DISSOLUTION KINETICS OF COMMERCIAL SPENT NUCLEAR FUELS
IN THE POTENTIAL YUCCA MOUNTAIN REPOSITORY ENVIRONMENT

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Abstract

This report provides a summary of a literature survey and an independent analysis of relevant experimental data on the dissolution rate of commercial spent nuclear fuel (CSNF) for disposal at the potential high-level waste repository at Yucca Mountain, Nevada. This analysis was carried out to identify models and parameters for the CSNF dissolution rate, for use in performance assessment (PA) models. The survey and analysis focused on three key areas that potentially control the CSNF dissolution rate: (i) in-package water chemistry; (ii) CSNF conditions before aqueous dissolution; and (iii) the modes of groundwater contact with CSNF from the failure of the drip shield and the waste package components. Based on the key findings, two models and their parameters for the CSNF dissolution rates are presented to be used in PA. Two models and their parameters for determining the exposed CSNF surface area for water contact are also presented. These models and parameters are used for determining the rate of radionuclide transfer from CSNF into the contacting water in the WP. PA was conducted to assess the sensitivities of the identified models and parameter values, and the associated uncertainties in radionuclide release.
Executive Summary

This report provides a summary of a literature survey and an independent analysis of relevant experimental data on the dissolution rate of commercial spent nuclear fuel (CSNF) for disposal at the potential high-level waste repository at Yucca Mountain, Nevada. This analysis was carried out to identify models and parameters for the CSNF dissolution rate for use in performance assessment (PA) models. The survey and analysis focused on three key areas that potentially control the CSNF dissolution rate: (i) in-package water chemistry; (ii) CSNF conditions before aqueous dissolution; and (iii) the modes of groundwater contact with CSNF from the failure of the drip shield and the waste package components. PA was conducted to assess the sensitivities of the identified models and parameter values, and the associated uncertainties.

A literature review of the in-package water chemistry suggests that the factors controlling the CSNF dissolution rate are primarily: (i) carbonate concentration, (ii) cation availability in the contacting water, (iii) pH, (iv) oxygen concentration, (v) iron concentration, and (vi) temperature. The carbonate concentration alone linearly increases the CSNF dissolution rate. The carbonate concentration effect is not discussed extensively in this report because the CSNF dissolution rate determined in pure carbonate solution is conservative and, therefore, easily adopted in PA. Calcium and silica cations (Ca$^{2+}$ and SiO$_4^{4-}$) in an amount to be present in the potential in-package water tend to decrease the CSNF dissolution rate by as much as, or more than, 2 orders of magnitude, at 25 °C [77 °F] compared to those in pure carbonate solutions of 2×10$^{-4}$ – 2×10$^{-2}$ M. As the pH decreases below ~5 under oxidizing conditions, the CSNF dissolution rate increases (e.g., a factor of 10 - 20 increase at pH ~ 3). In the carbonate solution, the increasing factor was extended to a factor of 10 - 20. In the presence of steel components (i.e., introducing dissolved iron ions in the groundwater), local oxygen concentration may decrease, which in turn could decrease the CSNF dissolution rate. Decrease in the CSNF dissolution rate by more than an order of magnitude was observed from the tests when iron was present in sodium chloride (NaCl) solutions at 25 °C [77 °F] in a German analog repository environment. The CSNF dissolution rate varies with temperature, by a combination of temperature itself and the activation energy in a rate equation of Arrhenius type. The activation energies extracted from appropriate literature test date to the in-package water vary depending on measurement conditions such as solution chemistry. The values extracted are 24 – 33 kJ/mol [6 – 8 kcal/mol].

Important processes related to the conditions of CSNF before aqueous dissolution begins include the opening up of grain-boundary internal surface areas to solution due to dry oxidation and hydration prior to the dissolution, leading to the formation of grain-size or sub-grain-size powders. These processes could effectively increase the surface area of CSNF for water contact, thus increase the release of radionuclides (RN) from the CSNF to the contacting water. Given the same exposed surface area, the oxidized matrix (i.e., UO$_2$ or U$_3$O$_8$) and unoxidized matrix (i.e., UO$_2$) show similar dissolution rates. Hydrated UO$_3$ xH$_2$O ($x = 0.8, 2$) dissolves at 10 - 20 times higher rates than the unhydrated UO$_2$ matrix. However, hydration rates forming UO$_3$ xH$_2$O are slower than the aqueous dissolution rates. UO$_2$ tends to open up more grain boundaries, increasing the RN release rate by a factor of ~10 in 2 ×10$^{-2}$ M of total carbonate anions, pH of 8, and temperature of 25 °C [77 °F], and no increase in 2 ×10$^{-3}$ M of total carbonate anions, pH of 9, and temperature of 50 °C [122 °F]. U$_3$O$_8$ or UO$_3$ xH$_2$O tends to become sub-grain-sized powder, increasing the exposed surface area by as much as 2 to 3 orders of magnitude, compared to the geometric surface area of the CSNF fragment. Prolonged oxidation or hydration may weaken deeper more grain boundaries. Mechanical impact would result in powdering of the fragment into grains. However, such mechanical
impacts depend on the magnitude of the event. Available data on the CSNF dissolution rate indicate that the release from RN from the grain boundary and gap inventory can be substantial along with the true long-term matrix dissolution rate. Therefore, the true long-term matrix dissolution rate could be lower than the measured (i.e., apparent) dissolution rate by as much as an order of magnitude. This indication is obtained by a detailed analysis of (i) RN inventory of the grain boundary inventory, (ii) the observed apparent CSNF dissolution rate, and (iii) the dissolution rate of the unirradiated UO₂ matrix or CSNF grains which exclude the grain boundary RN inventory.

For groundwater to contact CSNF, the drip shield and the waste package (WP) components (i.e., the WP inner and outer overpack containers and the CSNF cladding) must fail. Depending on the failure modes of these components (e.g., overpack containers) and the flow rate of groundwater into the WP, the groundwater contact modes for CSNF will vary. Dissolution rate for CSNF are significantly influenced by immersion versus drip condition for CSNF, the flow rate of groundwater into the WP, and the degree of partial protection by failed cladding could significantly influence. Immersion tests show lower CSNF dissolution rate than high-drip rate tests, in J-13 well water obtained near Yucca Mountain. However, under low-drip rate conditions which appear more representative of repository conditions, the CSNF dissolution rate could be lower than the immersion tests. In the presence of partial protection by failed cladding, the RN release decreases significantly. Slit- or hole-defected CSNF rods under immersion conditions in J-13 well water at 85 °C [185 °F], showed RN release rates that decreased by a factor of ~140 for Tc-99, ~7 ×10⁵ for I-129, and ~ 65 for Sr-90, compared to bare CSNF. Partially clad rod segments under drip conditions also showed decreased Tc-99 release rates, to a factor of 10–100.

