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Assessment of the potential for criticality in the far field of a used nuclear fuel repository

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ABSTRACT

This study aims to assess the likelihood for criticality in the far field of a repository for direct disposal of commercial light water reactor used nuclear fuel. Two models are used in combination: (1) a neutronics model to estimate the minimum critical masses of spherical, water-saturated depositions of fissile material; (2) a transport model to simulate the dissolution of waste packages arranged in an array and the subsequent transport of fissile solutes through fractured bedrock to a single accumulation location. The neutronics model shows that heavy metals from different types of used fuel present the same minimum critical mass behavior in the parameter space of initial enrichment and burnup, dictated largely by the fissile content. However, the magnitude of the minimum critical mass varies significantly within that parameter space, and secondary effects like the presence of absorbing nuclides play a minor role. The transport model employs various subsurface transport scenarios, and for each scenario the mass of each isotope and the overall fissile content of the accumulation is reported from the time of canister failure up to one hundred million years at various distances from the repository edge. Taking the results of the two models in concert, it is shown that even if the accumulation of a critical mass is possible under conservative conditions, these conditions are unlikely to be present in the vicinity of a carefully engineered repository. Based on the results of each model, recommendations for risk mitigation in terms of waste characteristics and repository design are given.

1. Introduction

Underground criticality is a plausible phenomenon resulting from the transport and accumulation of fissile radionuclides from extensively compromised waste packages in a repository for used nuclear fuel (UNF). If a deposition of fissile material in the repository far field attains the appropriate moderating conditions from the host rock and groundwater, positive reactivity feedback mechanisms can, in theory, lead to rapid fission power generation that can compromise the natural barrier. Although highly radioactive fission products are otherwise well-contained in the engineered and natural barriers, this rapid energy release (when criticality is autocatalytic) may, in theory, serve as a direct exposure pathway for fission products to the biosphere (Kastenberg et al., 1996). Even if a sustained criticality event is non-autocatalytic, fission products will be generated that can reach the biosphere; therefore, a criticality safety analysis (CSA) is necessary to increase confidence that a repository and the surrounding geologic formations remain subcritical at any time. As a part of a CSA, scenarios are developed to identify the features, events, and processes (FEPs) describing geologic behavior that could result in the accumulation of fissile material away from the repository. Once a site is selected, location-specific scenarios can be screened by some engineering measures based on geologic characteristics and on-site measurements. Those scenarios where criticality cannot be excluded should be further investigated to assess repository performance and fissile material accumulation (Ahn, 2006). An example of this approach is the detailed scenario and transport model development and analysis for the Yucca Mountain Repository (YMR) (Scaglione and Wagner, 2010; Barnard et al., 1997).

In the absence of site-specific information, the generic conditions under which criticality is achievable should be investigated

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Abbreviations: BWR, Boiling water reactor; CSA, Criticality safety assessment; FEPs, Features, events, and processes; HLW, High-level waste; HM, Heavy metal (uranium plus plutonium); HMVF, Heavy metal volume fraction; LWR, Light water reactor; MOX, Mixed oxide fuel; PWR, Pressurized water reactor; TTB, Transport-to-Biosphere code; UNF, Used nuclear fuel; UOX, Uranium oxide fuel; VVF, Void volume fraction; YMR, Yucca Mountain Repository.

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first and the geologic processes required for such conditions to be met can be examined in further studies (Ahn, 2006). This approach allows for the establishment of a scientific basis for future CSAs and engineering-informed decisions when repository site, design, and geologic conditions are known.

In a hypothetical CSA, neutronic analyses are first utilized to study the conditions required to achieve criticality in the far field of a geologic repository. The minimum critical mass is then used as a screening criterion: if a minimum critical mass of a deposition of fissile material in the far field is larger than the entire inventory of the repository, the deposition cannot become critical. Thus, the FEPs required for a critical formation can be eliminated from further consideration (Ahn, 2006). If instead criticality is conceivable, and a critical mass less than the repository inventory can be achieved, deterministic transport models are used to produce quantitative estimates of fissile mass accumulation in the far field of a geologic repository. Subject to the assumptions and conditions employed in the models, the comparison between minimum critical masses and estimated total accumulation allows for an informed juxtaposition needed to assess repository performance in the criticality safety context.

