

REPRESENTATION OF COPPER AND CARBON STEEL WASTE PACKAGE DEGRADATION IN A GENERIC PERFORMANCE ASSESSMENT MODEL

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The U.S. Nuclear Regulatory Commission (NRC) and the Center for Nuclear Waste Regulatory Analyses (CNWRA) jointly developed a generic performance assessment model named the beta-Scoping of Options and Analyzing Risk (β -SOAR), with the intention to provide risk and performance insights for a variety of potential high-level radioactive waste geological disposal options. One key model component considered in β -SOAR is the Waste Package Component model. In this paper, processes of degradation due to corrosion of copper and carbon steel waste packages are discussed as well as their simplified representations in a generic performance assessment model. The corrosion mechanisms considered in the current model include general and localized corrosion. Information on general corrosion rates was input to the model in the form of distribution functions to account for uncertainties in the rates as well as in the environmental setting. Localized corrosion was considered to be either a nonuniform type of general corrosion for copper or a form of pitting corrosion for carbon steel. Using the Waste Package model in β -SOAR, failure times of copper and carbon steel waste packages were calculated for the case of the reducing condition, and calculation results are presented.

I. INTRODUCTION

Copper and carbon steel are being considered candidate materials for waste packages (containers) in high-level waste geological disposal systems in several countries.¹⁻⁵ Metallic waste packages could undergo chemical degradation (i.e., corrosion) when contacted by groundwater. Copper is expected to exhibit either a very slow corrosion rate when exposed to groundwater or, theoretically, to experience no corrosion when in thermodynamic equilibrium in a reducing environment.² Carbon steel is a corrosion-allowance material that is expected to have a relatively low corrosion rate in a reducing environment.² In a potential disposal system, the waste package lifetime could be a key factor in limiting

radionuclide release to the biosphere. Accordingly, an evaluation of waste package material degradation due to corrosion and the impacts on disposal system performance is necessary. Recently, NRC and the CNWRA jointly developed a generic performance assessment model, β -SOAR with the intention to provide risk and performance insights for a variety of potential high-level radioactive waste geological disposal systems.^{6,7} Five key model components are considered in β -SOAR: Waste Form, Waste Package, Near Field, Far Field, and Biosphere. The Waste Package model accounts for chemical degradation due to corrosion of the waste package materials.

This paper discusses how the general and localized corrosion processes of copper and carbon steel waste packages are abstracted and conceptualized to estimate waste package failure times in either oxidizing or reducing environments in a potential geologic disposal system. Using the Waste Package model in β -SOAR, failure times of copper and carbon steel waste packages are calculated and results are presented.

II. CORROSION MODEL ASTRACTION

In the Waste Package model in β -SOAR, two corrosion failure mechanisms are considered: general corrosion and localized corrosion. Other degradation processes [e.g., stress corrosion cracking (SCC), microbially influenced corrosion, hydrogen embrittlement, creep, and material properties/fabrication effect] are not considered in the current model. Potential effects of some of the other degradation processes on long-term waste package degradation are addressed in this paper. General corrosion is modeled to represent progressive failure of waste packages as a function of time. The waste package failure time is calculated as the time at which the corrosion front penetrates the material thickness, using the equation where t_f is the waste package failure time by general corrosion, L is the thickness of waste package, and Corrosion Rate (CR) is the general corrosion rate.

$$t_f = \frac{L}{CR} \quad (1)$$

Information on general corrosion rates obtained from the literature was input to the model in the form of distribution functions to account for uncertainty in the rates and uncertainty in the environmental setting. General corrosion rates are assumed to follow either a log-normal or a log-uniform distribution for copper or carbon steel, respectively. The low and high bounds in the log-normal distribution correspond to the 0.1 and 99.9 percentiles of the corrosion rate distribution, while the low and high bounds in the log-uniform distribution correspond to the minimum and maximum corrosion rates.

In the model, localized corrosion was considered a relatively fast corrosion process compared to general corrosion. Initiation and propagation of the localized corrosion are not explicitly implemented in the model. Instead, a localized corrosion process was considered to be either a non-uniform type of general corrosion (i.e., unevenly distributed corrosion attack on the waste package surface) for copper or to take the form of pitting corrosion for carbon steel based on the knowledge gained from domestic and international geologic disposal research programs. Therefore, degradation by localized corrosion of copper and carbon steel waste packages was represented by an adjustment in the magnitude of general corrosion rates, accomplished by applying an enhancement factor to the base of general corrosion rates.