The rate of RN transfer from CSNF into groundwater in a WP is modeled as a function of the flow rate of groundwater contacting CSNF, water chemistry, and the RN solubility in this water. Fission- and activation-product RN normally have high-solubility limits that do not limit the release of these RN from a WP. These RN include Tc-99, I-129, Cs-135, C-14, and Cl-36. Np-237 may belong to this category also at high flow rates. These RN are considered to be released congruently with the dissolving CSNF matrix. Np-237 at low flow rates and other actinides such Pu-(239+240) are considered to be released at rates determined by solubility limits and groundwater flow rates. Two specific models for the CSNF dissolution are presented. The first model from the literature is based on an enhanced CSNF dissolution rate derived from tests in pure carbonate solutions. In this model, the dissolution rate is a function of carbonate concentration, pH, oxygen fugacity, and temperature. The second model is based on a CSNF dissolution rate considering all the above: (i) in-package water chemistry; (ii) CSNF conditions before aqueous dissolution; and (iii) the modes of groundwater contact with CSNF from the failure of the drip shield and the WP components. In this model, the dissolution rate is an Arrhenius form with a pre-exponential term incorporating other factors included in (i) to (iii).

The RN release is proportional to the surface area of CSNF exposed to groundwater because the CSNF dissolution rate is defined per unit exposed surface area. Two models for determining the CSNF surface area are presented in this report. The first model represents surface area, using fragmented pellets (i.e., particles), and the second model uses CSNF grain-size criteria. The particle model assumes that CSNF in a WP is fragmented into small spherical particles. The grain model assumes that smaller individual grains in particles are exposed. The effects of alpha-, beta-, and gamma-ray radiolysis were not considered. In the oxygen-buffered solution at the potential Yucca Mountain repository, radiolysis effects are offset by the oxygen effects within the uncertainty range. Ongoing work in Europe does not suggest the long-term
alteration of crystal structure/integrity or radionuclide redistribution. Nevertheless, PA models consider these uncertainties too.

To include the CSNF dissolution models in PA, various parameter values used in the model are considered and the associated ranges of the parameter values are presented to evaluate uncertainties in RN release. These models and parameters are used for determining the rate of radionuclide transfer from CSNF into the contacting water in the WP. PA was conducted to assess the sensitivities of the identified models and parameter values, and the associated uncertainties.
1. Introduction

Current plans for commercial spent nuclear fuel (CSNF) in the United States (U.S.) call for disposal in waste packages (WP), in drifts excavated nearly 300 meters (m) (984 feet, ft) below the ground surface, at the potential Yucca Mountain (YM) repository in Nevada. If the WP container, cladding, and other engineered barrier system components such as the drip shield (DS) fail, radionuclides (RN) such as technetium-99 (Tc-99), iodine-129 (I-129), and neptunium-237 (Np-237) could be mobilized upon the CSNF contact with groundwater and the subsequent dissolution of CSNF. The high solubility RN —Tc-99 and I-129—are released from the WP at the dissolution rate of the CSNF UO₂ matrix (i.e., dissolution rate). Under high-flow-rate conditions, Np-237 is also released from the WP at the dissolution rate. Np-237 at low flow rates and other actinides such Pu-(239+240) of low solubility are considered to be released by solubility limits and groundwater flow rates, after the continuous matrix dissolution and its subsequent precipitation back. The dissolution rate is a function of the evolved physico-chemical environment inside the WP, especially water chemistry inside a failed WP (i.e., in-package water chemistry) such as: (i) concentration of carbonate/bicarbonate (i.e., carbonate) anions; (ii) concentration of calcium (Ca) or silica (Si oxide) cations; (iii) pH; (iv) concentration of oxygen and iron; and (v) temperature. Further modifications of the in-package water chemistry may occur by groundwater interaction with materials inside the WP such as neutron poison materials. CSNF conditions before aqueous dissolution are also important to the dissolution process. Such conditions include the pre-hydration/oxidation of the CSNF UO₂ matrix (i.e., the matrix) prior to the dissolution, and the RN inventory of the grain boundaries. The dissolution rate is also a function of the mode in which groundwater contacts CSNF. The water contact mode influences the RN release primarily through the surface area and duration of contact between groundwater and CSNF. The mode of groundwater contact (immersion versus drip), the flow rate of groundwater past the CSNF, and partial protection by failed cladding influence the area and period of groundwater contact. The immersion test uses a small amount of CSNF sample in a batch of solution. The drip test uses a small volume of solution dripping onto the CSNF sample. The RN release is also proportional to the surface area of CSNF exposed to groundwater because the CSNF dissolution rate is defined per unit exposed surface area. The effects of alpha-, beta-, and gamma-ray radiolysis were not considered. In the oxygen-buffered solution at the potential YM repository, radiolysis effects are offset by the oxygen effects within the uncertainty range (Ferry, et al., 2005a,b; Poinssot, et al., 2005; Bechtel SAIC Company, 2004; Poinssot, 2001; Shoesmith, 2000). Ongoing work in Europe does not suggest the long-term alteration of crystal structure/integrity or radionuclide redistribution. Nevertheless, PA models consider these uncertainties too.

This report attempts to summarize the relevant key information, pertinent to the potential repository at YM, available in the literature on CSNF dissolution. This initial study is based on an acceptable “baseline-event sequence,” regarding the most abundant type of CSNF. This work helps inform NRC’s independent model for CSNF dissolution, potentially used in performance assessment. In the following sections, key findings are presented for: (i) the in-package water chemistry (ii) CSNF conditions before aqueous dissolution; and (iii) groundwater contact modes with CSNF from the failure of the DS and WP components. Based on the key findings, models and parameters for the CSNF dissolution rates are presented to be used in PA. Two models for determining the CSNF surface area are also presented. The first model determines surface area, using fragmented pellets (i.e., particles), and the second model uses CSNF grain-size criteria.
To include the CSNF dissolution models in PA, various parameter values and ranges used in the model are presented to treat uncertainties. These models and parameters are used for determining the rate of radionuclide transfer from CSNF into the contacting water in the WP. PA was conducted to assess the sensitivities of the identified models and parameter values, and the associated uncertainties.

