MODEL ABSTRACTION OF STAINLESS STEEL WASTE PACKAGE DEGRADATION

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The U.S. Nuclear Regulatory Commission and its contractor, the Center for Nuclear Waste Regulatory Analyses, have been developing a beta (preliminary) version of the performance assessment model Scoping of Options and Analyzing Risk (β-SOAR). This model is intended to provide risk and performance insights for a variety of potential high-level radioactive waste geological disposal concepts. The model includes a representation of the engineered barrier system, which includes submodels for the waste form, waste package, and engineered backfill material. The Waste Package Component model specifically accounts for chemical degradation (i.e., corrosion) of waste package materials, such as copper, stainless steel, carbon steel, and titanium materials, in some geological environmental conditions. Using stainless steel as one of the waste package materials, this paper describes how the general and localized corrosion processes were abstracted and conceptualized to estimate waste package failure times and the extent of damage to the waste package surface for disposal systems located in either an oxidizing or reducing groundwater environment of the hosting geologic disposal system.

I. INTRODUCTION

The U.S. Nuclear Regulatory Commission and its contractor, the Center for Nuclear Waste Regulatory Analyses, have been developing a beta (preliminary) version of the performance assessment model Scoping of Options and Analyzing Risk (β-SOAR). This model is intended to provide risk and performance insights for a variety of potential spent nuclear fuel and high-level radioactive waste geological disposal concepts. The model includes a representation of the engineered barrier system, which includes submodels for the waste form, waste package, and engineered backfill material. The Waste Package Component model specifically accounts for chemical degradation (i.e., corrosion) of waste package materials in possible geological environmental conditions.

Based on the knowledge gained from domestic and international geologic disposal options, copper, carbon steel, stainless steel, and titanium materials were selected as the waste package materials to represent different corrosion behaviors in the β-SOAR scoping analysis. Currently, most countries have selected carbon steel or copper as the base case or candidate waste package materials for the disposal setting in a reducing groundwater environment.[1–2] Carbon steel is a corrosion-allowance material that is expected to have a relatively slow corrosion rate in a reducing environment.[1] Copper is expected to have either a very slow corrosion rate when exposed to aqueous chemical species or theoretically to experience no corrosion when in thermodynamic equilibrium with a reducing environment.[1] A crevice-corrosion-resistant, palladium-containing titanium alloy has been considered as a waste package material in a rocksalt host environment,[1] in a bentonite-filled underground disposal facility in granite,[3] and in the Boom Clay host rock in Belgium.[1] Japan proposed to use Titanium Grade 17 (Ti-0.06Pd) as packaging material for transuranic waste disposal in deep underground drifts surrounded by concrete.[4] Several types of stainless steels have been considered as high-level waste or spent fuel canisters as part of waste package materials in a number of countries.[1] Stainless steel is also used as a canister material for the interim storage of nuclear waste (e.g., United Kingdom, Japan, and United States).

Using stainless steel as one of the waste package materials, this paper describes how the general and localized corrosion processes are abstracted in the β-SOAR and conceptualized to estimate waste package failure times and the extent of damage to the waste package surface for disposal systems located in either oxidizing or reducing groundwater environments of a potential host geologic disposal system.

II. MODEL ABSTRACTION

The Waste Package Component model simulated two waste package failure mechanismsn, representing general corrosion and localized corrosion. General corrosion was modeled to represent progressive failures of waste packages distributed over time. Conversely, localized corrosion was modeled to cause failure of a fraction of the waste packages at discrete times. Other degradation processes (e.g., stress corrosion cracking, microbial-influenced corrosion, hydrogen embrittlement, creep, and susceptibility to corrosion as a function of metal and container fabrication processes) were not explicitly considered in the current model version, but may be considered in later versions.

II.A. General Corrosion

General corrosion is assumed to be a gradual material thinning process proceeding in a relatively slow and uniform manner compared to localized corrosion. The waste package failure time is calculated as the time at which the corrosion front penetrates the material thickness, using the equation

$$t_{gc} = \frac{L}{R_{gc}} \qquad (1)$$

where t_{gc} is the waste package failure time by general corrosion, L is the thickness of waste package material, and R_{gc} is the general corrosion rate.

