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CLADDING DEGRADATION COMPONENT IN WASTE FORM DEGRADATION MODEL IN TSPA-SR

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INTRODUCTION

The U.S. Department of Energy (DOE) has prepared a total system performance assessment for a site recommendation (TSPA-SR), if suitable, on Yucca Mountain for disposal of radioactive waste.¹ Discussed here is the Cladding Degradation Component of the Waste Form Degradation Model (WF Model), of the TSPA-SR. The Cladding Degradation Component determines the degradation rate of the Zircaloy cladding on commercial spent nuclear fuel (CSNF) and, thereby, the CSNF matrix exposed and radioisotopes available for dissolution in any water present.^{2,3,4}

Since the 1950s, most CSNF has been clad with less than 1 mm (usually between 600 and 900 μm) of Zircaloy, a zirconium alloy. Zircaloy cladding is not a designed engineered barrier of the Yucca Mountain disposal system, but rather is an existing characteristic of the CSNF that is important to determining the release rate of radioisotopes once the waste package (WP) has breached. Although studies of cladding degradation from fluoride [F] began at Lawrence Livermore National Laboratory as early as 1984,⁵ cladding as a characteristic of the waste was not considered in TSPAs, conducted in the early 1990s. However, enough information on cladding performance has accumulated in the literature such that cladding was considered in 1993 when examining the performance of DOE spent nuclear fuel^{2,6} (DSNF) and most recently in TSPA for the viability assessment^{2,7} (TSPA-VA). The Nuclear Regulatory Commission (NRC) currently uses cladding data as the basis for extending the period of wet storage, for licensing dry storage facilities, and for licensing shipping casks for CSNF.

PERFORATION AND UNZIPPING OF CLADDING

For TSPA-SR, two steps for cladding degradation were included: perforation and unzipping⁸ (axial splitting) (Figure 1). The perforation of the cladding, through the initiation of small cracks or holes, can occur because the cladding (1) is damaged in the reactor (initial perforations),

(2) creeps and ruptures or cracks from stress corrosion during dry storage, transportation, or initial disposal in the repository, (3) corrodes locally when highly acidic environments develop along with the presence of iron and chlorides or fluorides, and (4) is damaged from rockfall or shaking motion from earthquakes. Once perforated, the cladding may unzip⁸ as the underlying UO_2 fuel matrix oxidizes further to uranium minerals that take up more volume (e.g., the uranium hydroxide, schoepite).^{3,4}

Other mechanisms of initiating cladding perforations have been examined (e.g., hydride failures, hydride embrittlement of cladding, delayed hydride cracking, water-logged rods, general corrosion of cladding, microbial corrosion of cladding, acid corrosion from radiolysis, and enhanced cladding corrosion from high, dissolved-silica content of waters, or diffusion-controlled cavity growth). These mechanisms, however, were omitted because of their low probability of occurrence or inability to substantially increase cladding degradation beyond those mechanisms considered.⁶

INITIALLY PERFORATED CLADDING

Zircaloy cladding perforations may occur in the reactor (including fuel handling operations), in pool storage or dry storage (including fuel handling), and in transportation from storage. To estimate this percentage of initially perforated clad, Siegmann⁴ defined a triangular distribution, with a minimum of 0.0155%, a mode of 0.0948%, and maximum of 1.285%. This distribution is based on data from 65,000 boiling water reactor (BWR) assemblies, with about 4 million fuel rods, and 47,000 pressurized water reactor (PWR) assemblies, with about 10 million fuel rods⁴ and includes projected burnup of the fuel to 75 GWD/MTU (gigawatt day per metric tonne of uranium). This distribution is sampled once for each TSPA simulation.

A small number of the CSNF packages will contain stainless steel clad fuel. All the stainless steel clad fuel is conservatively assumed to be perforated. In the

TSPA-VA,⁷ the 1.1% by weight of stainless steel clad fuel was assumed to be uniformly placed in each and every CSNF package, which resulted in only a portion of a rod being placed in an individual WP. For the TSPA-SR, a more realistic loading arrangement was used. About 3.5% of the containers were assumed to be 33% filled with stainless steel clad fuel. However, because the number of WPs containing stainless steel cladding is small, these packages are not distributed evenly throughout the repository. Rather, they are assigned to the percolation zone with the most WPs and thus covering the largest area of the repository.^a For the low percolation case (current climate conditions) the largest zone is the first bin (<3 mm/yr percolation). For the mean and high percolation cases, this is the fourth bin (40-60 mm/yr percolation). Furthermore, the stainless steel clad fuel is assigned to the "always dripping" condition; consequently, they contribute both to advective and diffusive releases.

