REVIEW OF THE U.S. DEPARTMENT OF ENERGY DISPOSAL CRITICALITY TOPICAL REPORT

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Request for Additional Information on DOE's Disposal Criticality Analysis Methodology Topical Report YMP/TR-004Q, Revision 0

Introduction

The U.S. Congress charged the U.S. Department of Energy (DOE) with managing the geologic disposal of high-level radioactive waste (HLW) and spent nuclear fuel (SNF) through the Nuclear Waste Policy Act of 1982 and the Nuclear Waste Policy Amendments Act of 1987. As part of DOE's efforts to license the geologic repository at Yucca Mountain, Nevada, a topical report has been submitted for review to the Nuclear Regulatory Commission (NRC) on the methodology that will be utilized in the License Application (LA) to demonstrate the potential for, and consequences of, the materials within the repository achieving a critical configuration. This methodology has been proposed to be applied to all waste forms (WFs) that will be placed in the repository including commercial SNF, DOE SNF including naval SNF, immobilized plutonium, and vitrified HLW glass. NRC staff have agreed to review this topical report in order to expedite the review of the DOE LA. NRC acceptance of this topical report will constitute approval of the methodology utilized by the DOE to demonstrate post closure criticality safety inside the waste package (WP), in the near-field and in the far-field, but will not close the issue of post closure criticality for the LA. The DOE LA must demonstrate that the methodology has been used to analyze all types of fuel that will be disposed in the repository and that the probability and consequences of criticality within the repository system are acceptable.

In fiscal year 1999, Center for Nuclear Waste Regulatory Analyses staff conducted a review of the topical report. Due to division of work with NRC staff, the focus of this review has been on the following aspects of the topical report: (i) master scenario list, (ii) methodology for developing configurations, (iii) probability of critical configurations occurring, and (iv) degradation models of the WP and WF. However, comments on other aspects of the topical report have been included, if identified. Based on this review and additional discussions, the following comments have been generated for the topical report.

Section 3.1

3-1.   Indicate how the effects of disruptive events will be considered in the evaluation of potential criticality events in the repository.

The scenarios listed in figures 3-1 and 3-2 appear to be comprehensive for an undisturbed repository. However, it is not clear whether the potential effects of disruptive events have been adequately considered. Failure to consider all potential scenarios that could result in a criticality event could result in an underestimation in the probability of a critical event occurring within the repository. Some potential effects of disruptive events include:

a.   Seismic events could cause the WPs to rotate on their invert, potentially allowing corrosion products to be released while the hole in the WP is facing down. Later, the hole could rotate back to the top of the package, allowing the package to fill with water.

b.   A volcanic event, although a low probability event, could fail many WPs and force their contents into a compact configuration encased in lava at the end of the tunnel.

Section 3.1.2

3-2.   Clarify whether credit will be taken in the criticality analyses for the assumption that there is no mechanism for completely sealing the fractures in the bottom of the drift so any in-drift accumulations of water will only be present for a few weeks.

Previous investigations have indicated that thermal alteration of the rock surrounding the repository or microbial growth has the potential to seal fractures, at least in local portions of the repository (Lin and Daily, 1989). Although it appears that the scenarios listed in figure 3-2 include the potential for water to pond on the bottom of the drift, if credit is taken for the short duration of ponding water in the drift, this assertion that fractures cannot become sealed will have to be justified.

3-3.   Confirm that far-field configuration classes FF-3c, 3d, and 3e are located in the saturated zone.

The text in item 1 of section 3.1.2 contradicts figure 3-2b in assigning these configurations to the unsaturated zone. The distinction is important in modeling hydrologic and geochemical processes.

3-4.   Explain why, in item 3, configuration NF-1b includes only a reducing reaction with tuff as a mechanism for precipitation of fissile solutes in the near-field below the WP.

Other chemical reactions should be considered as causing such precipitation, such as changes in aqueous chemistry related to the presence of concrete and tuff. This comment reflects the desire for completeness in modeling the configurations.

3-5.   Correct item 5 to state that the final two configurations are NF-3b and 3c, rather than FF-3b and 3c.

The context of the sentence implies incorrectly that the listed sites of colloidal accumulation are all in the far-field. The list of configurations should read "FF-2a, -2b, and -2c, and NF-3b and -3c." In addition, in the final sentence "open fractures" should be specified as being in concrete to be consistent with figure 3-2a. These changes will correct the impression from item 5 that all colloidal accumulation sites are far-field.