II.A. Copper Waste Package

For a potential disposal system located in a reducing environment, after saturation with groundwater, the environmental conditions surrounding the waste package (container) are expected to evolve from initially higher temperature and oxidizing conditions to ambient temperature and reducing conditions.^{1,3} As a consequence of feasible changes in the near-field geochemistry with time, several modes of copper corrosion could occur including general corrosion, localized corrosion, microbially influenced corrosion, and SSC.³ In particular, under the initial warm and oxidizing condition, the copper canister may exhibit a relatively high general corrosion rate and localized corrosion.

An oxidizing environment normally has an oxygen concentration of 8 parts per million (5×10^{-4} M), and a reducing environment has 10 parts per billion (6×10^{-7} M).² The initial transient oxidizing condition may last tens to hundreds of years.^{8,9} Long-term corrosion test results of copper coupons in compacted bentonite performed at the Äspö Hard Rock laboratory measured general corrosion rates of copper under oxidizing conditions ranging from 0.5 to 3.5 $\mu\text{m}/\text{year}$ (1.97×10^{-5} to

1.38×10^{-4} inch/year).¹ No signs of active pits were observed after 3 years of tests. However, there was a tendency for more localized corrosion attack. Copper was corroded in a non-uniform type of general corrosion, and the copper surface was roughened. The effect of surface roughness potentially developed in the long term may need to be further considered in the future in determining the distribution type for general corrosion and pitting corrosion rates. Based on a theoretical modeling approach of copper corrosion under oxidizing conditions for up to 300 years for the Swedish disposal setting, the upper bound of the general corrosion rate was selected to be 7 $\mu\text{m}/\text{year}$ (2.76×10^{-4} inch/year) as the conservative case with an enhancement factor of 100 to the reference general corrosion rate.⁹ From these data, the model for the general corrosion rate of copper under oxidizing conditions in β -SOAR is proposed to be 1 to 7 $\mu\text{m}/\text{year}$ (3.94×10^{-5} to 2.76×10^{-4} inch/year), following a normal distribution.

As the initially trapped atmospheric oxygen is consumed, the corrosion rate may become limited by diffusion of oxygen to the waste package surface. After several hundred years, disposal system conditions should become reducing, and the corrosion process of copper will be supported by the reduction of water if sulfide is present in the groundwater.² Therefore, the rate of copper corrosion could be limited by the rate of sulfide diffusion to the waste package surface. Theoretical modeling calculation results to predict the depth of the corrosion front due to sulfide, assuming 1 mg/L (8.34×10^{-6} lb/gal) of sulfide concentration, showed that the average corrosion rates of copper ranged from 5×10^{-5} to 2×10^{-2} $\mu\text{m}/\text{year}$ (1.97×10^{-9} to 7.87×10^{-7} inch/year).^{2,9} The upper bound of 2×10^{-2} $\mu\text{m}/\text{year}$ (7.87×10^{-7} inch/year) was estimated by multiplying the enhancement factor of 5 to the reference rate to account for potential localized corrosion.⁹ The lower bound of 5×10^{-5} $\mu\text{m}/\text{year}$ (1.97×10^{-9} inch/year) determined by modeling, however, appears to be underestimated considering the rates from the corrosion experiments under reducing condition. Taniguchi and Kawasaki¹⁰ conducted a corrosion test for copper. The measured general corrosion rate of a copper coupon surrounded by compacted bentonite was 0.55 $\mu\text{m}/\text{year}$ (2.16×10^{-5} inch/year) after 730 days immersed in simulated seawater containing 0.001 M sulfide {33 mg/L (2.75×10^{-4}) lb/gal} at 80 °C (176 °F). Considering a relatively high concentration of sulfide {33 mg/L (2.75×10^{-4}) lb/gal} in the test, the corrosion rate of 0.55 $\mu\text{m}/\text{year}$ (2.16×10^{-5} inch/year) could be a high bound in the general corrosion rate. For example, the upper limit of sulfide concentration in typical groundwater in Japan was reported to be 0.0003 M {1 mg/L (8.34×10^{-6}) lb/gal}.¹¹ The estimated sulfide concentrations in a potential disposal setting in Sweden are up to 3 mg/L (2.5×10^{-5} lb/gal).³ Therefore,