The distribution of general corrosion rates for the respective waste package materials is an input to the general corrosion model in the Waste Package Component. In a realization, each waste package is considered to exhibit a single corrosion rate. The complete distribution of corrosion rates is covered by the multiple waste packages. Thus, waste packages are computed to fail at different times at given realizations. In the model, general corrosion rates are limited to follow either a normal or uniform distribution or their linear or logarithmic variants. In the case of normal or log-normal distributions, the low and high bounds correspond to the 0.1 and 99.9 percentiles of the corrosion rate distribution. In the case of uniform or log-uniform distributions, the low and high bounds are the minimum and maximum corrosion rates. The effect of surface roughness potentially developed in the long term may need to be considered to determine the distribution type used for general corrosion rates in the β-SOAR.

II.B. Localized Corrosion

Localized corrosion is modeled as a relatively fast degradation process compared to general corrosion, leading to waste package failure in a stepwise manner with time. No explicit models for initiation and propagation of localized corrosion are implemented in the β-SOAR. Instead, failure times are directly sampled from input distributions. Localized corrosion is assumed to cause waste package failure at different, discrete times depending on whether the geologic disposal system is located in a reducing or oxidizing environment.

For a disposal system located in a reducing environment, an initial transient oxidizing period (referred to as Period I) is considered before establishing a stable reducing condition (referred to as Period II). The initial transient oxidizing period is assumed to be much shorter than the stable reducing period because the initial oxygen in the disposal system in a reducing environment is reported to be consumed primarily by the corrosion process in tens to hundreds of years.[5–6] For this type of disposal system, a two-step function defining the fraction of waste packages affected by localized corrosion (f_{lc}) as a function of time is constructed for each realization, as shown in Fig. 1. Two distributions to define the waste package failure time by localized corrosion (t_{lc}) are provided as inputs to the Waste Package Component model, one for Period I and the other for Period II. Other inputs to the Waste Package Component model are the fractions of waste packages affected by localized corrosion during Periods I and II. During Period I, the affected fraction is sampled from a distribution for f_{lc}, which was selected based on the possibility that a waste package will exhibit localized corrosion under the transient oxidizing conditions.

During Period II, it is assumed that the waste packages that did not fail in Period I could exhibit localized corrosion during this period. The fraction of waste packages failed during Period II is sampled from a distribution based on the probability for a waste package to exhibit localized corrosion under reducing conditions. This stepwise failure model allows the user to simulate localized corrosion failures at two discrete times. The total number of waste packages that fail by localized corrosion is the sum of the number of waste packages failed during both Periods I and II, as shown in Fig. 1.

In the case of a disposal system in oxidizing environment, only one failure time distribution for localized corrosion is provided as an input to the Waste Package Component model, along with the corresponding distribution for the fraction of waste packages affected by localized corrosion. With these inputs, a single-step function defining the fraction of waste packages failed by

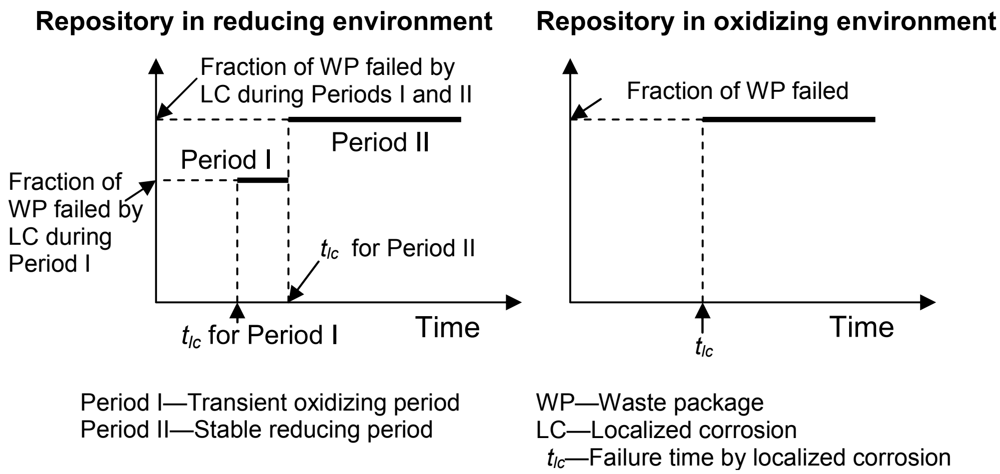


Fig. 1. Step functions used to model localized corrosion per realization

localized corrosion as a function of time is constructed for each realization, as shown in Fig. 1. The fraction of waste packages failed by localized corrosion is sampled from a distribution based on the probability for a waste package to exhibit localized corrosion under oxidizing conditions.

