2: Type 316L stainless steel after cleaning, front and back views.

RESULTS

Two coupon sets were retrieved in the fall of 2009, two in the fall of 2003, two in the fall of 2000, and two in the fall of 1998. The results of the corrosion evaluations are presented here. In all, 288 coupons were recovered, cleaned, and weighed. The average corrosion rates, by exposure year, for each metal type are presented in Table 2 and Table 3 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.

TABLE 2
AVERAGE CORROSION RATE (mm/y)—4 FOOT (1.22 m) BELOW SURFACE

Metal	1-Year	3-Year	6-Year	12-Year
UNS A96061	3.895E-05	9.969E-05	3.841E-05	3.111E-05
Beryllium	2.007E-03	4.635E-04	2.160E-04	1.581E-04
UNS G10180	3.175E-03	3.087E-03	3.947E-03	2.481E-03
UNS S32550	a.	7.620E-06	a.	a.
UNS NO7718	a.	8.255E-06	a.	a.
UNS S30403	a.	6.350E-06	a.	a.
UNS S31603	a.	8.890E-06	a.	a.
Welded UNS S31603	a.	5.080E-06	a.	a.
UNS R60804	a.	a.	a.	a.

a. Not Reportable/within tolerance of balance error.[6]

Pitting was evident in the aluminum, beryllium, and carbon steel coupons. Figures 3 and 4 are photographs of one carbon steel coupon, before and after cleaning (from the 10-ft (3.05-m)). Likewise, Figures 5 and 6 are photographs of beryllium coupons before and after cleaning (from the 10-ft (3.05-m)). Figures 7 and 8 are photographs of aluminum coupons before and after cleaning (from the 4-ft (1.22 m) depth. Table 4 details the pit depth measurements from 16 coupons with visibly deep-pitted surfaces. Aluminum exposed for one year showed no evidence of pitting and was not measured.

TABLE 3
AVERAGE CORROSION TEST DATA (mm/y)—10 FOOT (3.05 m) BELOW SURFACE

Metal	1-Year	3-Year	6-Year	12-Year
UNS A96061	a.	4.530E-05	1.602E-05	3.111E-05
Beryllium	4.540E-03	7.248E-03	6.633E-03	1.581E-04
UNS G10180	6.350E-03	1.131E-02	8.679E-03	2.481E-03
UNS S32550	1.101E-05	5.715E-06	a.	a.
UNS NO7718	a.	1.079E-05	a.	a.
UNS S30403	a.	8.255E-06	a.	a.
UNS S31603	a.	1.206E-06	a.	a.
Welded UNS S31603	a.	7.620E-06	a.	a.
UNS R60804	a.	5.080E-06	a.	a.

a. Not Reportable/within tolerance of balance error.[6]

Figure 3: Carbon steel coupon before cleaning (front and back views).