in β -SOAR, the reference rate of $4 \times 10^{-3} \mu\text{m}/\text{year}$ ($1.57 \times 10^{-7} \text{ inch}/\text{year}$) from the modeling calculation is selected as a lower bound to be conservative.⁹ The upper bound of the general corrosion rate accounting for enhancement due to potential localized corrosion was selected to be $2 \times 10^{-2} \mu\text{m}/\text{year}$ ($7.87 \times 10^{-7} \text{ inch}/\text{year}$). Corrosion rates were assumed to follow a log-normal distribution in β -SOAR based on the data from both theoretical long-term corrosion rates and relatively short-term corrosion experiments.

II.B. Carbon Steel Waste Package

Carbon steel is a non-passive metal in near neutral pH solution. Carbon steel is in a group of corrosion allowance metals used as container metals for geological disposal of high-level nuclear waste. The general corrosion rate of carbon steel in the oxidizing environment is very high: in the range of 10 to 100 $\mu\text{m}/\text{year}$ (3.94×10^{-4} to $3.94 \times 10^{-3} \text{ inch}/\text{year}$) at room temperature in simulated groundwater.^{5,12} Under a 30 year oxidizing condition applicable for carbon steel corrosion,¹³ a rate of 50 $\mu\text{m}/\text{year}$ ($1.97 \times 10^{-3} \text{ inch}/\text{year}$) will result in the penetration depth of 0.15 cm (0.02 inch). The thickness of carbon steel waste packages ranges from 3 to 25 cm (0.12 to 0.98 inch) with an average of 10 cm (0.39 inch) in proposed international disposal programs. The 0.15-cm (0.02-inch) penetration depth is relatively small and will not significantly affect the waste package lifetime. Additionally, carbon steel is known to be susceptible to pitting corrosion in a oxidizing environment.¹³ Therefore, carbon steel is considered as a candidate metal mainly in reducing environments.

The carbon steel corrosion is modeled in β -SOAR as slow general corrosion in the reducing condition with mild groundwater in the host rock. From substantial literature information on carbon steel corrosion,² the two most comprehensive models and data were chosen. The Japanese group conducted carbon steel corrosion tests at 50 and 80 °C (122 to 176 °F) in synthetic groundwater and synthetic sea water, which are both simulated solutions expected in a granite disposal setting.^{8,9} The solution contained $5.6 \times 10^{-1} \text{ M}$ chloride, $2.9 \times 10^{-2} \text{ M}$ sulfate, $2.4 \times 10^{-3} \text{ M}$ bicarbonate, and minor other species and was deaerated by nitrogen gas purging. The tests were conducted for up to 4 years. The data were correlated with archaeological analogue data of the 6th century Yamato Ancient Tomb. Fig. 1 shows a good correlation up to 1,000 years, taking test data from a longer term of 3 to

4 years. The corrosion rates decreased with time, ranging from 4–18 $\mu\text{m}/\text{year}$ (1.57×10^{-4} – $7.09 \times 10^{-4} \text{ inch}/\text{year}$) after 1 year to 0.18–1.9 $\mu\text{m}/\text{year}$ (7.09×10^{-6} – $7.48 \times 10^{-5} \text{ inch}/\text{year}$) after 4 years. From this correlation, the constant corrosion rates used in β -SOAR were estimated to be 0.1 to 10 $\mu\text{m}/\text{year}$ (3.94×10^{-6} to $3.94 \times 10^{-4} \text{ inch}/\text{year}$).

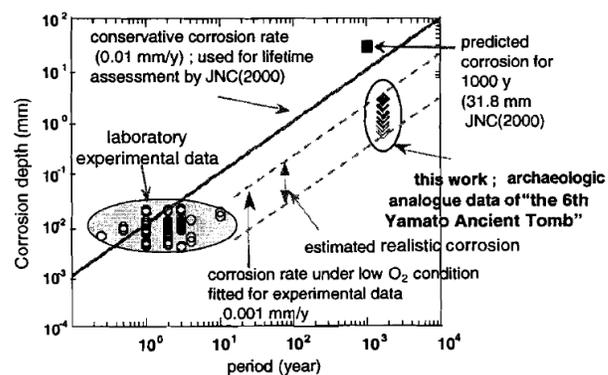


Fig. 1. Comparison of archaeological analogue data with laboratory and predicted corrosion data¹⁴ (used with permission of Elsevier, the Journal of Nuclear Materials, ©2010).