II.C. Waste Package Breached Area

An output of the Waste Package Component model is the total waste package breached area as a function of time. Radionuclide releases into the region surrounding the waste packages are proportional to this breached area. The model considers two distinct breached area fractions: one for localized corrosion and one for general corrosion. For waste package radionuclide release computations, however, a combined breached area per failed waste package is computed at each time step as a function of the general and localized corrosion breached area and the number of waste packages failed. Two different approaches in β-SOAR were implemented to compute the breached area per failed waste package as a function of time. The first is a stepwise approach in which the breached area is a constant after failure by localized corrosion. After the waste package with the highest general corrosion rate (or the 99.9 percentile corrosion rate if corrosion rates follow a normal distribution) fails, then the breached area per waste package steps to the larger number of the breached area due to localized or general corrosion even if other waste packages may not have failed. In this approach, for waste package materials where both localized and general corrosion occur, the breached area per waste package is overestimated because not all waste packages fail at the same time.

In the second approach, the breached area per failed waste package, $WP_{breached\ area}$, is computed as a weighted average

$$WP_{breached\ area} = \left(\frac{f_{gc}}{f_{failed\ WP}} f_{gc\ breached\ area} + \frac{f_{lc}}{f_{failed\ WP}} f_{lc\ breached\ area}\right) A \qquad (2)$$

where f_{gc} is the fraction of waste packages failed by general corrosion, $f_{failed\ WP}$ is the fraction of waste packages failed, $f_{gc\ breached\ area}$ is the general corrosion breached area fraction, $f_{lc\ breached\ area}$ is the localized corrosion breached area fraction, and A is the total area of a waste package.[7] In this equation, if few waste packages are failed due to general corrosion and the majority of the waste packages are failed by localized corrosion, the breached area per failed waste package is closer to the breached area associated with localized corrosion because f_{gc} in Eq. 2 is much smaller than f_{lc}. As more waste packages are failed by general corrosion, the breached area per failed waste package, $WP_{breached\ area}$, approaches the breached area associated with general corrosion, recognizing that $f_{gc\ breached\ area} \sim$ 10-100X $f_{lc\ breached\ area}$.

As stated previously, localized corrosion is modeled to be a faster degradation process than general corrosion. If localized corrosion failure occurs, it usually occurs prior to general corrosion failure. As such, the stepwise approach calculates a breached area that is greater than calculated with the weighted average approach.

III. MODEL INPUT PARAMETERS AND MODEL OUTPUT

For the general corrosion model, the key input parameters are the general corrosion rates for geologic disposal systems in oxidizing or reducing media [R_{gc} in (Eq. 1)], represented as probability distributions to reflect uncertainties in the disposal system environmental conditions. Environmental conditions affecting general corrosion (e.g., temperature, solution pH, and solution chemical composition) are not explicit inputs to calculate general corrosion rates. Nevertheless, their influence is implicitly accounted for by the specified ranges of the corrosion rate distributions (i.e., the lower and upper bounds for the distributions are obtained from literature data representing possible benign and aggressive environmental conditions for a variety of disposal sites). Other general corrosion inputs include the thickness of waste package material (L in Eq. 1) and general corrosion breached area fraction ($f_{gc\ breached\ area}$ in Eq. 2).

For the localized corrosion model, a key parameter is the probability of waste package failure by localized corrosion. The inputs to the localized corrosion model include localized corrosion failure time (t_{lc} in Fig. 1) and localized corrosion breached area fraction ($f_{lc\ breached\ area}$ in Eq. 2).

The general corrosion breached area fraction is assumed to be one, because general corrosion proceeds uniformly across the entire material surface. On the other hand, localized corrosion is assumed to affect a smaller fraction of the waste package surface area. Other inputs to the Waste Package Component model include the waste package surface area (A in Eq. 2), which is used to compute radionuclide releases away from the waste package, and the total number of waste packages, which is used to define the total initial inventory.

The simplified model abstractions and associated parameter inputs used in the Waste Package Component are built upon the knowledge and experience gained from domestic and international performance assessments for a variety of geologic disposal options. Independent analyses and engineering judgment were also used where data were lacking, such as to establish the probability and breach area fraction for localized corrosion of stainless steel. Table 1 shows the parameter values for the β-SOAR Waste Package Component with stainless steel as the representative waste package material.

