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Underground Corrosion Of Activated Metals 6-Year Exposure Analysis

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UNDERGROUND CORROSION OF ACTIVATED METALS

6-YEAR EXPOSURE ANALYSIS

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ABSTRACT

The subsurface radioactive disposal site located at the Idaho National Laboratory contains neutron-activated metals from non-fuel nuclear-reactor-core components. A long-term underground corrosion test is being conducted to obtain site-specific corrosion rates to support efforts to more accurately estimate the transfer of activated elements in the surrounding arid vadose zone environment. The test uses nonradioactive metal coupons representing the prominent neutron-activated materials buried at the disposal location, namely, Type 304L stainless steel (UNS S30403), Type 316L stainless steel (S31603), nickel-chromium alloy (UNS NO7718), beryllium, aluminum 6061-T6 (A96061), 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) are also included in the test. This paper briefly describes the ongoing test and presents the results of corrosion analysis from coupons exposed underground for 1, 3, and 6 years.

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

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 site at Idaho National Laboratory. Corrosion rates are based on 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 testing includes parameters known to influence underground metal corrosion. As well as 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 are part of the test program. 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 results, presented here, consist of corrosion rates after 1, 3, and 6 years of exposure from direct corrosion testing.

EXPERIMENTAL PROCEDURE

Direct corrosion testing using buried coupons is the most widely used and simplest method of underground corrosion testing.[1,2,3] 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 (UNS 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 (the 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 × 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 (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] The most recent coupons to be recovered, in 2003, were exposed 6 years to underground corrosion conditions.[6]

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.[7]

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.

RESULTS

Two coupon sets were retrieved 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, 216 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.

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 zirconium alloy coupon after 6-years of exposure and Figure 2 shows the same coupon after cleaning (color differences are an artifact of lighting). 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) — C.5.2 (for the coupons exposed 1 year) and C.1.1 (for the coupons exposed 3 and 6 years).

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.

CONCLUSIONS

Of the various metals subjected to long-term corrosion testing and evaluated after 1, 3, and 6 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 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 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 applied in this study.

Two salient points of interest has emerged from the results thus far. 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. The second 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.

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TABLE 1
MATERIAL CHEMISTRY

Metal	Chemical Composition, Weight %
Aluminum alloy 6061-T6 UNS A96061	Al: BALANCE C: 0.227 Cr: 0.145 Fe: 0.480 Mg: 0.955 Mn: 0.089 Ni: 0.007 Si: 0.644 Ti: 0.021 Zn: 0.048
Carbon steel 1018	Fe: BALANCE Al: 0.054 C: 0.163 Cr: 0.018 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 UNS NO7718	Fe: BALANCE Al: 0.620 B: 0.004 C: 0.040 Co: 0.240 Cr: 18.410 Ni: 52.700 Cu: 0.220 Nb: 5.400 Mn: 0.120 S: 0.002 Mo: 3.150 Si: 0.110 Ta: 0.030 P: 0.011 Ti: 1.120
316L stainless steel UNS S31603	Fe: BALANCE C: 0.010 Co: 0.140 Cr: 16.490 Cu: 0.290 Mn: 1.790 Mo: 2.060 N: 0.034 Ni: 10.170 P: 0.030 S: 0.013 Si: 0.380
Weld Filler	Fe: BALANCE C: 0.010 Cr: 16.490 Co: 0.140 Cu: 0.290 Mn: 1.790 Mo: 2.060 N: 0.034 Ni: 10.170 P: 0.030 S: 0.013 Si: 0.380
304L stainless steel UNS S30403	Fe: BALANCE C: 0.020 Co: 0.100 Cr: 18.230 Cu: 0.390 Mn: 1.760 Mo: 0.400 N: 0.086 Ni: 8.250 P: 0.030 S: 0.016 Si: 0.410
Duplex UNS S32550	Fe: BALANCE C: 0.010 Cr: 25.200 Cu: 1.940 Mn: 1.040 Mo: 3.100 N: 0.210 Ni: 5.880 P: 0.018 S: 0.002 Si: 0.400
Beryllium	Be: 99.000 Al: 0.030 C: 0.050 Fe: 0.100 Si: 0.020 Mg: <0.010
Metal	**Chemical Composition, PPM**
Zirconium alloy UNS R60804	Al: 38 B: 0.25 C: 146 Ca: 10 Cd: <0.25 Cl: 5 Co: <1 P: 8 Cu: 25 Mg: 10 Ta: 100 Fe: 2210 Cr: 1190 O: 1300 H: 7 Hf: 64 Mo: 10 N: 32 Na: 5 Nb: 50 Ni: 35 Pb: 25 Si: 96 Sn: 15400 Ti: 25 U: 1 V: 25 W: 50 Mn: 25

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)	6-YEAR AVERAGES Mass Loss (g)	6-YEAR AVERAGES Corrosion Rate (mm/y)
Aluminum 6061	0.0013	3.895×10^{-5}	0.0218	9.969×10^{-5}	0.0076	3.841×10^{-5}
Beryllium	0.0470	2.007×10^{-3}	0.0919	4.635×10^{-4}	0.0303	2.160×10^{-4}
Carbon Steel	0.3121	3.175×10^{-3}	0.6893	3.087×10^{-3}	2.0296	3.947×10^{-3}
Duplex SS	a.	a.	0.0022	7.620×10^{-6}	0.0018	a.
Nickel-chromium alloy	a.	a.	0.0025	8.255×10^{-6}	0.0016	a.
304 L SS	a.	a.	0.0018	6.350×10^{-6}	0.0015	a.
316L SS	a.	a.	0.0027	8.890×10^{-6}	0.0015	a.
316L SS Welded	a.	a.	0.0014	5.080×10^{-6}	0.0008	a.
Zirconium alloy	a.	a.	a.	a.	-0.0008	a.

