CRITICALITY ISSUES WITH HIGHLY ENRICHED FUELS IN A REPOSITORY ENVIRONMENT

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ABSTRACT

This paper presents preliminary analysis of a volcanic tuff repository containing a combination of low-enrichment commercial spent nuclear fuels (SNF) and DOE-owned SNF packages. These SNFs were analyzed with respect to their "criticality risks".

Disposal of SNF packages containing significant fissile mass within a geologic repository must comply with current regulations relative to criticality safety during transportation and handling within operational facilities.[1] However, once the repository is closed, the double contingency credits for criticality safety are subject to unremediable degradation, (e.g., water intrusion, continued presence of neutron absorbers in proximity to fissile material, and fissile material reconfiguration).

The work presented in this paper focused on two attributes of criticality in a volcanic tuff repository for near-field and far-field scenarios: (1) scenario conditions necessary to have a criticality, and (2) consequences of a nuclear excursion that are components of risk. All criticality consequences are dependent upon eventual water intrusion into the repository and subsequent breach of the disposal package. Key criticality parameters necessary for a critical assembly are (1) adequate thermal fissile mass, (2) adequate concentration of fissile material, (3) separation of neutron poison from fissile materials, and (4) sufficient neutron moderation (expressed in units of moderator to fissile atom ratios). Key results from this study indicated that the total energies released during a single excursion are minimal (comparable to those released in previous solution accidents), and the maximum frequency of occurrence is bounded by the saturation and temperature recycle times, thus resulting in small criticality risks.

INTRODUCTION

Regardless of the double contingency criteria established for any given spent nuclear fuels (SNF) package at loading (e.g., fissile limits, neutron "poisons," fuel spacing within a package, "dryness," etc., in a geologic time frame), one or more of any designed contingencies can be expected to be compromised. Prior studies have addressed the potential failure of one or more SNF packages that contain fission products and actinides within the 10,000-year isolation stipulated for the repository.[2,3]

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DISCLAIMER

"Criticality" in a performance assessment of a geologic repository corresponds to the risks resulting from a nuclear criticality excursion (where risk is the combined effect of consequences and probability). A wet environmental criticality generally requires water to breach a waste package, promote the necessary "reconfiguration" of package contents, and provide the necessary moderation/reflection needed to allow neutron propagation. An example of a criticality in a SNF package would be a top-breach of the waste package with long-term water drip. The breaching of the package may result in a criticality in one of several scenarios: internal (in situ), near-field (within several tunnel diameters of the waste package), or far-field (a significant distance away from the package).

THEORY

Eventually, all issues related to any possible occurrence of a criticality in a post-closure time frame of the repository rely heavily on the presence of water. Fissile loads distributed throughout the various DOE SNF canisters containing enriched material have no identified, credible events that cause fuel reconfiguration leading to criticality inside or outside the package without the presence of water. Water entering a breached waste package results in the addition of both reflection and moderation of the waste package contents as well as the necessary mechanism to provide reconfiguration of the fissile material, or separation from any neutron absorber within the package.

Reactivity features of ^{235}U, ^{239}Pu, and ^{233}U differ slightly in their nuclear resonances and fissioning for thermal neutrons. Of the three fissile isotopes ^{235}U, in enriched defense fuel canisters, dominates the risk for criticality. ^{239}Pu does not pose a significant issue for criticality potential because the fissile atom density is too low even in densely packed SNF canisters containing N-reactor fuels. ^{233}U has significant mass quantities in only one fuel category, however, its atom density is much less than that for ^{235}U and it is not considered significant.

Given the fact that any criticality experienced in the repository will require the presence of water, models that incorporate the physics associated with water-moderated critical systems should predict the characteristic behavior of the criticality. Doppler temperature coefficients, water density/saturation changes, and resonance behavior of the fissile materials with increasing temperatures all suggest that a natural shutdown mechanism exists for any criticality that might occur. The rate of water influx, rate of neutron production, and the thermal properties of the repository would govern initiation of any criticality. System shutdown would similarly be governed by the rate of heat generation (negative temperature coefficient) and desaturation based on water being driven out of the system.

RESULTS

During scenario development for criticality in a repository, an infinite number of configurations can be identified that result in a critical assembly. In this study, a systematic approach was used where different combinations of fissile mass and fissile concentrations were investigated for near-field and far-field criticality.