The Waste Package Component model output for a representative stainless steel waste package is shown in Fig. 2 for an example geologic disposal in oxidizing media and Fig. 3 for an example disposal in reducing media.

Fig. 2(a) shows that waste packages started to fail by general corrosion at 1.7×10^4 years and about 90 percent of the waste packages failed at 1.0×10^6 years, which are consistent with the values calculated based on Eq. 1 using the general corrosion rates and waste package thickness in Table 1. Fig. 2(b) shows that at about 300 years, 40 percent of the waste package failed by localized corrosion with no subsequent failures, which is consistent with the modeling approach in Fig. 1 and the selected parameters in Table 1. Fig. 2(c) shows that at 1.7×10^4 years the breached area stepped to the total surface area of 40 m^2 for each waste package, which is consistent with the conservative approach for general corrosion breached area discussed in Section II.C. Similar consistencies are observed from Fig. 3 and the reducing environment model. In the oxidizing environment (Fig. 2), the stainless steel waste package failed earlier by general corrosion and failed more by localized corrosion than in the reducing environment (Fig. 3). The sensitivity of these parameters on dose can be further understood by running the entire performance assessment model including other components in addition to the Waste Package Component.

IV. MODEL FLEXIBILITIES

The Waste Package Component model is flexible and can be modified to incorporate other waste package materials. For the general corrosion model, the general corrosion rates can be modified to assess performance of other waste package materials. For the localized corrosion model, a key parameter is the probability of waste package failure by localized corrosion, which differs for the four materials considered. For example, the titanium waste package material is assumed to be alloyed with noble metals (e.g., palladium or ruthenium), which effectively reduces the localized corrosion probability of this material.[24] In the current initial version of the model, the localized corrosion probability parameter is simply set to zero for this material. For copper and carbon steel, localized corrosion in the form of pitting corrosion is considered possible. The localized corrosion probability parameter, however, is also set to zero because localized corrosion of these materials is not explicitly modeled with the localized corrosion model. Instead, localized corrosion is indirectly modeled by enhancing the general corrosion rates provided as input, as pitting corrosion of these materials is more widespread.[24]

In addition to the default parameterization, the Waste Package Component model has several flexibilities to consider alternative geologic disposal system configurations. These corrosion rates can be modified to constrain uncertainty where environmental conditions in the disposal system are better characterized or to analyze different environmental regimes (e.g., increasing the rates due to higher temperatures). The distributed and stepwise

TABLE I. Parameters and values for β-s*coping of options and analyzing risk* waste package component model with stainless steel as the representative waste package material

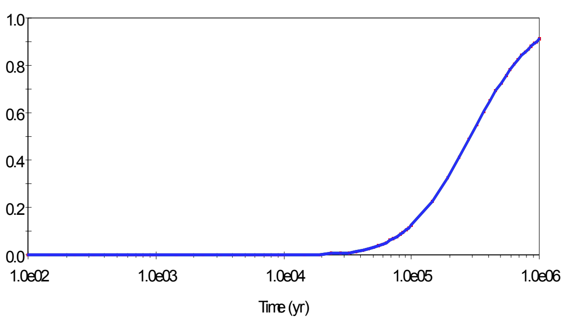
Description	Distribution Type & Values	Reference/Rationale
Stainless steel waste package thickness (cm)	Constant 5	Based on the inner container design of the proposed Yucca Mountain repository[7] because of lack of available data from other countries**.**
Total surface area of one waste package (m^2)	Constant 40	Based on waste package designs for the proposed Yucca Mountain Project[7]
Low bound for the stainless steel general corrosion rate in oxidizing medium (µm/yr)	Discrete 0.01	Based on stainless steel minimum corrosion rate for the proposed Yucca Mountain Project under in-package condition.[7]
High bound for the stainless steel general corrosion rate in oxidizing medium (µm/yr)	Discrete 3	Based on corrosion studies at pH of 1.9–13 and temperatures of 30–80 °C in cementitious backfill material reported in Ref. 1.[1] The corrosion rate increased with decreasing pH. At pH of 4 and 80 °C, the corrosion rate was 2.6 µm/yr. The data reported in the literature[8–10] are mostly distributed at the lower side, which supports a log-normal distribution between low bound and high bound of corrosion rates.
Low bound for the stainless steel general corrosion rate in reducing medium (µm/yr)	Discrete 0.003	Based on data reported by Kursten.[1] This value is also near the resolution limit of the weight loss analysis method to determine corrosion rate.
High bound for the stainless steel general corrosion rate in reducing medium (µm/yr)	Discrete 0.1	Based on *in-situ* corrosion studies relevant to the Belgian disposal concept in clay in anoxic condition, Kursten[1] reported that the maximum corrosion rate was 0.15 µm/yr at 170 °C with most of the corrosion rates less than 0.1 µm/yr. The limited data reported by Kursten[1] in anoxic condition also support a log-normal distribution between low bound and high bound of corrosion rates.
Failure time by localized corrosion for the early Period I in Fig. 1 (yr)	Log-uniform 30, 280	Period I in Fig. 1 is used to define an early transient oxidizing period over which the waste package may experience localized corrosion. Taniguchi[5] reported that for a geological disposal system design for high-level radioactive waste in Japan, the period until the environmental condition returns from oxidizing to reducing is expected to be less than 100 years. Wersin[6] reported a maximum period of 280 years for the oxidizing condition. Foct and Gras (Fig. 6)[11] reported the oxidizing period begins from 30 years. Based on these data, it is assumed that localized corrosion could initiate during the oxidizing period, up to 280 years.
Failure time by localized corrosion for the late Period II in Fig. 1 (yr)	Log-uniform 280, upper end of simulation period	Period II is used to define a late period over which the waste package may experience localized corrosion after the return of a disposal to reducing conditions.