A global estimation of the corrosion rate for archaeological artifacts was made, and it was more recently reviewed by Féron⁵ and David.¹⁵ The results are shown in Fig. 2. Féron and David listed 44 iron samples; however, their origins were different and often origin details were unknown. Despite the variety of artifacts in terms of origin and environmental conditions, most corrosion rates are 0.1 to 10 $\mu\text{m}/\text{year}$ (3.94×10^{-6} to $3.94 \times 10^{-4} \text{ inch}/\text{year}$) except for selected data in seawater. This is closer to the Japanese correlation. From Fig. 2, the data also suggest that the distribution of the corrosion rates is log-uniform, ranging from 0.1 to 10 $\mu\text{m}/\text{year}$ (3.94×10^{-6} to $3.94 \times 10^{-4} \text{ inch}/\text{year}$). Kursten² reviewed corrosion performance of various candidate metals under several different geologic environmental conditions and suggested similar corrosion rates for carbon steel. From these data, the general corrosion rate of carbon steel for β -SOAR is proposed to range from 0.1 to 10 $\mu\text{m}/\text{year}$ (3.94×10^{-6} to $3.94 \times 10^{-4} \text{ inch}/\text{year}$) and follow a uniform distribution.

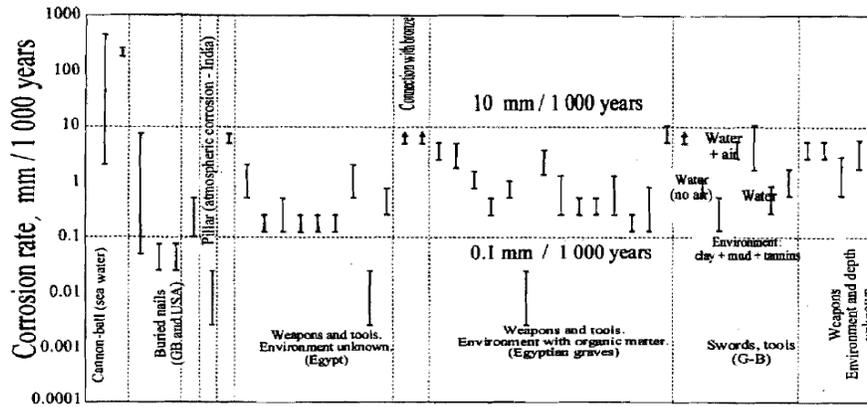


Fig. 2. Corrosion rate data for iron artifacts [David,¹⁵ used with permission of Maney Publishing (www.maney.co.uk), ©2002].

It is generally known that pitting corrosion will occur in carbon steel under the oxidizing condition. This pitting corrosion in carbon steel was widely studied in an empirical approach using a pitting factor.¹³ The pitting factor is defined by the ratio of the penetration depth by pitting to the penetration depth by general corrosion. The pitting factor in longer term, steady-state studies in carbon steel in the literature varies from 1.0 to 3.0 (e.g., Johnson and King¹⁸). As the corrosion proceeds, the pitting factor tends to 1.0 and the pitting converges with general corrosion at less than 1 cm of the average corrosion penetration.¹³ In β -SOAR, a pitting enhancement factor, 1.5, from the pitting factor of 1 to 3 (i.e., a half of a factor 3, 3×0.5) was applied to the general corrosion rates under the oxidizing condition. Pitting corrosion is not obvious under the reducing condition. Therefore, a pitting enhancement factor of 1.0 was applied to the general corrosion rates under the reducing condition.

Féron⁵ presented the temperature dependence of the general corrosion rates from ambient temperature up to 180 °C (356 °F). The apparent activation energy obtained was 11 kJ/mol (2.7 kcal/mol). The groundwater temperature in the disposal system under the reducing condition would likely to be ambient by ~1,000 years in a granite host rock environment.¹⁶ Sometimes, it was postulated to take up to 10,000 years⁵ to return to ambient conditions. The Japanese data in Fig. 1 already considered elevated temperatures in the testing. The corrosion rate increase between the ambient temperature and near the groundwater boiling point is only about a factor of 2 (Ref. 5). Even if pitting and the oxidizing environment affected the disposal system, the resulting penetration would not be significant, relative to an average waste package thickness of 10 cm (0.39 inch).