PERFORATION FROM CREEP AND SCC

To evaluate perforations resulting from creep rupture and stress corrosion cracking (SCC), the initial stress state of the fuel rods (i.e., the hoop stress from internal gas pressure) was conservatively estimated by assuming that all Zircaloy cladding was placed in dry storage for 20 yr where temperatures slowly decreased from an initial temperature of 350°C, then placed in a shipping cask for three weeks, where it again reached a maximum temperature of 350°C. From the statistical distribution for the initial internal pressures, temperature history during transportation and storage, the perforations from creep rupture and SCC were calculated as a function of peak temperature during disposal (Figure 2a). At a peak WP surface temperature greater than about 300°C, the fraction of rods perforated from creep rupture and SCC increases dramatically. The function developed is very conservative, based on a comparison with creep rupture failure observed in irradiated cladding (which experiences strain hardening) (Figure 2b). The failure-strain criterion for creep rupture was based on tests of unirradiated cladding (i.e., Murry criterion⁹).⁴ By varying the strain threshold between 0.4% and 11.8% at which rupture is assumed to occur an uncertainty in the creep rupture function is declined. SCC of Zircaloy requires an aggressive chemical environment and high stress. Zircaloy is not susceptible to SCC in NaCl, HCl, MgCl₂, and H₂S solutions, but is in iodine solutions, when the iodine area density concentration in the fuel-cladding gap is greater than 5 mg/cm². The free iodine concentrations is expected to be small in CSNF. However, for the TSPA-SR, it was conservatively assumed that a sufficient amount of iodine was present on the

cladding interior, the stress was high, and the duration of elevated temperatures was sufficiently long so that once cracking started, there was sufficient time to propagate through the cladding.

Estimation of the peak cladding temperature after disposal is based on a heat transfer model expressed as a function of WP surface temperature. As implemented in the TSPA-SR, the peak WP temperature that occurs during disposal in each percolation zone is used to select minimum, mode, and maximum of a triangular distribution on the creep rupture and SCC function (Figure 2a). The Cladding Degradation Component takes samples from this triangular distribution to determine the fraction of rods perforated by creep rupture and SCC.

PERFORATION FROM LOCALIZED CORROSION

Most known forms of localized corrosion of the cladding could be screened out based on the calculated *macroscopic*, in-package chemistry as calculated by the In-Package Chemistry Component.^{2,10} For example, the fluoride concentration present in Yucca Mountain groundwater (2.2 mg/L in the J-13 well)¹¹ is too low by several orders of magnitude to corrode Zircaloy. However, localized corrosion was still considered as a perforation mechanism, albeit generically, to account for uncertainty in the *microscopic*, in-package chemistry. The localized corrosion was assumed to be linearly dependent upon the cumulative amount of water^b (with anions such as F) that had entered a WP. On average, when 2,424 m³ of water flows through a WP, 100% of the rods are assumed perforated by localized corrosion.⁴ The uncertainty about the average was defined as a log-uniform distribution ranging by a factor of 0.1 to 10.

PERFORATION FROM EARTHQUAKES

Although, perforation of the Zircaloy cladding may also arise from mechanical and seismic loads, rockfalls on backfill supported by intact drip shields and waste packages, and even most earthquakes, would not perforate the cladding. Only severe seismic events, with a frequency of 10⁻⁶/yr, could potentially perforate the cladding;⁴ therefore, a disruptive event with this frequency is considered to cause the perforation of all cladding in TSPA-SR. Future iterations of TSPA-SR will incorporate mechanical perforation from rockfalls at late-times when the drip shield and WP have failed to better evaluate peak doses beyond 10⁵ yr.

^a WPs within the repository were grouped into five percolation zones ("bins") (<3mm/yr, 3-10 mm/yr, 10-40 mm/yr, 40-60 mm/yr and >60 mm/yr) and three drip conditions (always, intermittent, and never).

^b The water flow depends on the location of the WP because of different drip rates in different percolation zones of the repository. The water flow into the WP increases with time as additional patches open on the WP.

CLADDING UNZIPPING

Unzipping of the cladding occurs⁸ after perforation. The release occurs in two stages: fast release and unzipping (Figure 1). Fast release refers to the inventory of radioisotopes that are in the gap between the fuel pellets and the cladding of the CSNF matrix¹² plus the inventory in the matrix at the perforation for a specified volume.