2. Literature Survey

2.1 In-Package Water Chemistry

A literature review of the physico-chemical environment inside a failed WP suggests that the primary controlling parameters in the dissolution rate are: (i) concentration of carbonate anions; (ii) concentration of calcium (Ca) or silica (Si oxide) cations; (iii) pH; (iv) concentration of oxygen and iron; and (v) temperature [Shoesmith, 2000; Bechtel SAIC Company, 2004]. Table 1 summarizes CSNF dissolution rates from laboratory studies, selected from the literature, on the basis of their relevance to YM. Oversby and VMO Konsult [1999] indicate that the dissolution rate at the room temperature varies over a range of more than 2 orders of magnitude, mostly in the pH range of 5 – 10. The dissolution rate will vary by a factor of 10 – 20, if pH decreases to -3, from the near neutral values. Discussions about the effects of important environmental variables that were noted, follow.

2.1.1 Carbonate Effects

The carbonate concentration alone linearly increases the dissolution rate [Mohanty, et al., 2002]. The carbonate concentration effect is not discussed extensively in this report because the dissolution rates determined in pure carbonate solutions are generally higher than those in realistic groundwater containing magnesium, calcium, fluoride, nitrate, sulfate and silica ions, in addition to carbonate anions. Therefore, they are conservatively used in many performance assessment models. The discussion in this report focuses on more realistic dissolution rates considering various environmental and SNF conditions.

2.1.2 Cation Effects

Solutions with calcium and silica cations (Ca$^{2+}$ and SiO$_4^{4-}$) in an amount potentially present at the YM repository tend to decrease the dissolution rate by more than 2 orders of magnitude, at 25 °Centigrade (C) [77 °Farenheit (F)] compared to the pure carbonate solutions of 2×10^{-4} – 2×10^{-2} M [Shoesmith, 2000; Gray, 1997; Gray, 1992]. These tests were conducted at very high flow rates of a large volume of solution (i.e., flow-through tests). These decreased dissolution rates partly support the lower end of the distribution of the CSNF dissolution rates in PA. In the drip tests, these cations are consumed by forming uranosilicates. Consequently, reactions without cations are likely to form hydrated or dehydrated schoepite in drip tests. On the other hand, under a static immersion condition, the cations are expected to be available, although details of the localized chemistry between the uranium alteration (i.e., precipitation of continuously dissolved uranium) layer and the bare CSNF surface are not very well-known. Although the carbonate anions are known to ease the inhibition effect of these cations in the dissolution, the carbonate anions present in immersion tests or flow-through tests do not appear to be concentrated enough to affect the cation effect, as shown in Table 1. Additionally,
the presence of schoepite formed without the cations is also considered to decrease the
dissolution rates by impeding the transport of oxygen to the bare CSNF surface, possibly
forming less oxidizing local conditions between the schoepite and the bare CSNF surface.

2.1.3 pH Effects

Data on the pH dependence of the CSNF dissolution rates are not sufficient to cover a full
range of the pH in the expected repository environment. Nevertheless, an estimate of the pH
effect can be made, based on data from Torrero, et al. [1997] and Thomas and Till [1984],
under oxidizing conditions relevant to the potential YM environment. As the pH decreases
below \(-5\), the dissolution rate increases by a factor of \(10-20\) at pH \(\sim 3\), as shown in Table 1.
In the carbonate solution, the increasing factor was somewhat larger than \(10-20\), depending on
the choice of the reference pH above 6 [Röllin, et al., 2001]. The potential YM repository may
have low pH, because the reaction of WP components with limited amounts of groundwater
may result in forming stagnant groundwater films. These increased dissolution rates partly
support the higher end of the distribution of the CSNF dissolution rates in PA. The WP
components such as internal steel structure or WP itself include metallic ions, such as
chromium (Cr) ions, responsible for lowering pH. At low pH, cations such as calcium and silica
ions are theoretically known to be ineffective in lowering the dissolution rate [Shoesmith, 2000].
However, as shown in Table 1 [Forsyth, 1997; Forsyth and Wermer, 1985], Swedish tests in
Allard synthetic groundwater containing calcium and silica ions did not indicate any drastic
change of the dissolution rate resulting from ineffective cation effects at low pH.

2.1.4 Effects of Oxygen and Iron Concentration

Iron ions from the steel components into the in-package water may decrease local oxygen
concentration forming reducing environments, which in turn could decrease the dissolution rate.
The presence of iron in sodium chloride (NaCl) based groundwater and NaCl solution at
ambient temperature decreased the dissolution rates by more than an order of magnitude
[Quiñones, et al., 2001]. These decreased dissolution rates partly support the lower end of the
distribution of the CSNF dissolution rates in PA. However, the decreased dissolution rates in
the presence of iron may be localized and transitory. The production of oxidants from radiolysis
and abundant air inflow may buffer the reducing environment formed by iron. It is necessary to
understand better the iron effects with respect to the radiolysis and the air buffering. For
example, after 100–1000 years, gamma and beta radiation fields will decay away substantially,
wheras alpha fields will remain strong [Shoesmith, 2000].

2.1.5 Temperature Effects

The CSNF dissolution rate varies with temperature, by a combination of temperature itself and
the activation energy in a rate equation of Arrhenius type. The activation energies vary
depending on measurement conditions such as solution chemistry or CSNF conditions.
Literature data show activation energies of: (i) over 29–34 kilo-joule per mole (kJ/mol) [7–8 kilo-
calorie (kcal)/mol] in non-complexing neutral solutions; (ii) 47 kJ/mol [11 kcal/mol] in aerated
carbonate solutions; (iii) 20 kJ/mol [5 kcal/mol] in distilled water; and (iv) 26–31 kJ/mol [6–7
kcal/mol], in borate-buffered granite groundwater. In some experiments a decrease in the
dissolution rates was also observed with temperature; and insensitivity to temperatures was
additionally observed at low pH [Shoesmith, 2000]. The lower dissolution rate is obtained from
mineral deposits resulting in negative activation energies for strontium-90 (Sr-90) release and
very low activation energy values, \(10-18\) kJ/mol [2–4 kcal/mol]) for cesium-137 (Cs-137)
release. The release rates of these RN are used as markers for the CSNF dissolution, because they are not precipitated back to CNSF as uranium ions. The present work adopts activation energies obtained from the immersion tests of CSNF in J-13 well water (YM area groundwater), because this system is considered close to the potential YM environment. The obtained values are 24–33 kJ/mol [6–8 kcal/mol] for Tc-99 and I-129 releases. These values are within the range published in the other literature. The more likely in-package chemistry inside failed waste package may have variation in pH and limited availability of cations. Currently, data at different temperatures under these conditions are not available.