TABLE I summarizes corrosion rates of copper and carbon steel in oxidizing and reducing conditions reported in the literature.

TABLE I. Corrosion Rates of Copper and Carbon Steel in Oxidizing and Reducing Conditions in the Literature

Material		Corrosion Rate $\mu\text{m}/\text{year}$ (inch/year)	Ref.
Copper	Oxidizing	0.5 – 3.5 (1.97×10^{-5} – 1.38×10^{-5})	1
		0.04 – 7 (1.57×10^{-6} – 2.76×10^{-4})	9
	Reducing	5×10^{-5} – 0.02 (1.97×10^{-9} – 7.87×10^{-7})	1
		0.55 (2.17×10^{-5})	10
Carbon Steel	Oxidizing	10 – 100 (3.94×10^{-4} – 3.94×10^{-3})	5,12
		0.18 – 18 (7.09×10^{-6} – 7.09×10^{-4})	14
	Reducing	0.1 – 10 (3.94×10^{-6} – 3.94×10^{-4})	15

III. MODEL OUTPUT

A preliminary test of β -SOAR was conducted using either a 2.5-cm (0.1-inch)-thick copper or a 10-cm (0.39-inch)-thick carbon steel waste package. The redox condition surrounding the waste package condition was reducing, and a diffusive barrier of backfill was present with a homogeneous, saturated 5-km (3.13-miles) granite far field.^{6,7} TABLE II presents the input values of corrosion rates used. In the calculation, log-normal and log-uniform corrosion rate distributions were used for copper and carbon steel, respectively.

TABLE II. Corrosion Rates Used for Calculation

Material	Lower Bound	Upper Bound
Copper	$4 \times 10^{-3} \mu\text{m/year}$ (1.57×10^{-7} inch/year)	$2 \times 10^{-2} \mu\text{m/year}$ (7.87×10^{-7} inch/year)
Carbon Steel	$0.1 \mu\text{m/year}$ (3.94×10^{-6} inch/year)	$10 \mu\text{m/year}$ (3.94×10^{-4} inch/year)

The waste package model output of carbon steel is shown in Fig. 3.

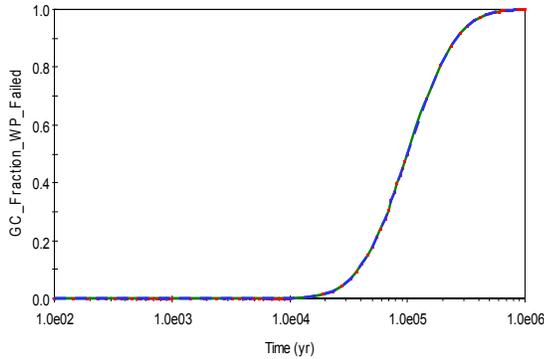


Fig. 3. Carbon steel waste package failure time for geologic disposal system in reducing environment

In the case of copper, the waste package does not fail during the period of up to 1 million years (not shown in the plot). This result is consistent with the calculated lifetimes of copper waste packages in the literature.² The lifetime of the copper waste package was calculated to be longer than 1 million years based on different modeling approaches (e.g., combined mass-balance/mass-transport model, steady-state mass-transport reaction model) from the various international corrosion programs.

On the other hand, relatively high corrosion rates of carbon steel in a reducing environment lead to earlier waste package failure (Fig. 3). The fractions of waste packages failed are 0.1, 0.5, and 1 at 40,000, 60,000, and 1 million years, respectively. In the model, corrosion rates are assigned to be distributed among all waste packages. Therefore, higher corrosion rates can result in earlier waste package failure. Lower corrosion rates contribute to waste package failure at a later time. Eventually, the fraction of waste packages failed increases as a function of time as shown in Fig. 3.