Reference

Lin, W., and W.D. Daily. 1989. *Laboratory Study of Fracture Healing in Topopah Spring Tuff–Implications for Near-Field Hydrology*. UCRL–100624. Livermore, CA: Lawrence Livermore National Laboratory.

Section 3.2

3-6. State whether temperature is included among the parameters quantified at this stage of the methodology and describe possible thermal variations.

Because equilibrium states and degradation/reaction rates for WP internal components (including WF) are temperature-dependent, all geochemical modeling should include sensitivity to temperature variations, including those caused by repository heating and cooling and by criticality events. With respect to internal configurations, the example analysis of appendix C refers to EQ6 calculations described in CRWMS M&O (1998e, appendix C, reference list). The discussion in this reference does not explicitly mention temperature constraints on models. Recent DOE modeling of the near-field (Hardin, 1998) predicts that temperatures of close to 100 °C may persist at the repository horizon 5000 yr after closure. A more recent repository design, EDA-II[1] yields lower drift temperatures, but the WP would still experience temperatures above 60 °C for at least 2000 yr. Thermal variations have a strong effect on degradation processes and rates for WP internal components. Furthermore, high temperatures would affect water chemistry [e.g., see composition of J-13 equilibrated with tuff at 90 °C in Wronkiewicz et al. (1992)]. This comment applies also to discussions of internal and external geochemistry models in topical report sections 3.3, 4.2.2, and 4.2.3.

References

CRWMS M&O. 1998e. *Evaluation of Codisposal Viability for Aluminum-Clad DOE-Owned Spent Fuel: Phase II Degraded Codisposal Waste Package Internal Criticality.* BBA000000–01717–5705–00017. Revision 01. Las Vegas, NV: CRWMS M&O.

Hardin, E.L. 1998. Near-Field/Altered Zone Models Report. UCRL–ID–129179. Livermore, CA: Lawrence Livermore National Laboratory.

Wronkiewicz, D.J., J.K. Bates, T.J. Gerding, E. Veleckis, and B.S. Tani. 1992. Uranium release and secondary phase formation during unsaturated testing of UO_2 at 90°C. *Journal of Nuclear Materials* 190: 107–127.

Section 3.4.3

3-7. Indicate whether consideration was given to isotopes that may activate to a fissile isotope in the selection of principal isotopes.

The selection of principal isotopes should consider all radionuclides that have the potential to have a significant positive effect on the reactivity of the system. Under storage conditions, the WPs will have a low neutron flux throughout the package due to (α, n) decays and fission within the package. Over long periods of time, this could lead to the creation of additional fissile material due to activation of other isotopes. For example, activation of Th-232 could yield additional U-233, which is fissile. DOE should indicate whether this

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[1]Snell, R. 1999. Presentation to the Advisory Committee on Nuclear Waste.

possibility was considered when determining principal isotopes or justify not considering this process.

Section 3.5

3-8.   Indicate whether the criticality calculations will account for neutron interactions between WPs.

The calculation of the effective neutron multiplication factor, k_{eff}, should account for all fissile material that can impact the modeled system. The EDA-II[2] design places the WPs much closer together in the line-loading formation, which will lead to greater neutronic interaction between the packages. It is not clear from the topical report whether these effects will be accounted for when calculating the k_{eff} of the fuel both inside the WP and in the near-field.

3-9.   Justify the assumption that Fe_2O_3 is the product that is formed by the corrosion of iron.

Credit is being taken for the filling of breached WPs by iron corrosion products, namely Fe_2O_3, thereby limiting the quantity of water present. It is unclear why the possibility that some of the iron corrosion product may be in the form of FeOOH was not considered. Justification of why the formation of FeOOH in lieu of Fe_2O_3 was not considered should be provided or else the effects of FeOOH formation on criticality control should be determined.

Section 3.6.1

3-10.   Provide justification for the claim that criticality transients that can occur in the repository will be sufficiently slow that significant kinetic energy will not be released.

The topical report outlines the conditions that would cause a transient to release significant kinetic energy, but does not demonstrate why the SNF casks will not experience these conditions. The release of significant kinetic energy from a transient criticality could damage either the fuel rods or the canister, thus increasing the degradation rate of the WF and the resulting dose following the criticality event.

Section 4.2.1

4-1.   Justify the extensive reliance on the wide range of corrosion rates utilized to determine the probability and location of a WP breach.

