DISPOSAL CRITICALITY ANALYSIS METHODOLOGY FOR FISSILE WASTE FORMS

J. Wesley Davis
Framatome Cogema Fuels

Peter Gottlieb
TRW

John Massari
Framatome Cogema Fuels

Civilian Radioactive Waste Management System, Management and Operating Contractor
1180 Town Center Drive
Las Vegas, NV 89134

Abstract

A general methodology has been developed to evaluate the criticality potential of the various types of waste form (WF) planned for geologic disposal. The waste forms include: commercial spent nuclear fuel (SNF), high level waste, Department of Energy reactor SNF (including highly enriched), mixed oxide SNF using weapons grade plutonium, immobilized plutonium, and naval reactor SNF.

The methodology can be summarized with the following steps, which apply to all waste forms.

1. The possible processes that can lead to criticality are identified and combined in a set of scenarios. Degradation scenarios are comprised of a combination of features, events, and processes, which result in degraded configurations to be evaluated for criticality. These scenarios were developed by a group of experts (representing the disciplines of geology, geochemistry, groundwater hydrology, metal corrosion, and neutronics) during a workshop.
2. The chemical and geochemical processes acting on the waste packages, the waste forms, and any criticality control material are analyzed theoretically, using the recognized geochemical computer code, EQ3/6, to determine the solubilities and concentrations of neutronically significant species released by the degradation of the initial waste forms.
3. Parametric criticality analyses, relying on calculations of k_{eff} , are performed on the potentially critical configurations, using variations to identify minimum amounts of degraded structural components (basket, containers) and neutron absorbers, which must remain mixed with the fissile material to prevent a criticality.

Background

A general methodology has been developed to evaluate the criticality potential of the various types of waste form (WF) planned for geologic disposal (Ref. 1). The waste forms include: commercial spent nuclear fuel (SNF), vitrified high level waste, Department of Energy (DOE) reactor SNF (including highly enriched), mixed oxide SNF using weapons grade plutonium, and immobilized plutonium. The disposal of these waste forms will be in a container with sufficiently thick corrosion-resistant barriers to prevent water penetration for upwards of several thousand years. The primary criticality control for SNF is neutron absorber incorporated into the basket holding the individual assemblies; for some SNF the absorber material may be carried in control rods inserted in the assemblies. A secondary control is metal oxide, from basket metal or specially added shot, which acts by displacing moderator (generally water). For the immobilized plutonium, the neutron absorber is incorporated into the waste form itself.

The disposal criticality analysis methodology includes analyzing the geochemical and physical processes that can breach the waste package and affect the waste forms inside. In order for a criticality to be possible, such processes must lead to the separation of the neutron absorber from the fissile material, and also allow the accumulation of water to provide moderation. The fissile material can be separated from the neutron absorber material in three ways: removing the neutron absorber from the waste package (leading to criticality inside the waste package); stratifying the degraded products from the fissile and absorber materials (leading to criticality inside the waste package); or removing the fissile material from the waste package, with subsequent re-concentration (leading to criticality external to the waste package). The period of interest

extends from 10,000 years to hundreds of thousands of years (with the specific time horizon to be determined by regulatory requirement).

The criticality analysis methodology guides the analysis of criticality control features of the waste package design, and demonstrates that the final design meets the licensing requirements for criticality control. The methodology can also be extended to analysis of criticality consequences (primarily increased radionuclide inventory), which will support the total performance assessment for the repository.

Methodology Summary

The methodology can be summarized with the following steps, which apply to all waste forms.

1. The possible processes that can lead to criticality are identified and combined in a preliminary set of scenarios. These scenarios were developed by a group of experts (representing the disciplines of geology, geochemistry, groundwater hydrology, metal corrosion, and neutronics) during a workshop (Ref. 2). The resulting set has been documented, including the values to be used for environmental and material performance parameters, generally specified as ranges or probability distributions (Ref. 3).
2. The chemical and geochemical processes acting on the waste packages, the waste forms, and any criticality control material are analyzed theoretically, using the recognized geochemical computer code, EQ3/6 (Ref. 4), to determine the solubilities and concentrations of neutronically significant species released by the degradation of the initial waste forms. Particular attention is given to estimating the composition and amounts of the following: (1) precipitates inside the waste package that contain neutronically significant material and (2) precipitates or adsorbates in fractures or other voidspace in the repository tuff. If the end results of such scenarios have sufficient fissile material, in the appropriate geometry, with sufficient moderator, and separated from most of the neutron absorber, they can then be designated as potentially critical configurations.
3. Parametric criticality analyses, relying on calculations of k_{eff} , are performed on the potentially critical configurations, using variations to identify minimum amounts of degraded structural components (basket, containers) and neutron absorbers, which must remain mixed with the fissile material to prevent a criticality. The values of k_{eff} for specific configurations are