TABLE I. Parameters and values for β-SOAR waste package component model with stainless steel as the representative waste package material (continued)

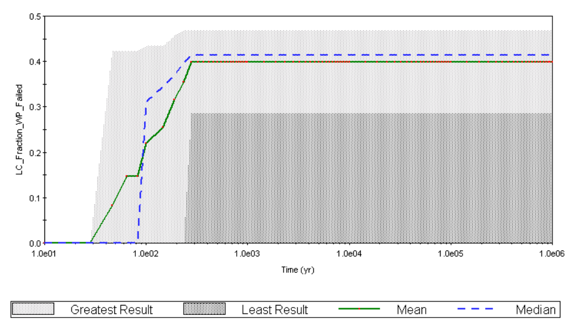
Description	Distribution Type & Values	Reference/Rationale
Fraction of stainless steel waste packages failed by localized corrosion during the early Period I in Fig. 1 in reducing environment (unitless)	Triangular 0, 0.125, 0.25	This defines the probability of stainless steel localized corrosion during Period I in Fig. 1 in a disposal system in reducing media. Based on literature information, this probability strongly depends on Cl^- concentration, pH, and temperature. Under various disposal system-relevant pH and temperature, the threshold Cl^- concentration for localized corrosion is reported to be in the range of 1,000-10,000 ppm (1,000 ppm for crevice corrosion and 10,000 ppm for pitting corrosion).[12–15, 9, 16–20] According to the Cl^- concentration range in potential disposal environments summarized in Ref. 1[1] Table 1, it appears that the probability to have such Cl^- concentration is about 50 percent. The backfill diffusive barrier could limit the Cl^- concentration in contact with stainless steel. Assuming 50 percent chance for backfill to limit water contact and considering the 50 percent chance of susceptible Cl^- concentration, the probability for stainless steel localized corrosion to occur is 25 percent. A triangular distribution is used to have the most likely value in the middle.
Fraction of stainless steel waste packages failed by localized corrosion during the late Period II in reducing environment (unitless)	Uniform 0.01, 0.1	This defines the probability of stainless steel localized corrosion during Period II in Fig. 1 in a disposal system in reducing media. Stainless steel localized corrosion data under anoxic conditions are very limited. Kursten[1] (Fig. 4-38) shows that crevice corrosion may occur even at Cl^- concentration of 10,000 ppm, but pitting corrosion will not occur until Cl^- concentration is greater than 50,000 ppm at 140 °C. According to the Cl^- concentration range in potential disposal environments summarized in Ref. 1[1] Table 1, it appears that the probability to have such Cl^- concentration is less than 10 percent. During the later period, backfill may be less effective to limit water contact, so no credit is taken for backfill to protect the waste package against localized corrosion. Therefore, the probability of stainless steel localized corrosion during Period II is selected to be 10 percent assuming a uniform distribution.
Fraction of stainless steel waste packages failed by localized corrosion in oxidizing environment (unitless)	Triangular 0.25, 0.45, 0.5	This defines the probability of stainless steel localized corrosion in a disposal system in oxidizing media where oxidizing conditions prevail in the short and long terms. Based on the arguments for the fraction of stainless steel waste packages failed by localized corrosion during the early oxidizing Period I in Fig. 1 in the reducing environment, 50 percent is considered to be the highest probability for localized corrosion to occur without taking any credit for backfill to protect the waste package against localized corrosion. By using a 50 percent chance of backfill to limit water contact, 25 percent is considered to be the lowest probability. A triangular distribution is selected.