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

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)	6-YEAR AVERAGES Mass Loss (g)	6-YEAR AVERAGES Corrosion Rate (mm/y)
Aluminum 6061	a.	a.	0.0051	4.53×10^{-5}	0.0032	1.602×10^{-5}
Beryllium	0.1098	4.540×10^{-3}	0.5072	7.248×10^{-3}	0.9331	6.633×10^{-3}
Carbon Steel	0.6453	6.350×10^{-3}	3.3061	1.131×10^{-2}	5.0539	8.679×10^{-3}
Duplex SS	0.0011	1.101×10^{-5}	0.0018	5.715×10^{-6}	0.0010	a.
Nickel-chromium alloy	a.	a.	0.0036	1.079×10^{-5}	0.0016	a.
304 L SS	a.	a.	0.0025	8.255×10^{-6}	0.0008	a.
316L SS	a.	a.	0.0036	1.206×10^{-5}	0.0012	a.
316L SS Welded	a.	a.	0.0022	7.620×10^{-6}	0.0007	a.
Zirconium alloy	a.	a.	0.0008	5.080×10^{-6}	-0.0004	a.

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

TABLE 4
CORROSION TEST DATA—PITTING MEASUREMENTS

Composition	Exposure Time (year)	Test Depth (ft)	Test Depth (m)	Pit Depth (um)
Carbon Steel	1	4	1.22	152
		10	3.05	272
Beryllium	1	4	1.22	113
		10	3.05	180
Aluminum	3	4	1.22	365
		10	3.05	275
Carbon Steel	3	4	1.22	99
		10	3.05	204
Beryllium	3	4	1.22	80
		10	3.05	115
Aluminum	6	4	1.22	218
		10	3.05	212
Carbon Steel	6	4	1.22	504
		10	3.05	379
Beryllium	6	4	1.22	108
		10	3.05	189

FIGURE 1 – Zirconium Alloy Before Cleaning, 6-year exposure at 1.22 m

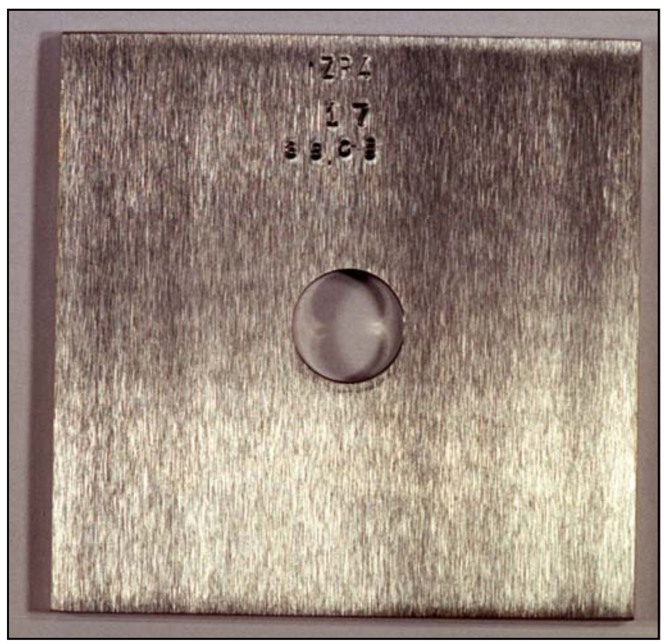

FIGURE 2 – Zirconium Alloy After Cleaning, 6-year exposure at 1.22 m

FIGURE 3 - Carbon Steel Coupon Before Cleaning, 6-year exposure at 3.05-m

FIGURE 4 - Carbon Steel Coupon After Cleaning, 6-year exposure at 3.05-m

FIGURE 5 - Beryllium Coupon Before Cleaning, 6-year exposure at 3.05-m

FIGURE 6 - Beryllium Coupon After Cleaning, 6-year exposure at 3.05-m

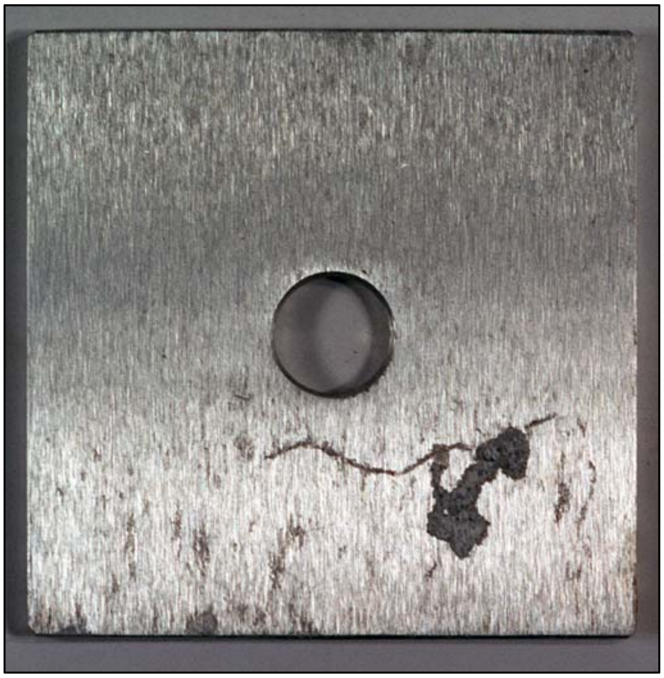

FIGURE 7 - Aluminum Coupon Before Cleaning, 6- year exposure at 3.05-m

FIGURE 8 - Aluminum Coupon After Cleaning, 6-year exposure at 3.05-m

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