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.\(^2\)

Since the 1950s, most CSNF has been clad with less than 1 mm (usually between 600 and 900 \(\mu\)m) of Zircaloy, a zirconium alloy. Zircaloy cladding is not a designed engineered barrier of the Yucca Mountain disposal system, but rather 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,\(^3\) 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\(^4\) (DSNF) and most recently in TSPA for the viability assessment\(^5,6\) (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 CNSF.

PERFORATION AND UNZIPPING OF CLADDING

For TSPA-SR, two steps for cladding degradation were included: perforation and unzipping\(^7\) (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 unzip\(^8\) as the underlying UO\(_2\) fuel matrix oxidizes further to uranium minerals that take up more volume (e.g., the uranium hydroxide, schoepite).\(^9\)

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, Siegmann\(^4\) 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 rods\(^1\) 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, 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. 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
criterion). 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
rupture started, there was sufficient time to propagate
through the cladding.

Estimation of the peak cladding temperature after
percolation 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
percolation in each percolation zone is used to select
minimum, mode, and maximum of a triangular distribution
of the repository. 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 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;
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 10³ yr.

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a WPs within the repository were grouped into five percolation zones
("bins") (<3 mm/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 occurs 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 matrix 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 UO₂ 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 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 respectively. Hence, presence of intact cladding directly affects 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 (f₀ₐₙ), (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 in 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 10⁵ yr. Uncertainty in the unzipping of the cladding, between 1 and 240 times the degradation rate of the CSNF matrix, is an important source of the uncertainty in the dose results after 10⁵ yr but not prior to 10⁵ 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 ~10⁵ yr. (In the TSPA-VA, perforation of the cladding from rock fall was the primary mechanism to damage cladding beyond 10⁵ yr.)

Prior to 10⁵ yr, the velocity (expressed as velocity divided by rod length) varies between 4 × 10⁻⁴/yr to 2 × 10⁻³/yr (Figure 3b). For comparison, the CSNF matrix degradation rate in TSPA-VA varied between 4 × 10⁻³/yr and 2 × 10⁻²/yr.

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REFERENCES


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


Figure 1. Cladding Degradation Component models perforation and unzipping as two steps\textsuperscript{3}
Figure 2. Perforation due to creep rupture and stress corrosion cracking (a) relationship based in TSPA-SR, and (b) comparison of Murry creep correlation (based on unirradiated cladding) with measurements of irradiated cladding.
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, and (b) unzipping rate for Always Drip condition.
Figure 4. Sensitivity of mean dose rates to degraded and enhanced cladding.