TABLE I. Parameters and values for β-SOAR waste package component model with stainless steel as the representative waste package material (continued)

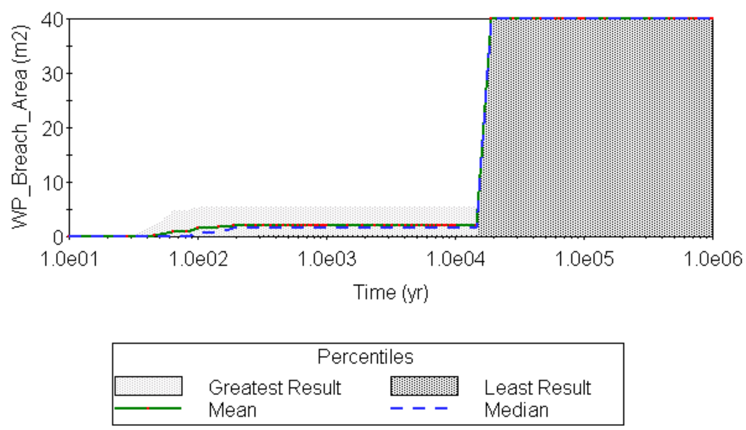
Description	Distribution Type & Values	Reference/Rationale
Fraction of waste package area breached by general corrosion (unitless)	Discrete 1	Because general corrosion is usually widespread, the percentage of the area damaged by general corrosion is considered to be 100 percent.
Fraction of waste package area breached by localized corrosion (unitless)	Log-triangular 0.001, 0.1216, 0.2	According to literature information on stainless steel pitting corrosion in chloride solution, the maximum breach area for the stainless steel waste packages does not exceed 5 percent of the total area.[21–23] Considering that the waste package may be in contact with buffer material to form a crevice and the buffer material may degrade with time to form fractures, stainless steel crevice corrosion is also considered as a possible localized corrosion form, especially for a disposal system in oxidizing media. To support metal dissolution reactions on anodic sites, it is conservatively estimated that the cathodic area needs to be at least five times that of anodic sites (corrosion sites) because both oxygen and water reduction reactions occur at much slower rates compared to metal dissolution reactions at anodic sites. Considering this, an upper bound of 20 percent is considered.