2.2 Condition of CSNF before Water Contact

Important processes related to the conditions of CSNF before aqueous dissolution begins include the opening up of grain-boundary surface area due to dry oxidation and hydration prior to the dissolution, leading to the formation of grain-size or sub-grain-size powders [Shoesmith, 2000; Gray, 1997; Gray and Wilson, 1995]. These could effectively increase the surface area of CSNF for water contact, thus increase the release of RN from the CSNF to the contacting water.

2.2.1 Pre-Oxidation/Hydration

Higher uranium oxides (UO$_{2.4}$ or U$_3$O$_8$) that form by oxidization of the uranium matrix dissolve at a rate similar to the unoxidized UO$_2$ matrix. Hydrated UO$_3$ xH$_2$O (x = 0.8, 2) dissolves 10–20 times faster than unhydrated oxides [TRW, 1997; Casas, et al., 1993]. However, the rate of hydration (i.e., the formation rate of UO$_3$ xH$_2$O) is slower than the aqueous dissolution rate [Finn, et al., 1998, 1996].

Tc-99 is used as a dissolution marker (including Tc-99 bounded in an $\varepsilon$-phase particle) because Tc-99 has high solubility and does not precipitate back to CSNF. Also, Tc-99 dissolves oxidatively in the higher ionic valence state, regardless of the pre-oxidation. These two characteristics show that Tc-99 is a good marker for the dissolution. Although UO$_{2.4}$ dissolves similarly to UO$_2$ per unit surface area, UO$_{2.4}$ tends to have more grain boundaries (i.e., greater surface area) in flow-through tests. This increases the release rate by a factor of ~10 in a solution at 25 °C [77 °F] with 2 ×10$^{-2}$ M total carbonate concentration and at pH of 8. No such increase was observed in 2 ×10$^{-3}$ M of total carbonate concentration at pH of 9 and temperature of 50 °C [122 °F] [Gray, 1997]. Tests at 50 °C [122 °F] could have alteration products that prevent grain boundary opening. Unlike flow-through tests at 25 °C [77 °F], it is generally understood that grain boundary openings are masked with alteration products in drip or immersion tests. U$_3$O$_8$ or UO$_3$ xH$_2$O tends to become sub-grain-sized powder, increasing the exposed surface area by as much as 2 to 3 orders of magnitude, compared to the geometric surface area of the CSNF pellet fragments. In addition to UO$_3$ xH$_2$O formation on the surface, extended vapor hydration or aqueous dissolution may weaken the grain boundaries by deeper preferential attacks along grain boundaries. Under the conditions of UO$_{2.4}$ formation or hydration, mechanical impact such as seismically initiated mechanical impact would result in powdering of the matrices into grains. With the current WP design being considered for the potential YM repository, dry oxidation or hydration of CSNF could occur prior to the dissolution in initially failed WP soon after the repository closure when the temperature peaks, especially if drift degradation would occur. There is a limited likelihood that the WP temperature would exceed 250 °C [482 °F] [Manepally, et al., 2003], which is necessary for U$_3$O$_8$ to form. UO$_{2.4}$ is likely to form below 250 °C [482 °F] during the early period in an initially failed WP. Typically,
UO$_2$ would form at relative humidity (RH) less than ~ 50, above which hydration would occur [Ahn, 1996].

Grain boundary attacks by hydration and aqueous dissolution would occur up to a depth of 20 - 30 grains [Finch, et al., 1999]. However, mechanical impacts depending on the magnitude of the event such as seismicity can break the fragment into grains. In PA, the large exposed surface area caused by oxidation or hydration may be exercised by sensitivity analyses, observing changes in the peak expected dose with respect to a base case.

2.2.2 Grain-Boundary Effects

In the current U. S. Department of Energy (DOE) [OCRWM, 2006; Bechtel SAIC Company, 2004] or NRC [Mohanty, et al., 2004] performance assessment models, the release of RN from its inventory at grain boundaries is conservatively assumed to be instantaneous. Because most measured RN releases include grain boundary inventory, it is necessary to determine the dissolution rate of the matrix alone to be used in long-term PA. With unirradiated UO$_2$, the true matrix dissolution rates are obtained by the uranium mass dissolved in flow-through test. In CSNF tests, powdered grains are used from which the grain boundary inventories are removed by pre-washing. Tests with fragments under all conditions of immersion, drip, or flow-through tests appear to measure a substantial amount of grain boundary RN inventory in determining the measured (i.e., apparent) dissolution rates, which are based on the release of Tc-99, I-129, Cs-137, or Sr-90 as markers for the matrix dissolution. A significant portion of the released RN could be from grain boundaries. The true matrix dissolution could be lower, compared to CSNF fragments, by at least an order of magnitude. This was indicated in the flow-through tests of unirradiated UO$_2$ grains, with/without addition of calcium/silica ions [Gray and Wilson, 1995; Gray, 1992; Wilson and Gray, 1990]. Unirradiated UO$_2$ grains do not contain RN in the matrix or grain boundaries. The true matrix dissolution rates partly support the lower end of the distribution of the CSNF dissolution rates in PA.

Although the initial instant RN release from grain boundaries of several-grain depths could be excluded in determining the long-term dissolution rate, further continuous inward penetration of groundwater along grain boundaries could possibly release the grain boundary inventory continuously. The grain boundary inventory could be greater by 2 percent, as shown in the footnote 1. Some evidence is shown in Figure 2, where grain tests showed no discrepancy between RN release and uranium dissolution. However, fragment tests always show higher release rates (note the release in log scale) of RN, compared to uranium. These fragments were supposed not to have grain-boundary contributions, because the samples were tested after initial immersion tests during which time most grain boundary RN were considered released. Johnson, et al. (1985) assessed the dominance of the grain-boundary inventory by a factor of 2$^{\pm}$10, in the observed RN releases, from the data of immersion tests longer than 8 years, in the Canadian repository system tests. Also, flow-through tests resulted in more RN release than immersion or drip tests. Flow-through tests are expected to have more aqueous

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1 This is based on the calculation of the surface area associated with grain boundaries attacked during aqueous dissolution. For fragment size ~ 1 millimeter (mm) [0.04 in.], grain size ~ 10 micrometer (µm) [4x10^-4in.], and an average 5-grain penetration (a maximum 20 - 30), ~ 15 percent of the total surface area of grain boundaries are attacked. Assuming a grain- boundary RN inventory ~ 2 percent (0.3 - 26.8 percent [Poinssot, 2001; Bechtel SAIC Company, 2004]) as an example in a linear scale, the grain boundary contribution will be ~ 0.3 percent of the total RN inventory, from 15 percent of grain-boundary surfaces. In the drip tests, at an average 10^-4/day fractional release [OCRWM, 2003] the RN release from the matrix for 8.5 years would be 0.3 percent.
penetration into grain boundaries. It is difficult to verify the true matrix dissolution rates from the micrographs of the altered layer of the fragments after dissolution tests. Determining a statistically averaged thickness of the altered layer over the entire sample requires an inordinate amount of time for the test and analysis.