In the second stage of release, unzipping is assumed to occur as dissolved UO_2 precipitates locally as schoepite. The Cladding Degradation Component models the unzipping rate/per surface area as equal to the degradation rate of the CSNF evaluated in the CSNF Matrix Degradation Component^{2,3} times the active surface area sampled from a triangular distribution with a mode of 40 and range of 1 to 240. Because the CSNF degradation rate varies with pH, total carbonate concentration, and temperature, the unzipping velocity and fraction of fuel exposed are evaluated at each time-step in TSPA-SR. It is conservatively assumed that a perforation occurs in the center of a fuel rod and propagates in both directions to the ends of a 3.66-m rod.

RESULTS

Until several tens of thousands of years after the WP begins to fail, the only cladding perforated is either that perforated by rare seismic events of that which arrives at the repository perforated from reactor operation, transportation, or because of creep rupture or SCC during dry storage. The mean perforation at 200°C is respectively 0.0045 and 0.0765 for initial perforation and creep rupture for percolation zone 4 (40 to 60 mm/yr) (Figure 2a).

Thus, on average about 8% of the cladding will be calculated to perforate because of creep rupture or SCC in TSPA-SR. This mean percentage is very conservative and above the amount of creep rupture and SCC that the NRC will tolerate of operators of dry storage facilities.^c

For the two most important radioisotopes in the groundwater release scenario, ^{99}Tc and ^{237}Np , the CSNF represents ~90% and ~86% of the total inventory, respectively.^{7,13} Hence, presence of intact cladding directly influences the dose by reducing the release rate. The cladding model had five parameters that were sampled for TSPA-SR: (1) the number of rods initially perforated in a CSNF waste package (f_{rod}), (2) the fraction of cladding perforated because of creep rupture and stress corrosion cracking (Figure 2), (3) the uncertainty in localized corrosion rate, (4) the uncertainty of the CSNF degradation rate per surface area, and (5) the uncertainty in the shape

of the active surface area. An estimate of the uncertainty that the cladding model causes in the dose was evaluated by setting all sample parameters except the fraction of cladding perforated by creep and SCC at the 5% and 95% values and observing the change in mean dose (Figure 4). Because cladding perforation is predominately from creep rupture (from dry storage) during the first 40,000 years, the dose is not strongly affected by changing other parameters that influence degradation of the cladding (i.e., the WP surface temperature of the WPs in the repository rarely exceeds 200°C, thus the creep rupture and SCC that occur are during dry storage and transportation. The mean dose increases a factor of ~1.5 when the four parameter values are set at the 95% percentile and decreases a factor 4 when the four parameters are set at the 5% percentile in the first 10^5 yr. Uncertainty in the unzipping of the cladding, between 1 and 240 times the degradation rate of the CSNF matrix,⁴ is an important source of the uncertainty in the dose results after 10^5 yr but not prior to 10^5 yr.

Only after 50,000 years, does the perforated fraction of cladding change due to localized corrosion and then only in those WP that have seepage (Figure 3a). The localized corrosion is a direct function of the seepage volume into the WP; because the intermittent drip case has a greater mean seepage volume, it has slightly more localized corrosion and greater perforation.

Thus, localized corrosion is important only after $\sim 10^5$ yr. (In the TSPA-VA, perforation of the cladding from rock fall was the primary mechanism to damage cladding beyond 10^5 yr.)

Prior to 10^5 yr, the velocity (expressed as velocity divided by rod length) varies between 4×10^{-6} /yr to 2×10^{-3} /yr (Figure 3b). For comparison, the CSNF matrix degradation rate in TSPA-VA⁵ varied between 4×10^{-5} /yr and 2×10^{-4} /yr.

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^c In future iterations of TSPA-SR, perforations from creep and SCC will more closely match NTC criteria for licensed dry storage facilities.

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Figure 1. Cladding Degradation Component models perforation and unzipping as two steps³

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Figure 2. Perforation due to creep rupture and stress corrosion cracking (a) relationship based in TSPA-SR, and (b) comparison of Murty creep correlation (based on unirradiated cladding) with measurements of irradiated cladding

(a)

(b)

Figure 3. Perforation and unzipping rate for cladding in percolation zone 4 (40-60 mm/yr) (a) Mean fraction perforated for all three drip conditions,¹ and (b) unzipping rate for Always Drip condition¹

Figure 4. Sensitivity of mean dose rates to degraded and enhanced cladding¹