Given sufficient criticality control in the as-fabricated WP, a breach in the WP is necessary for a criticality event to occur. The model used to determine the probability of a WP breach and its location was the WAPDEG code using the Total System Performance Assessment (TSPA)-Viability Assessment (VA) base case. The primary limitation of this case is that the input parameters for corrosion rate rely extensively on expert elicitation, with nearly five orders of magnitude variance in the corrosion rate utilized. Thus, the possibility exists for a wide range of WP failure times and a commensurately wide range of times in which

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[2]Snell, R. 1999. Presentation to the Advisory Committee on Nuclear Waste.

criticality control becomes important. The wide range of corrosion rates resulting from the heavy reliance on expert elicitation is considered a limitation to the utility and validity of the subsequent criticality analysis, since it leads to dilution of the probability of occurrence and resulting risks.

4-2.   Provide justification for the long-term credit being taken for the presence of fuel cladding in the degradation analysis.

It appears that credit is being taken for the presence of Zircaloy-4 cladding in terms of its corrosion resistance. Although Zircaloy-4 does have good corrosion resistance, it is known to suffer from localized corrosion under reasonably attainable conditions inside breached WPs. Additionally, the cladding can be degraded prior to disposal due to the effects of irradiation, reactor water chemistry, and predisposal storage conditions. Commercial SNF exhibits a wide range of Zircalloy material characteristics, including large variations in the degree of hydriding, oxidation, erosion thinning, embrittlement, crack formation, pellet-cladding interactions, crud depositions. Further information is requested on the technical basis that this degree of credit can be claimed for the Zircaloy-4 cladding and the effect on criticality control if no credit was taken.

Section 4.2.2

4-3.   Justify the use of J-13 well water as representative of the solution that would be present within the WP.

Once the WP is breached, corrosion and degradation of WP internals play important roles in criticality control. Although some testing data has been obtained, the results are based on experiments conducted in variants of simulated J-13 well water. It seems unlikely that the chemistry of the solution inside a WP would be J-13. Furthermore, it is unclear if the testing program and the subsequent analysis has considered the possibility of chemistry changes resulting from evaporative processes and dissolution products from WP components (e.g., acidification due to metal cation hydrolysis, alkalinization from dissolution of HLW glass, etc.). Because of possible chemistry changes, the corrosion mode and corrosion rates could be altered from the general corrosion case considered. For example, alkalinization could lead to the formation of a passive film on carbon steel components that could then experience localized corrosion in the form of pitting or crevice corrosion in the presence of chloride. Similarly, the corrosion mode of stainless steel components could change from relatively slow passive dissolution to more rapid localized corrosion, which could lead to unanticipated, catastrophic failure of WP internal components. These accelerated corrosion modes could make conditions for criticality more favorable by allowing fuel materials to coalesce. Further information justifying the environments chosen and any further work examining likely alternate chemistries and their effects on material degradation is requested.

4-4.   Clarify the internal geochemistry model treatment of U produced from WF degradation.

WF U is input into solution (along with other degradation products) according to fixed degradation rates and solution evolution is modeled with EQ6. What is not clear is if U secondary phases are allowed to precipitate, in effect lowering the U release rate and

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perhaps lowering the probability for potentially critical external accumulations. References cited in the topical report suggest that secondary U phases will be included in internal degradation models. These references include CRWMS M&O (1998e) and CRWMS M&O (1998q), the latter being cited in the former. For example, in section 6.3.2 of CRWMS M&O (1998e), EQ6 models of SNF degradation are said to lead to precipitation of the hydrated uranyl silicate soddyite. In contrast, retention by secondary U phases was not modeled in the TSPA-VA. Clarify whether these differing approaches will be reconciled in future work.

4-5. Compare and contrast the approach to modeling to be used for release and internal geochemistry in the criticality analysis with that employed in present and future TSPA models (which may or may not use EQ3/6).

It was stated in topical report section 4.2.1 that WF degradation modeling in the criticality analysis will employ TSPA model approaches, but it is not clear if this extends to modeling release from WPs. TSPA-VA did not explicitly employ EQ3/6 geochemical modeling of WF alteration, RN release, and secondary solid phase formation. Any deviations from the TSPA-VA approach in the criticality analysis should be demonstrably more conservative or supportable.

4-6. Specify what kinetic models will be used in the internal geochemistry models and whether default EQ6 values will be used.