(a) Fraction of waste packages failed by general corrosion

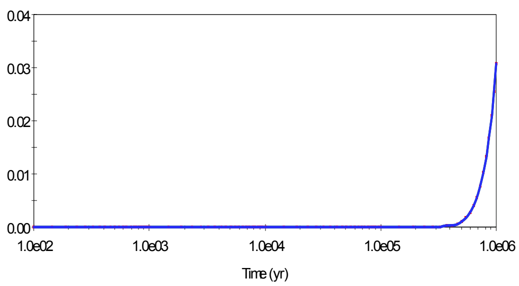


(b) Fraction of waste packages failed by localized corrosion

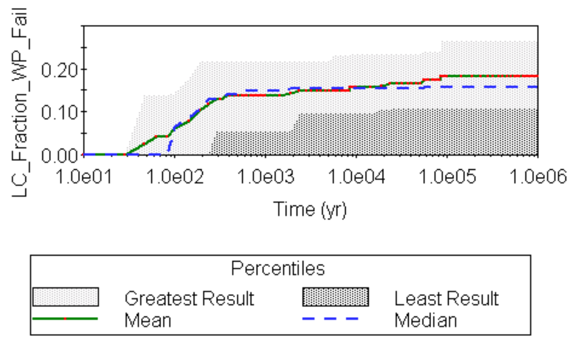


(c) Waste package breached area

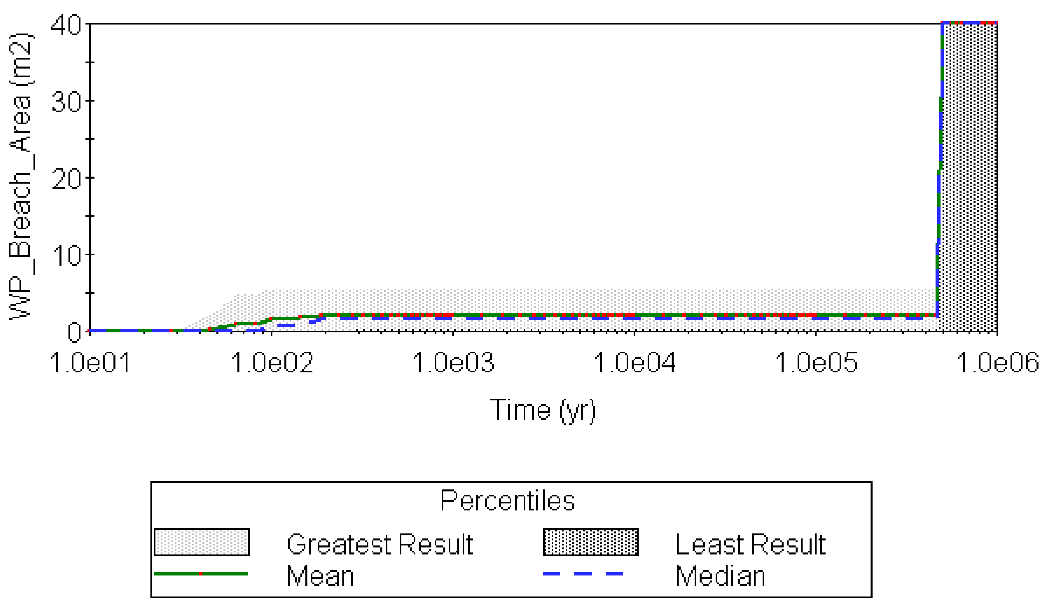
Fig. 2. Stainless steel waste package failure times and extent of damage to the waste package surface in the example case for a geologic disposal system in an oxidizing environment: (a) fraction of waste packages failed by general corrosion, (b) fraction of waste packages failed by localized corrosion, and (c) waste package breached area.



(a) Fraction of waste packages failed by general corrosion



(b) Fraction of waste packages failed by localized corrosion



(c) Waste package breached area

Fig. 3. Stainless steel waste package failure times and extent of damage to the waste package surface in the example case for a geologic disposal system in a reducing environment: (a) fraction of waste packages failed by general corrosion, (b) fraction of waste packages failed by localized corrosion, and (c) waste package breached area.

failure can be modified to approximate additional failure mechanisms beyond general and localized corrosion. Given that any waste package failure mechanism can be characterized by two quantities, i.e., the failure time and the extent of damage to the waste package surface, the default input parameters can be adjusted to evaluate consequences of other failure mechanisms. For example, consequences of stress corrosion cracking could be evaluated, provided technical data were available to define the extent of waste package surface damage and other input parameters selected to cause failure in expected time frames. Similarly, the stepwise failure time, probability, and breach area fraction can be modified to approximate discrete events such as early failure, human intrusion, or other disruptive events. However, because the β-*SOAR* is an abstracted scoping tool with limited consideration of coupling among processes leading to waste package failure and radionuclide release to the environment, the results should be carefully interpreted.

I. CONCLUSIONS

A Waste Package Component model using stainless steel as a one of the waste package materials was developed in the beta (preliminary) version of the performance assessment model β-SOAR. The general and localized corrosion processes of stainless steel were abstracted and conceptualized to estimate waste package failure times and the extent of damage to the waste package surface for disposal systems located in either oxidizing or reducing environments.

General corrosion was assumed to be a relatively slow and uniform process resulting in waste package thinning. Localized corrosion, on the other hand, was modeled as a relatively fast degradation process compared to general corrosion, leading to waste package failure at different times depending on whether the geologic disposal system is located in a reducing or oxidizing environment. A simplified description for the extent of damage to the waste package surface (i.e., breached area) was adopted in the model. Localized corrosion was assumed to affect a smaller fraction of the waste package surface area than general corrosion. Damage due to general corrosion was assumed not to significantly restrict radionuclide releases away from the waste package.

The simplified model abstractions and associated parameter inputs were built upon the knowledge and experience gained from domestic and international performance assessments for a variety of geologic disposal options. This model is flexible to account for chemical degradation (i.e., corrosion) of various waste package materials, such as stainless steel, copper, carbon steel, and titanium materials, in some environmental conditions.

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