Near-field criticalities are defined as those events that occur outside, but in proximity to the disposal package (within a few tunnel diameters of the package). A totally degraded outer package would be expected to cause significant accumulation of iron oxide on the floor of the drift. Intermittent groundwater flow across exposed fissile material in a breached package would then create incremental fissile material additions to any rust or concrete material encountered on the drift floor. Alternatively, fissile accumulation might occur from a gross breach in the bottom of the package where partially intact fuel shards could drop onto the drift floor. In either case, the ability to maintain adequate water in proximity to effect a critical system is certainly problematic. Neither chemically bound water nor hydrated compounds can provide the atom density of hydrogen necessary to significantly affect neutron moderation in such an "open" system.

To evaluate the consequences of criticality in a moderated system, this study used an uncoupled far-field thermal-hydraulics (THX) conceptual model and a set of assumed conditions. These conditions were a mixture of thermal fissile material (TFM), water, and tuff. The uncoupled calculations were required to determine the time history of the water saturation given the groundwater transport through interconnected pores and fractures of the host rock. This flow is indicated by changes in fluid density and/or vaporizing the liquid phase because of heat generated in a criticality. BRAGFLO_T (X1.00), a multiphase fluid and energy simulator, predicted the uncoupled water saturation and (moderator) temperature changes resulting from a single critical event of an assumed system size (energy release) and power output. A total of 45 uncoupled calculations examined a range of initial saturation (65, 75, and 85 percent), and heat-generating-zone radii of 0.5, 1.0, and 1.5 m. The size of the modeled critical events ranged from 2.0×10^{18} to 1.0×10^{20} total fissions per event. From these calculations, both the average saturation change and average temperature history changes were used to determine the recycle (or reflux) times. These recycle times are simply the periods of time for the change in average saturation and temperatures to return to near initial conditions. Saturation recovery times generally ranged from 150 to 10,000 days, and temperature recovery times ranged from 12 to 110 days.

The dynamic system feedback for criticality comprises several major components consisting of prompt shutdown mechanisms. Because the temperature recycle time is shorter than the saturation recycle time, the prompt shutdown mechanism dominates the feedback processes.

In support of these criticality calculations, Sandia National Laboratories (SNL) developed a NARK (NucleAR Kinetics and dynamics) code that uses self-adaptive ordinary differential equation (ODE) solution algorithms to solve sets of "stiff" or "nonstiff" coupled ODEs common to nuclear kinetics and dynamics. These calculations were uncoupled from the THX calculations; hence, they were denoted as uncoupled nuclear dynamics (UDX) calculations. The subsequent calculations were performed for a range of thermal feedback coefficients. The goal of these calculations was to demonstrate that prompt negative coefficients dominate and control the shutdown mechanisms of a

moderated system that might develop in near-field or far-field scenarios. The values used in these calculations are identified in Table 1.

Table 1: NARK UDX Sensitivity Input Parameters

NARK Sensitivity Input Parameter	Units	Sensitivity Input Parameters
TFM Mass	kg	25.0, 50.0, 100.0
TFM Volume Faction	dimensionless	262.155×10^{-6}
Effective Neutron Lifetime, l	s	1.0×10^{-4}, 5.0×10^{-4}
Excess Reactivity Insertion, $\rho_o(\$)$	$\$$	0.1, 1.0, 10.0
Initial Power, N_o	w	0.001, 0.01, 10.0
Fuel-Doppler Temperature Coefficient, α^T_F	$\Delta k_{eff}/k_{eff}/K$	-7.5×10^{-3}, -10^{-4}, -2.5×10^{-4}

Several uncoupled NARK calculations were run to calculate power excursions (rise and fall above initial power condition). The calculations also yielded total fissions that ranged from 2.0×10^{18} to 1.5×10^{20}. These values are similar to documented criticality accidents in aqueous systems.[7]

The output from the UDX calculations revealed how fast power excursions shut down because of prompt feedback mechanisms. UDX results yielded a range of integrated fissions from 1.30×10^{16} to 5.54×10^{19}, and excursion durations ranged from 3.17×10^3 to 3.46×10^5 seconds. Selected time behavior results are presented in Figure 3.