calculated using the Monte Carlo neutral-particle transport code MCNP. When a criticality event is possible, the probability distribution functions of the required conditions or processes are used to calculate the probability of the occurrence of the configuration. The confirmation of a critical configuration ($k_{eff} \geq$ upper subcritical limit) becomes the starting point for design modification(s) to minimize the probability of criticality. Minimizing the potential for criticality is a principal objective of the defense-in-depth policy.

Figure 1 shows how the methodology is applied in the analysis of generic internal and external scenarios. Note that there are three decision criteria (in the lower right corner of the figure) for feeding back to Investigate Design Options for Reducing k_{eff} . These criteria are the following: (1) whether the calculated probability of the critical configuration satisfies a threshold criterion, (2) whether total radionuclide increment from a criticality is sufficiently below the radioactivity that would have existed in the waste form in the absence of any criticality, and (3) whether the expected increase in radioactivity would cause failure of the repository performance objectives. Further details on the methodology, particularly the integration with the design process, are given in Ref. 1.

Standard Degradation Scenarios

Degradation scenarios are comprised of a combination of features, events, and processes (FEPs), that result in degraded configurations to be evaluated for criticality. Features are defined as topographic, stratigraphic, physical, or chemical characteristics of the site that may influence the criticality analysis; examples of features that may effect criticality are faults that may focus or block groundwater flow, or topographic lows in geologic strata that may provide locations where fissionable solutes can accumulate. Processes are physical or chemical interactions that can take place between the emplaced material and the surroundings; examples include groundwater flow, corrosion, precipitation, etc. Events are similar to processes, but have a short duration, and possibly a more extreme intensity or effect on the emplaced material. The event is considered indivisible with respect to time, and is a convenience in relating the standard concept of event to the overall methodology. The following are examples of events: the sudden collapse of a basket due to corrosion of structural members, or rock-fall(s) onto a waste package.

The great variety of possible configurations is best

understood by grouping them into classes. A configuration class is defined as a set of similar configurations whose composition and geometry are defined by specific parameters that may vary over a given range. The purpose of the configuration class concept is to focus the criticality evaluation methodology on the range of configuration parameters that result from a single scenario or set of related scenarios. The configuration classes are specifically defined to cover all possible configurations that can result from physically realizable scenarios. This capability will enable those not familiar with the overall degraded mode criticality to understand and review the results of the methodology.

Scenarios based on those FEPs associated with Yucca Mountain that may affect disposal criticality have been reviewed as part of a workshop on post-closure criticality for the Total System Performance Assessment - Viability Assessment (TSPA-VA) abstraction/testing effort (Ref. 2). The result of this workshop has been the development of a standard set of degradation scenarios that must be considered as part of the disposal criticality analysis of any waste form (Ref. 3). The degradation scenarios are conveniently grouped according to the three general locations for potentially critical degraded configurations: (1) internal to the waste package, (2) external to the waste package in the near-field environment (within the drift), and (3) external to the waste package in the far-field environment (external to the drift).

The internal scenarios initiate with water reaching the vicinity of the waste package. This is the usual requirement to start any degradation process leading to criticality because water is necessary to drive the processes waste package/waste form degradation, separation of fissionable and neutron absorber materials, and transport of fissionable material to the external environment.

Internal Scenarios and Configurations

Tracing the internal degradation scenarios provides information on the classes of internal configurations that result from the effects of degradation events and processes on the internal contents of the waste package. The events and processes that most directly impact the potential for criticality are the following: a) changes to a more optimum geometry, b) retention of moderator, and c) separation of neutron absorbers from fissionable material. Other events and processes are of importance if they are precursors to such direct impact events and processes. For convenience in describing the internal

degradation scenarios and the techniques for analyzing them, the waste package contents are divided into two categories: (1) the fissionable waste form(s) (FWF), and (2) other internal components (OICs). The second category includes various structural, thermal, or neutron absorber components of the intact basket, as well as any co-disposed non-fissionable waste forms.