Previous work in this area has focused on detailed analysis of criticality risk from a single source term. Excess weapons-grade plutonium, used naval reactor fuel, and most recently, the damaged fuel from the Fukushima Daichii nuclear reactors in Japan have been of particular interest due to the higher fissile content and thus smaller critical masses (Kastenberg et al., 1996; Liu et al., 2014; Greenspan et al., 1997; Vujic and Greenspan, 1998). This study, instead, focuses on the direct disposal of UNF discharged from commercial light water reactors (LWR)—either from uranium dioxide or uranium-plutonium mixed oxide (MOX) fuel—as it represents a larger quantity of fissile material with wide variety of compositions derived from the type of reactor the fuel is used in, the initial enrichment and burnup, and other factors. The scope of this paper is to determine if criticality can be excluded or not for the far field of an LWR UNF repository by (1) determining the minimum mass of fissile material necessary to achieve criticality in a deposition in the repository far field as a function of the UNF composition, and (2) estimating the mass that could be accumulated in the far field as a consequence of the transport of radionuclides through the host rock.

Section 2 of this manuscript describes the neutronics model and reports the estimates for the minimum critical mass of the deposition as a function of UNF composition. Section 3 describes the transport model and illustrates results obtained for representative compositions of used fuel in terms of the mass of fissile material accumulation in the far field. Section 4 takes the results of the criticality analysis and of the transport analysis in concert and presents an integrated discussion on far-field criticality risk from the direct disposal of commercial LWR UNF.

2. Minimum critical mass in a far-field deposition

2.1. Postulated scenario for fissile material accumulation

The minimum critical mass is determined for a fictitious deposition of fissile material located in the far field of a hypothetical, water-saturated geologic repository. It is postulated that at some time after repository closure, groundwater will corrode and infiltrate the canisters, gradually dissolving the used fuel and transporting it beyond the vicinity of the canister. From this point, the radionuclides in the used fuel will be carried by groundwater into the geologic formations surrounding the repository and eventually precipitate to form a deposition. After canister failure, uranium and plutonium may become separated from neutron absorbing materials included in the canister or present in the UNF itself because the transport behavior of uranium and plutonium typically differ from that of neutron absorbing materials. Thereafter, they can intermix with neutron moderating materials such as rock and water.

In oxidizing environments, uranium is mobilized in the hexavalent state and can be transported away from the repository, whereas plutonium will likely precipitate in the vicinity of the canister given its very low solubility and the strong sorption with rock (Ahn, 1997). Over time, however, plutonium isotopes (in particular, Pu-239 with a half-life of 24,100 years) will decay to uranium isotopes and be transported along a similar pathway, adding to the fissile content of the radionuclide plume. Unless the concentration or composition of the plume is significantly altered—for example, by a high degree of dispersion or a criticality event during transport—it may become subject to localized geochemical environments that favor tetravalent uranium, where the resulting precipitate may continue to grow until a critical mass is reached. This study did not consider the possibility that uranium and plutonium might accumulate and form critical masses separately from each other.

2.2. Neutronics model

A neutronics analysis is performed to determine the minimum mass required to achieve criticality in a far-field deposition. It is conservatively assumed that all of the uranium in the repository feeds into a single, water-saturated deposition. Any plutonium that has not yet decayed to uranium isotopes at the time of analysis is lumped into the deposition to ensure that all fissile contributions are included. Plutonium and uranium are assumed to precipitate uniformly as oxides and will be collectively referred to as heavy metal (HM). The repository and surrounding geologic formations are assumed to be water-saturated.

The computational model represents the deposition as a spherical, homogenous mixture of heavy metal, host rock, and groundwater in variable ratios. A 1-m thick layer of water-saturated host rock is assumed surrounding the deposition to account for neutron reflection. The host rock for the neutronics model is assumed to be sandstone (grain density 2.71 g/cm^3) with composition given in Table 1. From the neutronics perspective, sandstone is conservative due to low concentrations of neutron-absorbing elements and minerals. Later, in Section 3, the model for radionuclide transport through granite, a common repository host rock, is described.

The temperature is assumed constant at 20℃, although the ambient temperature will certainly be higher due to both the natural geothermal gradient and decay heat. To accurately determine the temperature of the system, complex analyses are required for heat and mass transfer based on site-specific information

Table 1
Composition of sandstone host rock in the far-field deposition (Liu et al., 2014).

Component	Weight fraction (%)
SiO_2	78.70
TiO_2	0.25
Al_2O_3	4.80
Fe_2O_3	1.10
FeO	0.30
MnO	0.03
MgO	1.20
CaO	5.50
Na_2O	0.45
K_2O	1.30
H_2O	1.30
P_2O_5	0.08
CO_2	5.00