2.3 Groundwater Contact Modes for CSNF

For groundwater to contact CSNF, the DS, the WP, and the cladding must fail. Depending on the failure modes of these components and the flow rate of groundwater, the groundwater contact modes for CSNF will vary. It is recognized that the immersion versus drip condition, the groundwater flow rate, and partial protection by failed cladding could significantly influence the CSNF dissolution rate.

2.3.1 Immersion versus Drip, and Flow Rate

The WP failure will form perforations and cracks on the WP surface. Depending on the location of the perforations or cracks, CSNF would be subject to immersion in groundwater or drips of groundwater.\(^2\)

To provide a basis for models developed for representing the CSNF dissolution rate over a long period of time, the experiments conducted to date can be divided into two broad categories – immersion tests and drip tests. Immersion tests have been conducted in a variety of groundwater chemistries from different locales [Wilson, 1990; Forsyth, 1997], because most candidate repositories are saturated with groundwater. However, drip tests are unique to the U.S. YM Project, where only a small volume of groundwater is expected to drip in an unsaturated repository. Nevertheless, immersion tests are relevant to the U.S. YM Project too, because dripping groundwater could accumulate in a failed WP. The drip tests were mainly conducted at Argonne National Laboratory (ANL) [Bechtel SAIC Company, 2004]. To understand mechanistically the results of immersion tests and drip tests, auxiliary flow-through tests are also used as laboratory-controlled (rather than repository-relevant) tests. Flow-through tests were designed to determine the true (i.e., intrinsic) dissolution rates on the surface of the CSNF, with an unlimited supply of reacting solutions. High flow rates were used, artificially, to remove any alteration-product deposits blocking the reacting surface.

Current ANL drip tests involve drips from 0.075 mL [4.6x10\(^{-3}\) in.\(^3\)]/3.5 days to 0.75 mL [4.6x10\(^{-2}\) in.\(^3\)]/3.5 days, over about 10 CSNF individual fragments, with each fragment 7.0 – 8.5 g [(1.5 - 1.9)x10\(^{-2}\) lb] [Finn, et al., 1998]. Considering the initial geometric surface area of each fragment as 2.1 cm\(^2\)/g [148 in.\(^2\)/lb] [Wilson, 1990] the drip rates per fragment

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\(^2\) NRC PA [Mohanty, et al., 2004] models the groundwater entry perforation on the WP crown and the groundwater exit perforation located at any place between the bottom and the crown of the WP. The distance between the entry and the exit is randomly sampled.
area would be $1.3 \times 10^{-3} \text{ mL/cm}^2 \text{-day}$ [5.1x(10^{-4} \text{-} 10^{-3}) \text{ in.}^2/\text{in.}^2 \text{-day}].$ On the other hand, the potential repository flow rate per unit CSNF surface area would be $\sim 1.7 \times 10^{-4} \text{ mL/cm}^2 \text{-day}$ [6.7x10^{-5} \text{ in.}^2/\text{in.}^2 \text{-day}] under conservative focused flow scenarios. This is an order-of-magnitude lower value than the laboratory conditions. ANL data show several orders of magnitude decrease in the RN release rates when the drip rates are lowered by an order of magnitude, $0.75 \text{ mL} [4.6x10^{-2} \text{ in.}^3]$ to $0.075 \text{ mL} [4.6x10^{-3} \text{ in.}^3]$ for 3.5 days, as shown in Figure 1. This suggests that the RN release rates under the repository conditions would be even lower because the flow rates would be lower than even the lowest value of the ANL laboratory test drip rates. These lower CSNF dissolution rates partly support the lower end of the distribution of the CSNF dissolution rates in PA.

Table 1 includes relevant CSNF dissolution rates under different water contact modes. The water contact mode can be categorized as: (i) immersion; or (ii) dripping, of groundwater containing calcium/silica, expected at the potential YM repository. The data for drip contact were obtained at the higher drip rates (i.e., $0.75 \text{ mL} [4.6x10^{-2} \text{ in.}^3]/3.5 \text{ days}$). Immersion tests show lower dissolution rates than high-drip rate tests in J-13 well water. Data from the auxiliary flow-through tests were included to determine the true dissolution rates of bare CSNF by high-flow, eliminating deposits of alteration products.

### 2.3.2 Partial Cladding Protection

In the presence of partial protection from failed cladding, the dissolution rate decreases significantly. Wilson [1990] conducted tests with slit (-0.015 cm [5.9 x 10^{-3} in.] diameter x - 2.54 cm [1 in.] length) - or hole ( - 0.02 cm [7.9 x 10^{-3} in.] diameter)-defected CSNF under immersion conditions in J-13 well water at 85 °C [185 °F]. The tests were intended to simulate cladding partially failed by localized corrosion or SCC. The RN release rates decreased by a factor of -140 for Tc-99, -7 x 10^{5} for I-129, and - 65 for Sr-90 [Wilson, 1990], compared to bare CSNF. Later ANL tests, with partially clad rod segments under drip conditions, also showed decreased Tc-99 release rates, to $2.6 \times 10^{-8} \text{ f/d} (\sim 0.04 \text{ mg/m}^2\text{-d} [5.7 \times 10^{-11} \text{ lb/in.}^2\text{-d}])$ [OCRWM, 2003]. Such decreases in dissolution rates, compared to bare CSNF, partly support the lower end of the distribution of the CSNF dissolution rates in PA.