Preliminary static calculations using the MCNP code (via a pre-processor "RKeff") revealed that a critical state (i.e., $k_{eff} = 1.0$) could be achieved at a concentration of 5.0 kg/m^3 (i.e., concentration = TFM mass/volume of the mixture [TFM + rock + water]) for Topopah Springs welded tuff (saturation = 100%, porosity = 13.9%). Therefore, the static criticality concentration was incorporated into the uncoupled dynamic code calculations by varying the TFM mass consistently with the concentration of 5.0 kg/m^3. This constraint, using TFM mass values (25, 50, 100 kg) to bound the UDX calculations, resulted in radii of 1.0067, 1.3365, and 1.6839 m, respectively. The uncoupled nuclear dynamic calculations showed that excursions resulted in small quantities of release energy and additional fission yield products. To achieve a criticality in a far-field location, multiple HEU package failures (interspersed with commercial packages) and transport to a common accumulation point are necessary to accumulate a fissile mass needed to support a criticality. Similar criteria may exist for accumulated TFM both in situ (inside the breached package) and near-field. The latter situation is depicted in Figure 1. This figure shows what combinations of fissile mass and fissile concentrations are necessary to reach a critical assembly. Fissile mass would be that coming from either a single package or a combination of multiple SNF package failures.

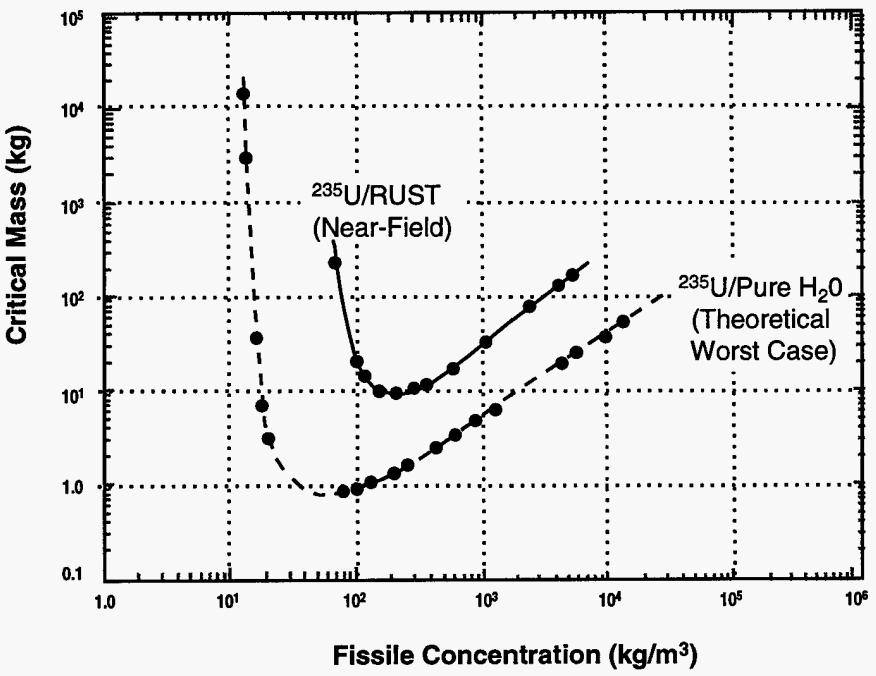

Figure 1. S-Curve for pure ^{235}U (ideal cases)

The net integrated fissions from a criticality were a simple function of a combination of initial reactivity, TFM mass, and the prompt feedback temperature coefficient. This relationship is depicted in Figure 2. The nuclear dynamics scaling parameter $(\rho_o m_{TFM}/|\alpha_T|)$, where

ρ_o = initial reactivity insertion
m_{TFM} = thermal fissile mass
$|\alpha_T|$ = thermal feedback coefficient

versus the maximum number of integrated fission displays as a linear trend in log-log space.

