

Localized corrosion of Alloy C22 nuclear waste canister material under limiting conditions

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Received 30 November 2005; received in revised form 22 June 2006; accepted 22 June 2006

Abstract

Localized corrosion behavior of Alloy C22 in simulated Yucca Mountain (YM) repository environments was studied at the highest achievable but realistic temperatures under boiling and dripping scenarios. Temperatures measured in concentrated boiling solutions of KCl and NaNO₃ were found to be stable at 140 °C, although transient boiling before dryout was observed at temperatures as high as 160 °C, as the electrolyte became progressively more concentrated. Experiments that simulated a dripping scenario with simulated J13 well water confirmed the existence of concentrated solutions stable at 142 ± 3 °C under controlled drip conditions leading to pit initiation in Alloy C22 after only a few hours. The polarization experiments conducted at 140 °C in a solution with 0.5 mol L⁻¹ chloride concentration showed that the critical potential for localized corrosion was 250 mV (versus Ag/AgCl). Potentiostatic tests confirmed that active metal dissolution occurred only in the crevice region at this potential. The crevice corrosion of Alloy C22 required an incubation time to develop a critical crevice solution, and it was triggered by severe local chemistry (enrichment of Cl⁻ and H⁺) aided by the high temperature.

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Keywords: Waste package; Drip test; Crevice corrosion; Alloy C22

1. Introduction

Alloy C22 is a high nickel base alloy that has been selected for waste package (WP) outer barrier because of its excellent corrosion resistance in various aqueous solutions [1–5]. The waste package is a major component of the engineered barrier system (EBS), and contributes an essential ingredient in the isolation of high level wastes during the pre-closure and post-closure control periods.

A key issue is to determine the maximum temperature at which liquids can exist in the Yucca Mountain (YM) repository setting. Corrosion of Alloy C22 was initially studied at temperatures up to 90 °C in simulated J13 well water with maximum test duration of about 3 years [1,2]. Under these conditions, the general corrosion rate of Alloy C22 was acceptably low, and no evidence of localized corrosion was observed. However, a recent review of the previous corrosion studies recommended

that these simulated tests should be carried out not only at the maximum plausible temperatures, but also at higher temperatures than 90 °C and more aggressive environments to determine the envelop of acceptable conditions for the repository [3].

It was assumed that the maximum water temperature to which the canister is subjected is determined by the deliquescence point of precipitated NaNO₃ [1–5] since NaNO₃ is assumed to be the first species to reach its deliquescence point at a relative humidity of 50%. Based on this assumption, the maximum temperature for aqueous corrosion is limited to 120 °C [1–5]. Although the temperature of the simulated solutions were higher than the critical temperature (~100 °C [6]) for the crevice corrosion of Alloy C22, no localized corrosion attack was reported at 120 °C in the simulated solutions for YM environments since the solutions contained several inhibitors (nitrate and carbonate), which moderate corrosiveness even at this temperature.

Recently, it has been suggested that the temperatures could be as high as 150 °C based on the deliquescence of CaCl₂ to produce brines at 22.5% relative humidity (RH) [4]. In this solution, localized corrosion attacks are evident; however, this temperature is little plausible because the environments at YM are

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Table 1

Chemical compositions of test materials; Alloy C22 (ASTM B-575-97, UNS06022, Hastelloy C22)

Element	Mo	Cr	Fe	W	Co	C	Si	Mn	V	P	S	Ni
Maximum	14.5	22.5	6.0	3.5	2.5	0.015	0.08	0.50	0.35	0.02	0.02	Bal.
Minimum	12.5	20.0	2.0	2.5	0.0	0.000	0.00	0.00	0.00	0.00	0.00	Bal.
Actual (wt.%)	12.8	21.3	4.1	3.2	1.8	0.0010	0.04	0.26	0.10	0.01	<0.01	Bal.

expected to contain a lot of nitrate and carbonate rather than pure CaCl_2 [5].