However, in the segment tests of clad CSNF for light-water reactor CSNF immersed in Swedish Allard groundwater, the RN release rates did not decrease, compared to bare CSNF [Forsyth, 1997; Forsyth and Wermer, 1985]. In these tests, nearly half of the surface area was exposed. Similar conclusions can be drawn from tests with Canadian Deuterium-Natural Uranium Reactor CSNF, immersed in Canadian granitic groundwater [Stroes-Gascoyne, 1985]. These results suggest that cladding would not inhibit the dissolution rates if it fails catastrophically exposing a substantial surface area to the solution. In the most likely scenario, the unzipping is likely to occur in slit- or hole-defected cladding. Unzipping was observed in the ANL 1.5-year long tests, caused by stress generated by corrosion-product accumulation in the gap of cladding and the

---

3Under DOE’s design for a potential repository, one WP would have 21 Pressurized Water Reactor (PWR) assemblies. Each assembly would have 264 rods of ~ 1.1 cm [0.43 in.] diameter and ~ 400 cm [157 in.] length. Considering an approximate average size of an individual CSNF pellet as ~ 0.2 cm x 0.2 cm x 0.2 cm [0.08 in. x 0.08 in. x 0.08 in.], one WP would have ~ 2.4 x 10^{6} \text{ cm}^2 [3.7 x 10^{7} \text{ in.}^2] surface area. The modeled flow rate of groundwater is (1.5 - 150 L/year [9.2 x (10 - 10^{3}) \text{ in.}^3/\text{year} [NRC, 2002]. Focused flow on a conservative basis would go up to 1.5 \times 10^{4} \text{ L/year} [9.2 x 10^{3} \text{ in.}^3/\text{year}]] [Lin, 2003]. In the presence of focused flow, dry area without CSNF dissolution will also be present.
matrix from uniform corrosion of Zircaloy cladding at 175 °C [347 °F] [Bechtel SAIC Company, 2004]. The activation energy for Zircaloy uniform corrosion is large, ~93 kJ/mol [22 kcal/mol] [Wescott, 1995]. Thus, at lower repository temperatures, uniform corrosion would not be fast enough to build enough stress for cladding to unzip from the corrosion products. Below 100 °C [212 °F], estimated unzipping times based on this activation energy may exceed 10,000 years. Localized corrosion is likely to occur only at very low pH (less than 3) and temperatures below 95 °C [203 °F] [Brossia, 2002], which is needed to stabilize oxidants such as ferric ions. Such low pH conditions would occur with very low probabilities. In fact, in long-term CSNF storage facilities, cladding damage such as unzipping has not been identified even after 8 years, although inerting the environment inside the storage container may be contributing to cladding integrity [Wasywich and Frost, 1992].

3. Conceptual Models for CSNF Dissolution

The rate of RN transfer from CSNF into water in the WP is modeled as a function of the flow rate of water containing CSNF, the composition of water contacting CSNF, and the element solubility in that water. Fission- and activation-product RNs normally have high-solubility limits that do not limit the release of these RN from a WP. These RN include Tc-99, I-129, Cs-135, C-14, and Cl-36. Np-237 also may belong to this category of RN, depending on the chemistry and the flow rate of water in contact with CSNF. These RN are considered to be released congruently with the dissolving CSNF matrix in contact with groundwater. Np-237 at low flow rates and other actinides such Pu-(239+240) are considered to be released by solubility limits and groundwater flow rates, after the continuous matrix dissolution and its subsequent precipitation back. Gray and Wilson (1995) determined the dissolution rate of the CSNF matrix from flow-through tests. The flow-through tests use artificially high flow rates to eliminate the precipitation of alteration phases on the CSNF surface. Alteration phases may modify the dissolution rate of the CSNF matrix by altering the area of the reactive surface, or may otherwise affect radionuclide release by solubility limits of alteration phases/co-precipitation.

3.1 Specific Dissolution Models

Two models are presented below for PA. The first model, referred to as Model 1, estimates the CSNF dissolution rates in solutions containing pure carbonate anions, which are conservative values. The pure carbonate solution is representative of altered in-package groundwaters. Model 1 was obtained from the DOE’s CSNF dissolution model for Total System Performance Assessment [Gray and Wilson, 1995]. The CSNF dissolution rate in this model is a function of temperature, pH, total carbonate concentration, and oxygen fugacity. Gray and Wilson (1995) express this model as:

\[
\log r = 9.310 + 0.142 \log \left[ \text{CO}_3^{2-} \right] - 16.7 \log \left( p_{O_2} \right) + 0.140 \log \left[ \text{H}^+ \right] - \frac{2130}{T} + 6.81 \log \left( T \right) \log \left( p_{O_2} \right)
\]

(1)

The models presented here are based on an NRC’s PA (Mohanty, et al., 2002). Recently, updated formulae are being used, although the modifications are minor.
where \( r \) is the dissolution rate \([\text{mg m}^{-2} \text{ d}^{-1}]\) \([1.42 \times 10^{-9} \text{ lb/in.}^3 \text{d}]\), \([\text{CO}_3^{2-}]\) is the total carbonate concentration \([\text{M}]\), \([P_{\text{O}_2}]\) is the oxygen partial pressure \([\text{atm}]\) \([1 \text{ atm} = 101 \text{ kPa}]\), \([H^+]\) is the concentration of hydrogen ions \([\text{M}]\), and \( T \) is the temperature \([\text{K}]\). The second model (Model 2) is derived from the assessments presented previously in this report. In Model 2, the CSNF dissolution rate, \( r \) \((\text{mg/m}^2\text{-d})\) is represented by:

\[
 r = r_0 \exp \left( -\frac{E_a}{RT} \right)
\]

where \( E_a \) is the activation energy, 29 kJ/mol \([7 \text{ kcal/mol}]\), from immersion tests and \( r_0 \) is the pre-exponential coefficient that varies between \(1.2 \times (10^3 - 10^6) \text{ mg/m}^2\text{-d} \) \([1.7 \times (10^6 - 10^3) \text{ lb/in.}^3\text{-d}]\). These values were selected to simulate the dissolution rates in the range 0.01 - 10 mg/m^2-d \([1.4 \times (10^{-1} - 10^{-8}) \text{ lb/in.}^3\text{-d}]\) at room temperatures and in mineral waters extending to low pH. They are extracted from Table 1, considering cation effects, low pH effects, iron effects, the effects of grain boundary inventory, the effects of low-drip rates, and partial protection by failed cladding. In PA, \( r_0 \) values have a log-uniform distribution and is sampled randomly.