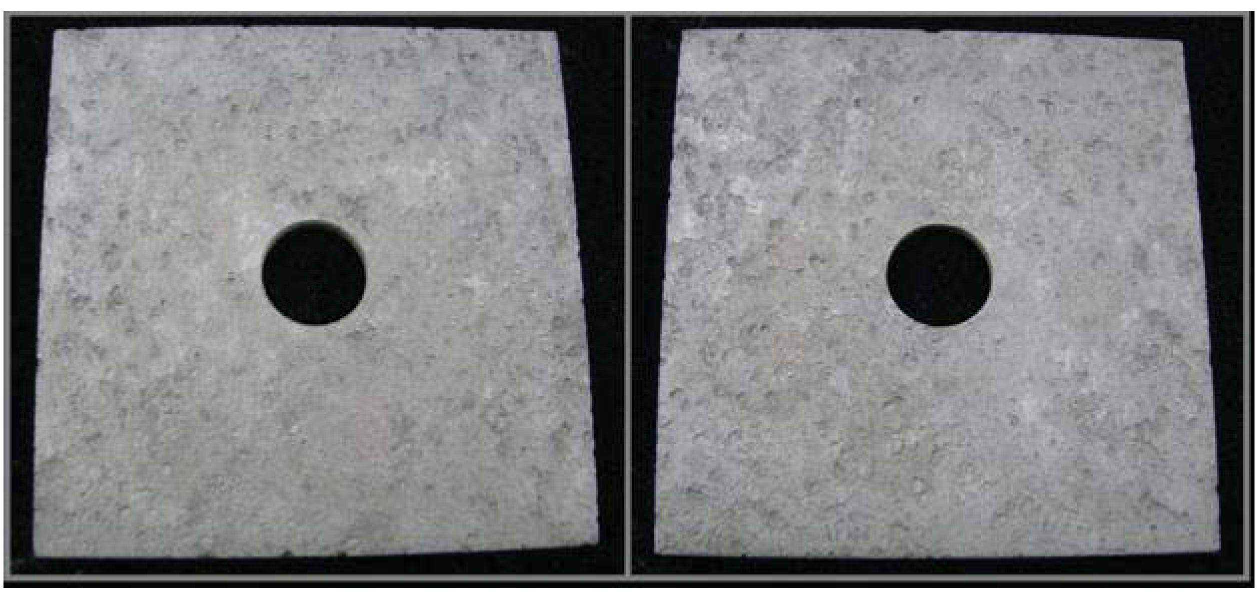

Figure 4: Carbon steel coupon after cleaning (front and back views).

Figure 5: Beryllium coupon before cleaning (front and back views).

Figure 6: Beryllium coupon after cleaning (front and back views).

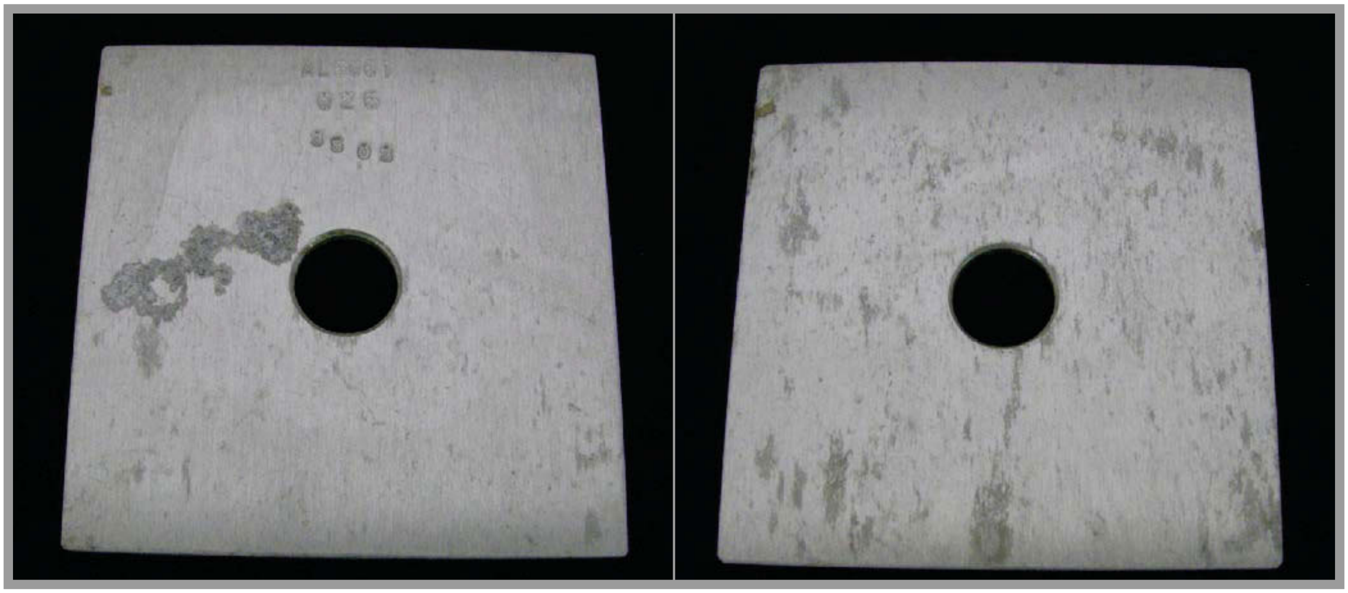

Figure 7: Aluminum coupon before cleaning (front and back views).