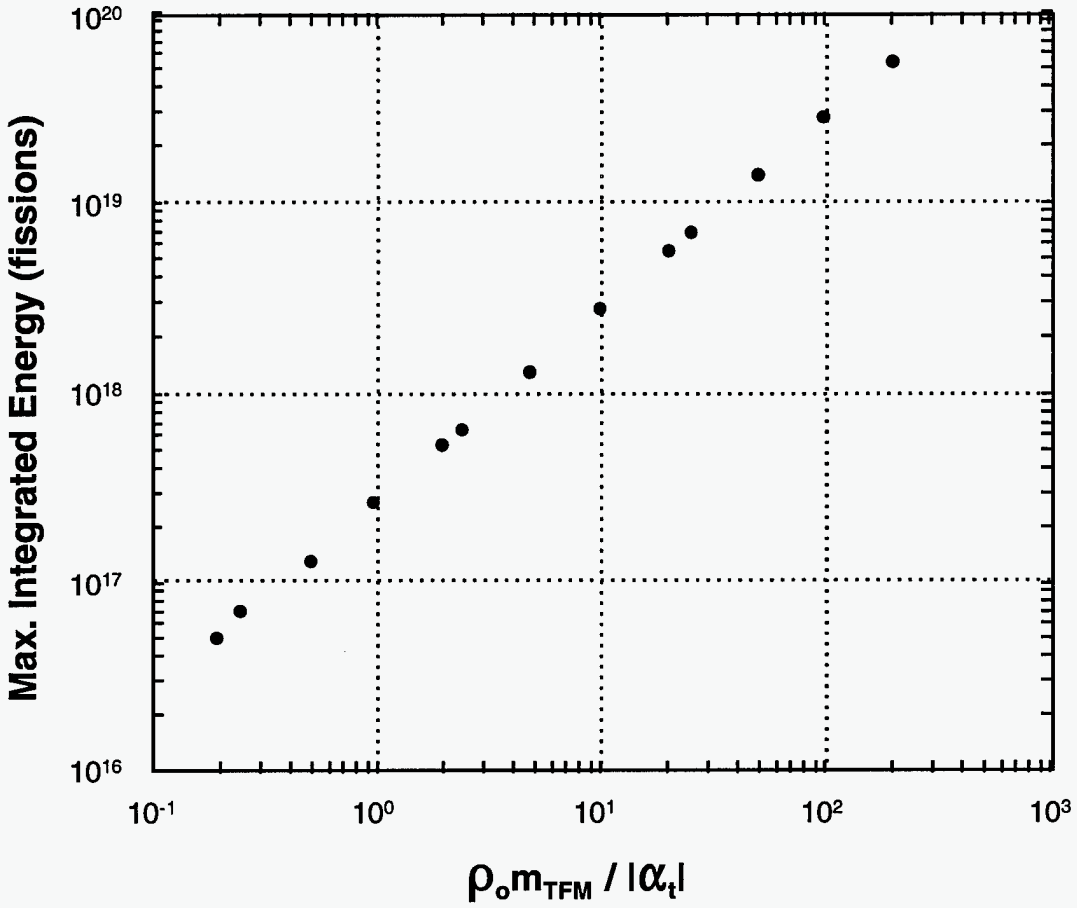

Figure 2. Integrated Energy

Associated with any SNF package, including commercial fuels, will be the potential presence of neutron "poisons." Neutron poisons may be introduced to ensure subcriticality for any package in transport or after emplacement and prior to repository closure as part of the double contingencies associated with enriched fuels in operating facilities. The established concentration levels of any neutron absorber material will probably be predicated on the required retention time within the 10,000-year life of the repository. Retention in this case is defined as the ability to provide some degree of assurance that adequate neutron absorber material will remain within proximity of any fissile material. Differential separation because of differing solubilities can result in movement of one material away from another that might allow formation of a critical mass. Such a separation can occur both within a breached package and in the geologic setting. Materials under consideration for inclusion in SNF packages for the purpose of criticality control include boron within a borated stainless steel matrix, hafnium, cadmium (where it is already integral to the fuel assembly), and, perhaps, gadolinium in an amorphous metal-glass composite.