### 3.2 CSNF Surface Area

Two models for determining the surface area are available in PA. The first model determines surface area, using fragmented pellets (i.e., particles), and the second model uses grain-size criteria. The particle model assumes that CSNF in a WP is fragmented into small spherical particles \((-1 \text{ mm [0.04 in.]}\) diameter), and the intergranular porosity does not contribute to the surface area. The total surface area for the particle model is then computed as:

\[
 A_p = \left( \frac{M_{\text{SF}}}{M_p} \times \frac{4\pi r_{po}^2}{3} \right) f_{\text{wet}}
\]

where \( M_p = \frac{4}{3} \pi r_{po}^3 \rho_{go} \) is particle mass \([\text{kg}]\) \([1 \text{ lb} = 0.4536 \text{ kg}]\), \( A_p \) is the particle surface area \([\text{m}^2] \) \([1 \text{ m} = 39.4 \text{ in.}]\), \( M_{\text{SF}} \) is the CSNF inventory/WP \([\text{kg}]\), \( r_{po} \) is the particle radius \([\text{m}]\), \( \rho_{go} \) is the density of oxidized CSNF \([\text{kg m}^{-3}]\), and \( f_{\text{wet}} \) is the volume fraction of CSNF immersed in the WP water.

If subgranular fragmentation takes place through conversion from \(\text{UO}_2\) to \(\text{UO}_{2.4}\) and \(\text{U}_3\text{O}_8\), a smaller particle size (i.e., equivalent) can be considered in the particle model, to represent additional exposed surface area. In the case of the second model, in which grains are exposed, the following expression is used to compute the total surface area:

\[
 A_g = 4 \times 10^{-12} \pi \left( \frac{M_{\text{SF}}}{M_p} \right) \left[ (r_g - w)^2 + \frac{3}{r_g} \left( 3r_g^2w - 3r_gw^2 + w^3 \right) \right] f_{\text{wet}}
\]
where \( A_g \) is the grain surface area \([\text{m}^2]\), \( r_g \) is the grain radius \([\mu \text{m}]\), and \( w \) is the width of the oxidized zone \([\mu \text{m}]\).

The surface area available for dissolution is conservatively held constant throughout the dissolution period, even though the radius of the unoxidized grains or particles would decrease with time. This simplification is most likely to make a difference to peak dose. In addition, preferential attacks of grain boundaries in the particles were not considered, since the effect could be masked by the presence of alteration phases under the potential repository conditions of low flow rates.

### 3.3 Treatment of Uncertainty

Table 2 shows parameter values and ranges used in the models 1 and 2 for dissolution rate and surface area. Most parameters are assigned constant values, except three: (1) initial radius of UO\(_2\) particle \([\text{normal distribution}]\); (2) subgrain fragment radius of UO\(_2\) particle after transgranular fracture \([\text{normal distribution}]\); and (3) pre-exponential factor for CSNF dissolution rate from \([\text{log-uniform distribution}]\). Other parameters are fixed at constant values either because these are better determined \(\text{(i.e., less uncertainty)}\) or the model output is less sensitive to uncertainty.

### 4. System-Level Sensitivity Analysis

The sensitivity of the CSNF dissolution rates in PA was exercised to illustrate the influence of the dissolution rate on the peak expected dose, which is the system-level performance metric. Sensitivity was studied by observing changes to the peak expected dose with respect to a base case. The details and results of an example exercise are given in Mohanty, et al [2004]. For example, dissolution models 1 and 2 were exercised to see the effects of various factors involved on the dose. Similarly, the two models of surface area were exercised to see the effects of particles and grains on the dose. Finally, various potentially possible distributions of the parameter values were also exercised to see the sensitivity of the choice on the dose.

### 5. Summary

The literature survey and independent analysis of CSNF dissolution kinetics relevant to the potential YM repository provide several key understandings. Presence of calcium and silica cations could decrease the CSNF dissolution rates more than an order of magnitude. Low pH, near 3, increase the CSNF dissolution rates by as much as a factor of 20. Iron components could form reducing environments, which reduce the CSNF dissolution rates by more than an order of magnitude. Dry oxidation and hydration in the early period for initially failed WPs may occur with limited likelihood. These processes could increase the effective surface area by a factor of 10–1000. The radionuclide release from the grain boundary and gap inventory appears to be substantial and the true matrix dissolution rates could be lower than the apparent dissolution rates by as much as an order of magnitude. Both immersion and drips are possible groundwater contact modes with CSNF. Drip tests were conducted at drip rates much higher
than the repository conditions. The CSNF dissolution rates at lower drip rates could be lower by more than an order of magnitude. Partial protection by failed cladding could decrease the CSNF dissolution rates more than 50 times. Based on these key observations, models and parameters for the CSNF dissolution rates are presented for use in PA. Two models for representing the CSNF surface area are also presented. Finally, some independent PA exercises for system-level sensitivity are introduced to illustrate the influence of the dissolution rate on the dose.

6. Acknowledgments

The NRC staff views expressed herein are preliminary and do not constitute a final judgment or determination of the matters addressed or of the acceptability of a license application for a geological repository at Yucca Mountain. The paper was prepared to document work performed by CNWRA for NRC under Contract No. NRC–02–02–012. The activities reported here were performed on behalf of the NRC Office of Nuclear Material Safety and Safeguards, Division of High-Level Waste Repository Safety. This report is an independent product of the CNWRA and does not necessarily reflect the view or regulatory position of the NRC.

7. References


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OCRWM, CSNF Waste Form Degradation: Summary Abstraction, ANL-EBS-MD-000015, Revision 01, 2003


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Table 1. Relevant CSNF Dissolution Rates in the Potential YM Repository