IV. OTHER FACTORS POTENTIALLY INFLUENCING LONG-TERM WASTE PACKAGE DEGRADATION

IV.A. Copper Waste Package

SCC of copper may be a concern in a potential disposal environment. Together with an aggressive near-field water chemistry, the copper waste package may also

be subject to considerable strain as the hydrostatic load develops.³ Several chemical species, such as ammonia, acetate, nitrite, and sulfide, have been known as SCC agents for copper.^{3,10} These species may be present in the disposal environment. Phosphorous as an impurity (up to 140 parts per million) in copper also has a detrimental effect on SCC resistance.³ The decrease of waste package temperature with time could render the copper waste package more susceptible to SCC.¹⁷ More extensive analyses on SCC of copper may be necessary for including or excluding SCC in performance assessments.

In addition to sources of sulfide species present in groundwater or bentonite, a high sulfide concentration could be achieved by a spatial process of sulfate reduction due to microbial action by sulfate-reducing bacteria (SRB). Although microbial action in compacted buffer materials is not expected to occur because SRB can hardly proliferate in the buffer, the possibility of SRB action in groundwater outside the buffer may increase effective sulfide activity in the groundwater.³

The possible effects of gamma radiation on corrosion behavior of copper due to radiolysis of gas or aqueous solutions are considered to be negligible because of the primarily self-shielding nature of the container walls {i.e., 2.5 or 5-cm (0.1 or 0.2-inch)-thick copper shell with cast-iron insert}.^{2,3}

IV.B. Carbon Steel Waste Package

Carbon steel is known to be susceptible to SCC in certain aqueous environments. Caustic and carbonate solutions may cause SCC in carbon steel.¹⁸ However, this condition for SCC also requires cyclic loading. Both caustic and cyclic loading conditions are unlikely to be established in a geologic disposal system. Another possible cause for SCC is hydrogen generation during the corrosion process. Ahn and Soo¹⁹ observed SCC at 80 °C (176 °F) in concentrated groundwater with cathodically charged hydrogen in slow strain testing. More detailed analyses on hydrogen-induced SCC may be necessary for including or excluding SCC in performance assessments.

Literature information indicates that microbially influenced corrosion (MIC) may not have a significant effect on degradation of carbon steel.¹⁸ The extent of MIC due to remotely produced sulfide and organic acids would be minimal. The intrusion of these remotely produced species would not be significant with respect to carbon steel corrosion, because of compacted bentonite backfill, low water activity, initially elevated temperature, and lack of continuous supply of nutrients. Some combined effects of these species with iron ions may need to be further studied.²⁰ Effects of bicarbonates on the general corrosion rates are also minimal.²¹ Although the groundwater may not have high concentrations of chloride, the backfill may induce salt deposits on the waste package surface in the initially high temperature regime. Data show

some increased rates,² which may need to be studied further. Experimental data on carbon steel corrosion at different gamma radiation suggest that below ~ 3 Gy/hour (300 rad/hour), radiolysis has little effect on corrosion rate.²² With exceptions, most disposal system designs would have less strength than this value.²² At 1.3×10^4 Gy/hour (1.3×10^6 rad/hour), the corrosion rate decreased compared with the corrosion rate without radiation.¹⁹ Finally, corrosion rate data show some variation with various weld methods.² This variation may not be significant and deleterious effects may be mitigated by choosing appropriate welding methods.

The extent of the damaged surface area on the waste package is potentially an important factor that could control the release rate of radionuclides from the waste form dissolution inside the failed waste package. There could be two sources of damaged surface area. The first type would be localized corrosion including crevice corrosion and pitting. The damaged surface area in this case may be determined by the density of pits on the open surface or inside the crevice, and the pit size. Recent studies show cathode capacity outside the pit may determine these two values.²³ Although some metals such as stainless steel have been studied,^{24,25} more work may be needed for other metals such as carbon steel. Another type of damaged surface area may come from SCC due to weld residual stress. The weld area and the density of crack number and crack size need to be studied further. Damage to the surface area by SCC is discussed in a paper in this proceeding.²⁶

IV. CONCLUSIONS

A corrosion model considering copper and carbon steel as waste package materials was developed as a part of β -SOAR, a flexible, scoping performance assessment tool to develop risk and performance insights for a wide range of potential geologic disposal systems. Two corrosion failure mechanisms (i.e., general corrosion and localized corrosion) were considered. Technical bases in selecting the range of corrosion rates and distribution types for these waste package materials were discussed. Model output results are consistent with the calculated lifetimes of waste packages in the literature. Other factors that affect waste package degradation were also reviewed to be considered in assessing long-term performance of copper and carbon steel waste packages in geologic disposal settings.

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