Selection of kinetic models profoundly influences model results regarding degradation products and water chemistry. Cited documents discussing EQ6 degradation models (CRWMS M&O, 1998e, appendix C reference 1998e, 1998q,) do not address kinetic models affecting the rates at which WP and WF degradation products precipitate. It appears that either default EQ6 kinetic parameters are utilized or kinetics are not included. Because degradation products are integral to criticality models, calculations predicting their formation should rely on supportable or conservative kinetic data. This comment applies also to the external geochemistry models discussed in section 4.2.3 of the topical report.

References

CRWMS M&O. 1998e. *Evaluation of Codisposal Viability for Aluminum-Clad DOE-Owned Spent Fuel: Phase II Degraded Codisposal Waste Package Internal Criticality.* BBA000000–01717–5705–00017. Revision 01. Las Vegas, NV: CRWMS M&O.

CRWMS M&O. 1998q. *Geochemical and Physical Analysis of Degradation Modes of HEU in a Codisposal Waste Package with HLW Canisters.* BBA000000–01717–0200–00059. Revision 01. Las Vegas, NV: CRWMS M&O.

Section 4.2.3

4-7. Describe how colloidal deposition will be incorporated into modeled chemical deposition.

Indicate whether the approach will be the same as those adopted under TSPA. Evaluation of models of fissile material accumulation requires full understanding of colloid modeling. A

previous analysis of external criticality (CRWMS M&O, 1998p) concluded that colloidal transport and accumulation of fissile materials would be insignificant. It should be clear how new analyses will differ and to what extent they are supportable and conservative.

Reference

CRWMS M&O. 1998p. *Report on External Criticality of Plutonium Waste Forms in a Geologic Repository.* BBA000000–01717–5705–00018. Revision 01. Las Vegas, NV: CRWMS M&O.

Section 4.2.4.1

4-8. Provide more information on validation methods for the "pseudo flow-through" internal and "open system" external EQ6 models.

With regard to the "pseudo flow-through" model, topical report section 4.2.4.1 refers only to hand calculations supporting the solute concentration adjustments (CRWMS M&O, 1998q). This exercise only partially addresses the question of the validity of the model results.

In discussing the "open system model," it is stated that the results are conservative, but the pertinent reference (CRWMS M&O, 1997f) is missing from the chapter 6 reference list. The report acknowledges that validation has not yet been done, but does not describe how it will be done. This information is vital to assessing the methodology (see also discussion of topical report section 4.2.4.2 below). Validation approaches should provide confidence that models will not underestimate the effects of processes that could lead to criticality.

Reference

CRWMS M&O. 1998q. *Geochemical and Physical Analysis of Degradation Modes of HEU in a Codisposal Waste Package with HLW Canisters.* BBA000000–01717–0200–00059. Revision 01. Las Vegas, NV: CRWMS M&O.

Section 4.2.4.2

4-9. Provide more information on the validation of EQ3/6 for the specific applications.

The validation examples provided in topical report section 4.2.4.2 (Wolery and Daveler, 1992; Bourcier, 1994; Bruton and Shaw, 1988; Bruton, 1996) do not adequately cover the conditions and processes to be included in the models. For example, the validated SNF and HLW models (table 4-3) did not include the other WP components (e.g., metal plates) to be included in the internal models. In addition, no examples are given that are comparable to the external models of low-temperature interaction between drift effluent waters and fracture walls. DOE should state whether or not any new analyses will be performed that would support validation under the conditions to be modeled and, if not, how model confidence will be improved.

4-10. Provide more information on the validity or conservatism of geochemical parameters to be used in EQ3/6 models.

As acknowledged in the report, there are large uncertainties in thermodynamic and kinetic data used by EQ3/6. The report states that a range of reaction rate values will be used so that conservative cases may be identified. This analysis should take account of any synergistic effects of varying rates for the numerous solid phases involved in this complex system. Such analysis should also be applied to thermodynamic data, particularly with regard to actinide phases. (Note, for example, that much uncertainty exists regarding appropriate thermodynamic data for U and Pu phases.) Only in this way can the model results be interpreted with confidence.

References

Bourcier, W.L. 1994. *Critical Review of Glass Performance Modeling.* ANL9417. Argonne, IL: Argonne National Laboratory.

Bruton, C.J. 1996. *Near-Field and Altered-Zone Environment Report, Volume II.* Livermore, CA: Lawrence Livermore National Laboratory.

Bruton, C.J., and H.F. Shaw. 1988. *Geochemical Simulation of Reaction Between Spent Fuel Waste Form and J-13 Water at 25 °C and 90 °C.* ISBN 0931837–80–4. Pittsburgh, PA: Materials Research Society.