<table>
<thead>
<tr>
<th>Dissolution Rate, (mg/m²-d)</th>
<th>Sample Tested</th>
<th>Solution Chemistry</th>
<th>pH</th>
<th>T (°C)</th>
<th>Surface Area*</th>
<th>Test Method</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2 - 1.0**</td>
<td>CSNF</td>
<td>J-13</td>
<td>8.4</td>
<td>25</td>
<td>1</td>
<td>I</td>
<td>Wilson, 1990</td>
</tr>
<tr>
<td>0.2</td>
<td>Archived CSNF</td>
<td>J-13</td>
<td>8.4</td>
<td>22</td>
<td>1</td>
<td>FT</td>
<td>Gray, 1992; Gray and Wilson, 1995</td>
</tr>
<tr>
<td>3 x (10⁻² - 1)</td>
<td>UO₂</td>
<td>NaHCO₃ +CaCl₂ +Silicic Acid</td>
<td>8.4</td>
<td>25</td>
<td>2</td>
<td>FT</td>
<td>Gray and Wilson, 1995</td>
</tr>
<tr>
<td>(0.8 - 2.5) x 10⁻²</td>
<td>UO₂</td>
<td>Silicate Solution</td>
<td>near neutral</td>
<td>25</td>
<td>1</td>
<td>FT</td>
<td>Tait and Luht, 1997</td>
</tr>
<tr>
<td>0.07</td>
<td>CSNF</td>
<td>Allard Synthetic Ground Water</td>
<td>8.1 (3.2)</td>
<td>20 - 25</td>
<td>1</td>
<td>I</td>
<td>Forsyth, 1997; Forsyth and Werner 1985</td>
</tr>
<tr>
<td>x (10 - 20) at pH=3</td>
<td>UO₂</td>
<td>Dilute KCl or KOH NaClO₄</td>
<td>- 3 w.r.t neutral</td>
<td>25</td>
<td>1</td>
<td>FT</td>
<td>Torrero, et al., 1997</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>- 3 w.r.t neutral</td>
<td>20 - 30</td>
<td>2</td>
<td>FT</td>
<td>Thomas and Till, 1984</td>
</tr>
<tr>
<td>25</td>
<td>CSNF</td>
<td>2 x 10⁻² M total carbonate</td>
<td>8</td>
<td>25</td>
<td>1</td>
<td>FT</td>
<td>Gray, 1997</td>
</tr>
<tr>
<td>2 - 12</td>
<td>UO₂, CSNF</td>
<td>5 x (10⁻³ - 10⁻²) M total carbonate</td>
<td>8 - 10</td>
<td>25</td>
<td>1</td>
<td>FT</td>
<td>Bechtel SAIC Company, 2004</td>
</tr>
<tr>
<td>0.023 - 0.165</td>
<td>SIMFUEL</td>
<td>Allard and Granite Groundwater</td>
<td>near-neutral</td>
<td>25</td>
<td>2 (1.52x 10⁻³ m²/pellet)</td>
<td>***</td>
<td>Oversby and VMO Konsult, 1999</td>
</tr>
<tr>
<td>3.7x10⁻²</td>
<td>CSNF (Segment)</td>
<td>Carbonate Groundwater with Ca and Si</td>
<td>7.2</td>
<td>25</td>
<td>1</td>
<td>I</td>
<td>Jégu, et al., 2004, 2001</td>
</tr>
<tr>
<td>0.19 - 0.57</td>
<td>CSNF</td>
<td>Clay, Granite Groundwater</td>
<td>near-neutral</td>
<td>25</td>
<td>2</td>
<td>I</td>
<td>Grambow, et al., 2000</td>
</tr>
</tbody>
</table>
I= Immersion, FT= Flow-through; CSNF=Commercial Spent Nuclear Fuel; 
Tc (Sr if Tc is unavailable) was used as a marker; all under oxidizing conditions otherwise specified 

* Two surface areas were used. The specific surface area (e.g., Brunauer, Emmett, and Teller, BET, measurement) considering the surface roughness is presented by 1 (normally ~7.2 cm²/g for CSNF fragments). The simple geometric surface area is presented by 2 and it is normally smaller by a factor of 3 for CSNF fragments. Except for flow-through tests, it is unlikely to have a roughness factor as the alteration phase masks the surface. It is user’s choice in the performance assessment.  
**( - x 1/140, partially cladded); 
***static tests, increase - x2.5 in flow-through tests;  

\[ \text{mg m}^{-2} \text{d}^{-1} = 1.42 \times 10^{-9} \text{lb/in}^{2} \text{-d} \]

\[ ^\circ F = ^\circ C \times \frac{9}{5} + 32 \]
Table 2. Parameter values selected for the TPA code based on literature survey (Mohanty et al., 2002)

<table>
<thead>
<tr>
<th>Parameter Description</th>
<th>Distribution and Range</th>
<th>Rationale</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spent fuel density (kg/m³)</td>
<td>Constant 10,600</td>
<td>Wilson (1990); based on ( \text{UO}_2 )</td>
</tr>
<tr>
<td>Negative log₁₀ of carbonate concentration in surrounding water (M)</td>
<td>Constant 3.71</td>
<td>Revised value that accounts for reinterpretation of literature data. Previous MULTIFLO runs based on only ( \text{CO}_3^{2-} ); new MULTIFLO runs based on the total carbonate concentration.</td>
</tr>
<tr>
<td>Radius of ( \text{UO}_2 ) grain (m)</td>
<td>Constant 1.25×10⁻⁵</td>
<td>Einziger, et al. (1992); Belle (1961). Sensitivity Analysis indicates low sensitivity.</td>
</tr>
<tr>
<td>Cladding correction factor</td>
<td>Constant 1.0</td>
<td>A factor that describes the fraction of cladding protection lost. Assumes no cladding protection when the factor is 1.0.</td>
</tr>
<tr>
<td>Subgrain fragment radius of ( \text{UO}_2 ) particle after transgranular fracture (m);</td>
<td>Normal 5.0×10⁻⁷, 2.0×10⁻⁶</td>
<td>Used only if fuel conversion takes place from ( \text{UO}_2 ) to ( \text{UO}_2.4 ) and ( \text{U}_3\text{O}_8 ); used only by the CSNF dissolution models that are dependent on exposed surface area. Mohanty, et al. (2002). Distribution based on distribution of grains or particles. Sensitivity Analysis indicates high sensitivity. Continue evaluating.</td>
</tr>
<tr>
<td>Pre-exponential factor for spent fuel dissolution rate from (mg -2 d⁻¹)</td>
<td>Log-uniform 1.2×10³, 1.2×10⁹</td>
<td>See the text and Table 1.</td>
</tr>
</tbody>
</table>

English conversion in the text
Figure 1. CSNF Drip Test Results of ANL with J-13 Well at 90°C [high-drip-rate: 0.75 mL, low-drip-rate: 0.075 mL, for 3.5 days, Cunnane, 1999]
Figure 3. Dissolution Rates of ATM-105 Spent Fuel in 2 x 10^{-3} M NaHCO3/Na2CO3, pH = 9, O2 = 0.2 atm, 50°C [Gray and Wilson, 1995]
Figure 6. Schematic of Bathtub Model with Incoming and Outgoing Water Conduits [Mohanty et al., 2002]