On the other hand, non-equilibrium wetting conditions may exist under drip or episodic flows that could dissolve the deposited salts on the surface to form concentrated aqueous solutions with much higher boiling points. For example, the infamous discovery of a “new” stable form of water called “polywater” was found in capillary tubes. Polywater was found to boil at 300°C and had a high density and viscosity [7], but was later shown to be merely a concentrated solution consisting of 60% sodium, 15% chloride, and 15% sulfate and 10% water (the composition of concentrated human perspiration that became stable in a fine capillary as may be possible in crevices or pits). Stable boric acid solutions in confined locations were discovered in upper head recesses at the Davis Besse nuclear plant which led to aggressive localized corrosive attack [8,9].

In the present study, an attempt was made to investigate the maximum temperatures possible in concentrated solutions under two scenarios: pool boiling and drip/evaporation conditions. After determining the most plausible maximum temperature, the critical potential for localized corrosion of C22 was measured by polarization and potentiostatic tests at this temperature in an autoclaved solution of simulated J13 + $0.5\text{ mol L}^{-1}\text{ Cl}^-$. Although this test solution was different than the high concentrated solutions from the boiling test and the drip test, the solutions in the crevice during the corrosion tests may represent those solutions in view of solution concentration. Under these conditions both the initiation and propagation of pits and crevice attack were studied.

2. Experimental procedures

2.1. Test material

The Alloy C22 disk-shaped test specimens, 0.78 mm thick \times 48 mm diameter were supplied by Haynes International. The nominal chemical composition and mechanical properties of these coupons are shown in Tables 1 and 2, respectively [6]. The microstructure of these specimens was observed on a polished area which was electrochemically etched in a 10 wt.% Cr_2O_3 solution at 3 V for a few seconds.

Table 2

Mechanical properties of Alloy C22 at RT [6]

Young's modulus (GPa)	206
Y.S. (MPa)	406.8
UTS (MPa)	799.8
Hardness, Rockwell (R_B)	93
Elongation (%)	57

2.2. Boiling point measurement

The major cations in the waters at YM are sodium and potassium, and the major anions are chloride and nitrate [4,5]. In accordance with the definition shown in Table 3 [4], a test solution was prepared by diluting by 10 times the standard simulated saturated water (SSW) [4] at room temperature, by adding 180 g of NaNO_3 and 27 g of KCl to 1000 mL of deionizer water. About 500 mL of this solution was then slowly heated in an open flask while continuously stirring and monitoring the temperature with a type T thermocouple.

2.3. Drip test

The formation and stability of liquids at temperatures above 100°C were examined under experimental conditions that closely simulated drip conditions on a hot canister. In a dripping scenario, the solution temperature on the surface of the waste canister may depend on many parameters such as the solution dripping rate and its regularity, rate of heat dissipation from the canister, composition and quantity of the solution and morphology of the salt precipitate, etc. An Alloy C22 disk sample was placed at the bottom of an alumina crucible and heated to $200 \pm 1^\circ\text{C}$ using a PID controlled hot plate (ECHOtherm™ Model HP30, Torrey Pines Scientific, Inc.). Thermal insulation was used to obtain nearly isothermal conditions and to keep the rate of evaporation low. The drip solution was again the diluted SSW that was heated to 80°C by an UV lamp prior to dripping. The drip rate was controlled by adjusting a valve so as to obtain a stable liquid pool at or near the point of impingement. The temperature of the liquid pool and next to the specimen was measured using sheathed type T thermocouples.

Table 3

Defined composition of J13 well water [4]

Species	Concentration (mg/L)	
	J13 well water	Simulated saturated water (SSW)
Ca^{2+}	13.0 ± 0.99	0
Cl^-	7.14 ± 0.61	128400
F^-	2.18 ± 0.29	0
HCO_3^-	128.9 ± 8.6	0
K^+	5.04 ± 0.61	141600
Mg^{2+}	2.01 ± 0.21	0
NO_3^-	8.78 ± 1.03	1310000
Na^+	45.8 ± 2.29	487000
SO_4^{2-}	18.4 ± 1.03	0
Si (aq)	28.5 ± 1.85	–
pH	7.41	7

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