Wolery, T.J., and S.A. Daveler. 1992. *EQ6, a Computer Program for Reaction Path Modeling of Aqueous Geochemical Systems: Theoretical Manual, User's Guide, and Related Documentation (Version 7.0).* UCRL–MA–110662 PT IV. Livermore, CA: Lawrence Livermore National Laboratory.

Section 4.3.1

4-11. Indicate how correlations between sampled parameters will be identified, quantified, and accounted for in the criticality configuration generation code.

Use of the Monte Carlo method requires that correlations between sampled parameters are taken into consideration if they are not truly independent variables. For example, the drip rate onto the package may affect the WP lifetime. Failure to account for these correlations could result in erroneous results.

Section 4.3.2

4-12. Justify the assumption that it is acceptable to only consider the potential for one external criticality for a given realization.

DOE argued that the small probability of a realization yielding a critical configuration obviates the need to analyze the realization for multiple criticalities. This argument is acceptable only if each criticality is an independent event. Since having a single criticality in a realization requires that several sampled parameters are favorable to produce a criticality, additional

criticalities are not independent events and the probability of having multiple criticalities for a single realization may not be small enough to be ignored. Failure to consider the potential for multiple criticalities in a realization may lead to an underestimation of the probability of a criticality event occurring.

4-13. Regarding item A of the External Criticality list on page 4-39, justify the exclusion of water chemical parameters other than pH.

Water chemistry will be greatly altered during WP and WF interaction, and concentrations of other components such as carbonate influence the geochemical behavior of U and Pu.

4-14. Clarify how the path selection process does not constitute an additional, nonconservative reduction in probability for a given configuration.

In item B of the External Criticality list, it is stated that random selection of external pathway is weighted according to probability. Subsequent transport modeling utilizes probability sampling of parameters. It should be made clear that this approach does not constitute redundant application of probability screening of external pathway.

Section 4.3.3

4-15. Regarding item II.C. of section 4.3.3 on the invert configuration generation code (CGC) geochemistry modeling, justify the exclusion of water chemical parameters other than pH in computing solubility dependence.

In the WP CGC, solubility dependence on other species such as carbonate is included (item I.D.). Such dependence—which, for example, is strong for carbonate content in computing U solubility—should also be included in external cases.

4-16. Clarify how matrix-fracture distribution of water below the WP is calculated (items III.B. and III.C.). Reconcile the distinction between fracture and matrix travel times discussed in section 4.3.3 with the attribution of all flow to the fractures apparent from the discussion in section 4.2.3.

The distinction between matrix and fracture flow has profound implications for modeled travel times and water-rock interaction. For example, it is typically assumed that solutes are not sorbed during fracture flow. The distribution of groundwater flow between the fracture and the matrix will strongly affect U and Pu transport because of contrasting sorption and groundwater travel times. U and Pu transport rates and concentrations are central to models of external criticality.

Section 4.4.1.2

4-17. Indicate whether the consequences of the transient criticality will include an assessment of the degradation of the fuel rods due to the heat or pressures generated by a transient criticality.

If the SNF or the cladding was degraded by a short period of extremely high temperature and pressure, during the event, an additional consequence of the criticality event would be to increase the release rate of radionuclides from the fuel because more water can come into contact with the SNF WF. It does not appear that this possibility is addressed in the topical report.

Section 4.4.3.2

4-18. Justify the claim that the maximum reactivity insertion rate is from the settling of iron oxide particles to the bottom of a degraded WP.

A fuel assembly falling due to a seismic event from a position above the water level in the canister into the water would lead to a much faster reactivity insertion, with more severe consequences. Even if the iron oxide particle settling has been shown to be the maximum reactivity insertion rate for spent nuclear fuel, unless this has been shown to be true for all waste types, this statement should not be included in the topical report.

Appendix C EXAMPLE APPLICATION OF THE METHODOLOGY FOR COMMERCIAL SPENT NUCLEAR FUEL

C-1. Provide information on plans for geochemical model validation in this example analysis and the appendix D example analysis (see discussions above on model validation). This information will make clearer the scope and rigor of the validation approach.

Appendix C, Section 1.4.3 Waste Form Degradation Characteristics

C-2. Support the potentially misleading statement (page C-21) that "At sufficiently high temperatures in an oxidizing environment, the fragments will oxidize...." Oxidation of UO_2 does not, in general, require elevated temperatures.