The degradation of the OICs is an important process with respect to criticality because the degradation products may remain as: (a) insoluble neutron absorbers, (b) insoluble corrosion products that displace moderator, (c) hydrated clayey materials, and (d) solutes affecting the internal chemistry such that the solubility and/or degradation rates of the FWF and/or OICs are altered. The purpose of this step of the methodology is to identify the internal configuration classes that are applicable to the waste form being evaluated. Additional details necessary to perform criticality analyses for the range of configurations in each class (e.g., the condition of the FWF, the amount of moderator, and the amount, composition, and physical distribution of the remaining FWF and OIC corrosion products) will be determined as part of the internal degradation analysis for each specific WF/WP combination.

The internal degradation scenarios branch into six general groups according to aspects of two processes: 1) whether there is accumulation of water within the waste package (two alternatives), and 2) the relative rates of the degradation processes affecting the FWF and the OICs (three alternatives). A minimum accumulation of water is important because nearly all the waste forms will not be capable of criticality without moderation, and water is the most effective form of moderator. Relative degradation rates of FWF and OIC are important because different effects on the geochemistry of the system may result from a different order of degradation, altering the solubility of the corrosion products of the FWF and/or the OICs.

The following is a list of the potentially critical configuration classes, internal to the waste package, together with some discussion of the degradation scenarios that can lead to them.

1. The basket (OIC) is degraded, but the waste form is relatively intact. For criticality, the following additional conditions are required: (1) sufficient moderator (e.g., standing water in the waste package), (2) neutron absorber flushed from the waste package, and (3) most of the fissionable material remaining in the package. This

configuration arises from scenarios in which the basket containing the neutron absorber degrades before the waste form. Such a situation may arise with the current waste package design for commercial SNF.

2. Both basket and waste form are degraded with the same three additional conditions (water, absorber removal, and fissionable material remaining) as configuration #1, above. In general, this configuration will have the fissionable material collected at the bottom of the container (waste package or canister). Since both FWF and OIC are fully degraded, it does not matter which one degrades first.
3. The fissionable material from the waste form is mobilized and moved away from the neutron absorber, which remains in the largely intact basket. As with configuration #2, above, the fissionable material will be most likely accumulated at the bottom of the container, but, unlike the configuration #2, the physical opportunities for this transport/accumulation are limited because the basket is still largely intact. This configuration results from a scenario group that involves the FWF degrading at a faster rate than the basket (OIC). An alternative configuration, with these relative degradation rates, has the fissionable component of the FWF not moving significantly upon degradation. This configuration is included only for completeness, since it does not differ significantly from the intact configuration. These configurations have been analyzed for the aluminum clad research reactor SNF (Ref. 5).
4. Fissionable material is accumulated at the bottom of the container, with the moderator provided by water either trapped in clay or by hydration of metal corrosion products, so that criticality can occur without standing water in the waste package. A variation of these configurations does not have the neutron absorber flushed from the waste package, but only a relative displacement between fissionable material at the bottom of the container and neutron absorber distributed throughout the container. These configurations can result from scenario groups that have penetrations in the bottom of the waste package, preventing standing water in the waste package. This flow-through removes soluble products, but leaves the insoluble ones. In all these scenarios a path representing removal of fissionable material from the waste

package through holes in the bottom provides a source term for the external criticality scenarios. These configurations have been analyzed for the aluminum-clad research reactor SNF (Ref. 5).

5. As with configuration #4 above, the moderator is provided by water trapped in clay, but in this case the fissionable material is distributed throughout a major fraction of the container volume. This configuration class can only be reached if the FWF degrades faster than the OIC, so that the fissionable material remains in place to be locked in by its own hydration or by the hydration of OICs. This configuration has been analyzed for the aluminum-clad research reactor SNF (Ref. 5).

External Scenarios and Configurations

The scenarios leading to near-field configuration classes begin with the source term (the outflow from the waste package bearing fissile material). This source term includes any fissionable material from the waste package in a form (either as solutes, colloids, or slurry of fine particulates) that can be transported into or over the concrete/crushed tuff invert beneath the waste packages.

The external criticality configuration classes are listed below in approximate order of priority for current evaluation, with near-field and far-field intermixed.