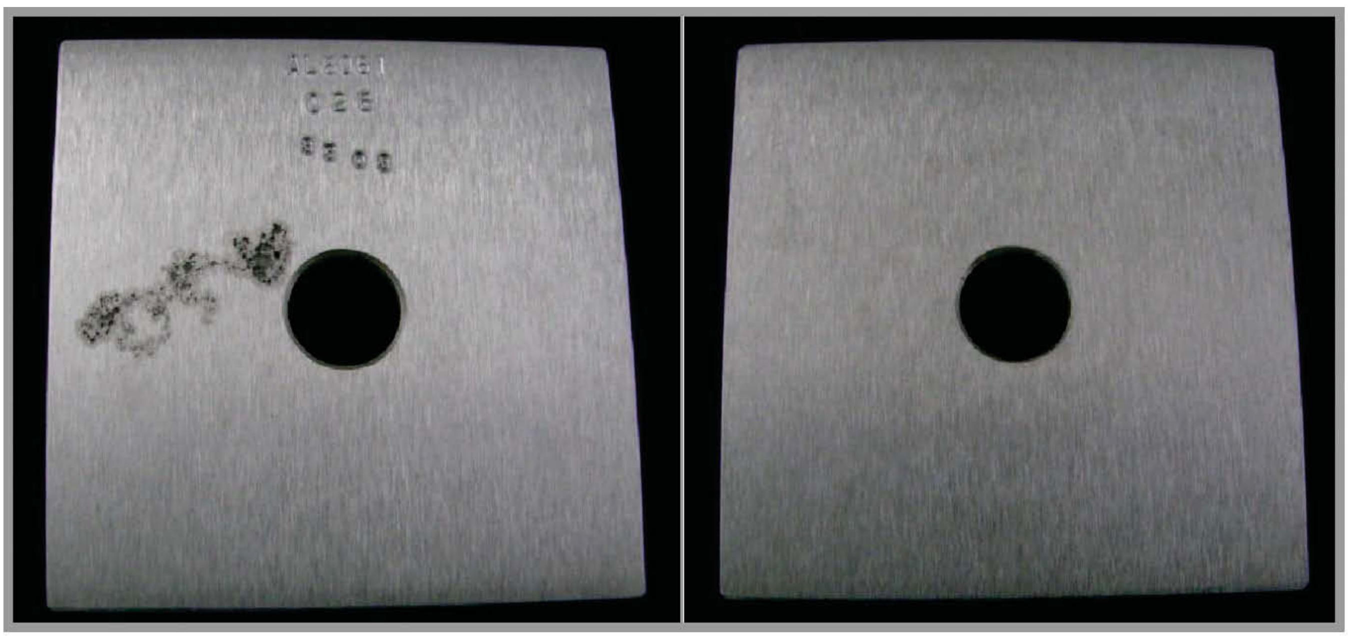

Figure 8: Aluminum coupon after cleaning (front and back views).

TABLE 4
CORROSION TEST DATA—PITTING MEASUREMENTS

Metal	**Exposure Time** (year)	**Test Depth** (ft)	(m)	**Pit Depth** (µm)
UNS A96061	3	4	1.22	365
		10	3.05	275
	6	4	1.22	218
		10	3.05	212
	12	4	1.22	550
		10	3.05	65
Beryllium	1	4	1.22	113
		10	3.05	180
	3	4	1.22	80
		10	3.05	115
	6	4	1.22	108
		10	3.05	189
	12	4	1.22	220
		10	3.05	920
UNS G10180	1	4	1.22	152
		10	3.05	272
	3	4	1.22	99
		10	3.05	204
	6	4	1.22	504
		10	3.05	379
	12	4	1.22	200
		10	3.05	440

CONCLUSIONS

Of the various metals subjected to long-term corrosion testing and evaluated after 1, 3, 6, and 12 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 exhibited low but detectable corrosion rates with the higher corrosion rates being evident on coupons at the lesser depth (4 ft [1.22 m]). The corrosion rates of the austenitic stainless steel (Type 304L and Type 316L), nickel-chromium alloy, and duplex stainless steel were very low but detectable after 3 years but after 6 and 12 years of exposure the rates were not significant. Corrosion rates for the zirconium alloy coupons were very low, below detection limits in most cases.

Pitting caused by corrosion was evident on the carbon steel, beryllium, and aluminum coupons. The contributions of pitting rather than uniform corrosion are of interest when applying the corrosion results to underground structures or containment. In instances where pitting occurs, the 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 as were applied in this study.

Two salient points of interest has emerged from the results. The first is the underground corrosion behavior of beryllium, a metal for which the authors are unaware of any available underground corrosion data except for this study. 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. As a direct result of this ongoing study, performance assessments at the Idaho site are using the beryllium corrosion rate results as part of the radionuclide transport model. The second point is the affirmation of low or not reportable corrosion rate of the stainless steels, nickel chromium alloy and zirconium alloy, indicating the long-lived radionuclides contained in those metal matrixes have a low potential for release. Even though more conservative literature values are currently being used in the performance assessments at the Idaho site for radionuclide transport modeling, continue testing increase confidence in the models.

ACKNOWLEDGEMENTS

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