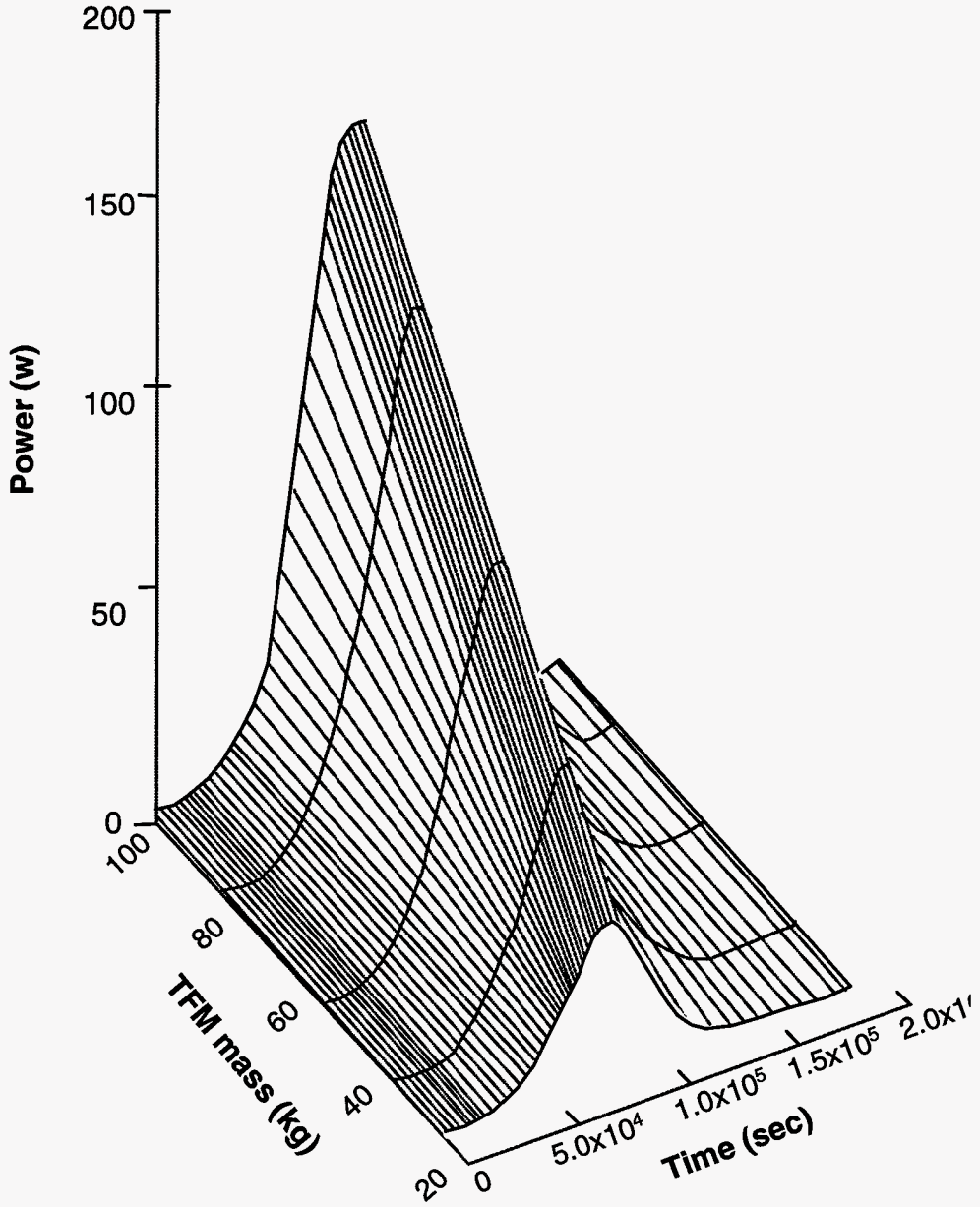

Figure 3. Select UDX results

CONCLUSIONS

Significant technical findings from this study showed that large fissile concentration values on the order of 70 kg/m^3 are necessary to yield a criticality assembly (Figure 1). In addition, should a criticality (nuclear excursion) occur, the total fissions would be very small (2.0×10^{18} to 1.5×10^{20}) and are comparable to values seen in the literature for previous aqueous criticality accidents.

In situ criticalities pose the highest probability of a criticality for any enriched package because they represent the greatest TFM concentration for all time within the repository. In addition, the in situ event also provides the most significant opportunity for both moderation *and* reflection within a package. It is also possible to study various scenarios that provide the "optimal" hydrogen-uranium atom ratio for a given fuel type or enrichment.

Perhaps fewer than 100 HEU SNF packages containing HEU contribute to a significant risk in the near-field. In this case, the fuel assemblies inside the SNF package would last longer than the disposal package itself because the corrosion rate for the fuel and fuel cladding is slower than that for the package. Water and corrosion will completely destroy the waste package to the point where solid fuel "debris" accumulates on the drift floor. But water may not be present in adequate quantities to either reflect or moderate such a mass or both.

Far-field criticalities are even more problematic, given that both sufficient water and contact time with the contents of a breached waste package are needed to transport TFM. Furthermore, all of the following conditions may be required: (1) no significant TFM retention along the drift floor, (2) multiple HEU package failures that are themselves interspersed with commercial packages, (3) a single collection point with adequate volume and suitable geometry, (4) geochemistry favorable to TFM retention, and (5) retained water quantities adequate to promote a criticality. Mechanisms that promote TFM dilution for transport are unlikely to also favor reconcentration to the degree needed to create a critical system. Additionally, any package failure and transport of uranium with low enrichment to a common point drain will tend to denature any HEU at the common location.

The presence (and retention) of neutron poisons during initial packaging may be necessary to ensure safety of a fully flooded SNF canister with intact fuel assemblies for transportation and handling within operational facilities. However, any failure scenario must necessarily address the degraded package condition, where changes in the spatial atom densities of both the fissile material and neutron poison(s) must be analyzed because of chemical alteration by an aqueous system. Such an analysis would require that a new S-curve (similar to that shown in Figure 1) be generated to incorporate the presence of added neutron poisons to the system.

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