1. Accumulation, by chemical reduction, of fissionable material by a mass of organic material (reducing zone) located in the unsaturated zone: (1) beneath the repository, (2) at a narrowing or pinchout of the tuff aquifer, and (3) at the surface outfall of the saturated zone flow, e.g., Franklin Lake Playa. The probability of a flow bearing fissionable material encountering such a mass is lower still. Alternatively, accumulation of fissionable material may occur by chemical reduction of fissionable material by a mass of organic material (reducing zone) located in the unsaturated zone beneath the repository, at a narrowing or pinchout of the tuff aquifer, or at the surface outfall of the saturated zone flow (e.g., Franklin Lake Playa). The initial assessment is that these are all of very low probability.
2. Accumulation, by sorption, onto clay or zeolite.
3. Precipitation of fissionable material in fractures and other voidspace of the near-field, either from adsorption or from a reducing reaction. The two

- configurations are considered together because they are both limited by the same buildup of non-fissionable deposits in the fractures of the near-field.
4. Accumulation of fissionable material in a standing water pond in the drift. This scenario involves waste packages which may not have been directly subjected to dripping water, but are located in a local depression such that a small amount of water flowing from other dripping sites may collect around the bottom of the package during periods of high flow. Since there is no mechanism for completely sealing the fractures in the bottom of the drift, such a pond would be expected to occur only within a short time (weeks or less) following an unlikely-high infiltration episode.
 5. Accumulation by processes involving the formation, transport, and eventual breakup (or precipitation) of fissionable material containing colloidal particles. It has been suggested that the colloid forming tendency of plutonium will enhance its transport capability, providing the potential for accumulation at some significant distance from the waste package.
 6. Accumulation at the low point of the drift/repository. The scenario leading to this configuration must have a mechanism for sealing the fractures in the drift floor so that the effluent from individual waste packages can flow to, and accumulate at, a low point in the drift or repository, possibly in combination with effluent from other waste packages. This is the principal configuration which could accumulate fissionable material from several waste packages. In order to minimize the probability of such an accumulation, the repository underground design team is considering a zero slope for the emplacement drifts, which differs from the 0.5% typical of many mines (typically beneath the water table).
 7. Fissionable material (uranium) accumulated by precipitation, in the saturated zone, at the contact between the waste package plume and a hypothetical upwelling reducing fluid or a redox front. This configuration is considered unimportant because there is no evidence for any such bodies below Yucca Mountain that would have sufficiently different chemical or redox characteristics to significantly concentrate fissionable material from the contaminant plume.
 8. Accumulation at the surface of the invert (or whatever it has degraded into) due to filtration by the degradation products, or remnants, of the waste package and its contents. The fissionable material may be carried as a slurry or colloid. The first case is considered unlikely because the solution moves too slowly to carry a slurry any significant distance.
 9. Accumulation/precipitation from encountering perched water having significantly different chemistry from the fissionable carrier plume. It is likely to be significant only during the very early stages of plume contact with the perched water, before the plume changes the perched water chemistry to its own. With such a limited duration, it is not likely to accumulate much fissionable material.
 10. Accumulation/precipitation from the chemistry changes made possible by carrier plume interaction with the surrounding rock. The amount of material that could be precipitated in this manner is limited by the fact that chemistry changes in the carrier plume itself would precipitate non-fissionable material from the carrier plume before any precipitation of fissionable material from the waste package plume. The result would be fracture filling with non-fissionable material, as configuration #3, above.
- Unlike the configuration classes associated with internal degradation scenarios, all of the near-field and far-field configuration classes are applicable to all waste forms, although the variation in source term may preclude criticality for certain waste forms for certain configurations. However, since the configurations associated with these scenarios are less dependent on the original waste form and waste package design, analyses that bound multiple (or all) waste forms may be used to quickly eliminate some scenarios.

Conclusion

The disposal criticality analysis methodology described here has been applied to possible configurations in the degraded waste package and some potentially critical external configurations. The results have shown that if the standard scenarios and configuration classes are properly defined and analyzed, it is possible to organize the criticality scenarios into a manageable set which can be easily evaluated according to the expected range of SNF characteristics.

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References

1. *Disposal Criticality Analysis Methodology Technical Report*, DI Number: B00000000-01717-5705-00020 REV 01, CRWMS M&O, Sep. 1997.
2. *Criticality Abstraction/Testing Workshop Results*, DI #:B00000000-01717-2200-000187 REV 00, CRWMS M&O.
3. *Construction of Scenarios for Nuclear Criticality at the Potential Repository at Yucca Mountain, Nevada*, DI #:B00000000-01717-2200-000194 REV 00, CRWMS M&O.
4. Wolery, T. J., EQ3/6, *A Software Package for Geochemical Modeling of Aqueous Systems: Package Overview and Installation Guide (Version 7)*, UCRL-MA-110662 PT I, September 1992.
5. *Evaluation of Codisposal Viability for Aluminum-Clad DOE-Owned Spent Fuel: Phase II Degraded Codisposal Waste Package Internal Criticality*, DI#:BBA000000-01717-5705-0017 REV 01

Figure 1. Disposal criticality analysis methodology.