Disposal Criticality Analysis for Aluminum-Based DOE Fuels

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Introduction

This paper describes the disposal criticality analysis for canisters containing aluminum-based Department of Energy fuels from research reactors. Different canisters were designed for disposal of high enriched uranium (HEU) and medium enriched uranium (MEU) fuel. In addition to the standard criticality concerns in storage and transportation, such as flooding, the disposal criticality analysis must consider the degradation of the fuel and components within the waste package. Massachusetts Institute of Technology (MIT) U-Al fuel with 93.5% enriched uranium and Oak Ridge Research Reactor (ORR) U-Si-Al fuel with 21% enriched uranium are representative of the HEU and MEU fuel inventories, respectively. Conceptual canister designs with 64 MIT assemblies (16/layer, 4 layers) or 40 ORR assemblies (10/layer, 4 layers) were developed for these fuel types. Borated stainless steel plates were incorporated into a stainless steel internal basket structure within a 439 mm OD, 15 mm thick XM-19 canister shell[1]. The Codisposal waste package contains 5 HLW canisters (represented by 5 Defense Waste Processing Facility canisters from the Savannah River Site[2]) with the fuel canister placed in the center. Figures 1 and 2 show the waste package after emplacement in the repository.

Approach

The range of environmental parameters, corrosion rates, and failure mechanisms were evaluated to develop degradation scenarios. The chemistry/geochemistry of the system was analyzed using the EQ3/6 program[3] with successive runs linked to simulate water dripping into, and flowing out of, the waste package with time[4]. The typical sequence of degradation starting with the assumption of water dripping on a waste package would be: 1) corrosion and eventual breach of waste package barriers allowing water accumulation; 2) aqueous corrosion of stainless steel HLW and fuel containers; 3) degradation of HLW glass to clay; 4) degradation of Al-based fuel concurrent with or after the HLW glass; and 4) degradation of fuel canister basket materials including criticality control material coupled with absorber flushing from the waste package.

Parametric analyses were run on a range of possible configurations of fuel within the waste package using MCNP[5] to identify the most reactive configurations and determine the minimum amount of neutron absorber required to be distributed with the fuel in its degraded state[6].

Results

The aluminum clad/aluminum matrix fuel would be expected to degrade through oxidation within a few decades of breach of the fuel canister. If the fuel canister were penetrated while the HLW glass were degrading, the chemistry (primarily pH > 10.0) would be such that most of the uranium could dissolve. The uranium concentration could be as high as 10 g/liter but more than 2 g/liter of boron would be present from the degradation of the HLW glass. The minimum critical concentration of HEU is 11.6 g/liter under ideal conditions (no absorbers)[7]; therefore, this scenario would not be a criticality concern inside the waste package.

Should the fuel canister be penetrated after the HLW glass were degraded when the pH would be near neutral, then the uranium would no longer be soluble and would remain in the canister or waste package. Three configurations could result based on level of degradation of the other components and the location of the canister as it degraded within the waste package: 1) degraded (oxidized) homogenized fuel material in intact or degraded basket in the fuel canister; 2) layers of hydrated aluminum, uranium, and iron oxides from the degraded fuel canister above the degraded HLW glass; and 3) degraded products from the fuel mixed with various fractions of the degraded HLW glass. The volume fraction of water in the degraded HLW and fuel, as well as the mass of iron oxide from the degraded canisters and basket, were varied over the likely ranges. As the borated stainless steel in the basket degrades, the borides may be dissolved and carried away in solution[8]. If gadolinium (Gd), which is relatively insoluble, is used in the absorber plates, a significant fraction would remain with the degradation products from the fuel. The bounding geochemistry analysis of this sequence indicates that no more than 1 kg of Gd could be removed from the waste package over a 70,000 year time period.

Analysis of the most reactive degraded fuel mixture in configuration 1 using MCNP indicates that approximately 1 kg of Gd is required to be distributed in the MIT fuel intact canister basket. After degradation of the basket, less than 0.5 kg of Gd would be required. If carbon steel were utilized for the basket fabrication, then less than 0.25 kg of Gd would be required. Stainless steel typically undergoes localized attack (e.g., pitting, crevice corrosion) that is likely to leave some small pieces of uncorroded material that are free to settle to the bottom of the canister and not become uniformly mixed with the degraded fuel. On the other hand, most of the iron oxides from the corrosion of carbon steel will be uniformly distributed with the degraded fuel because carbon steel undergoes general corrosion.

If stainless steel were utilized in the ORR fuel canister, then less than 0.1 kg of Gd would be required to remain in the debris mass; however if carbon steel were utilized with borated stainless steel separator plates between layers of assemblies, then no Gd would be required.

Parametric analysis of configurations 2 and 3 using MCNP indicates less than 0.2 kg of Gd is required to remain with the degraded MIT fuel and no Gd is required for the ORR fuel.

Conclusions

Without the presence of a fairly insoluble neutron absorber, the long-term action of infiltrating water can lead to a small, but significant, probability of criticality for both the HEU and MEU fuels. The MIT fuel (HEU) canister design which has 1.5 kg of Gd distributed throughout the basket will reduce the probability of criticality, during the first several hundred thousand years following emplacement, to virtually zero. The ORR fuel (MEU) canister design which either uses carbon steel for most of the structural basket or has 1.1 kg of Gd distributed in the basket structure will also reduce the probability of criticality to virtually zero.

References

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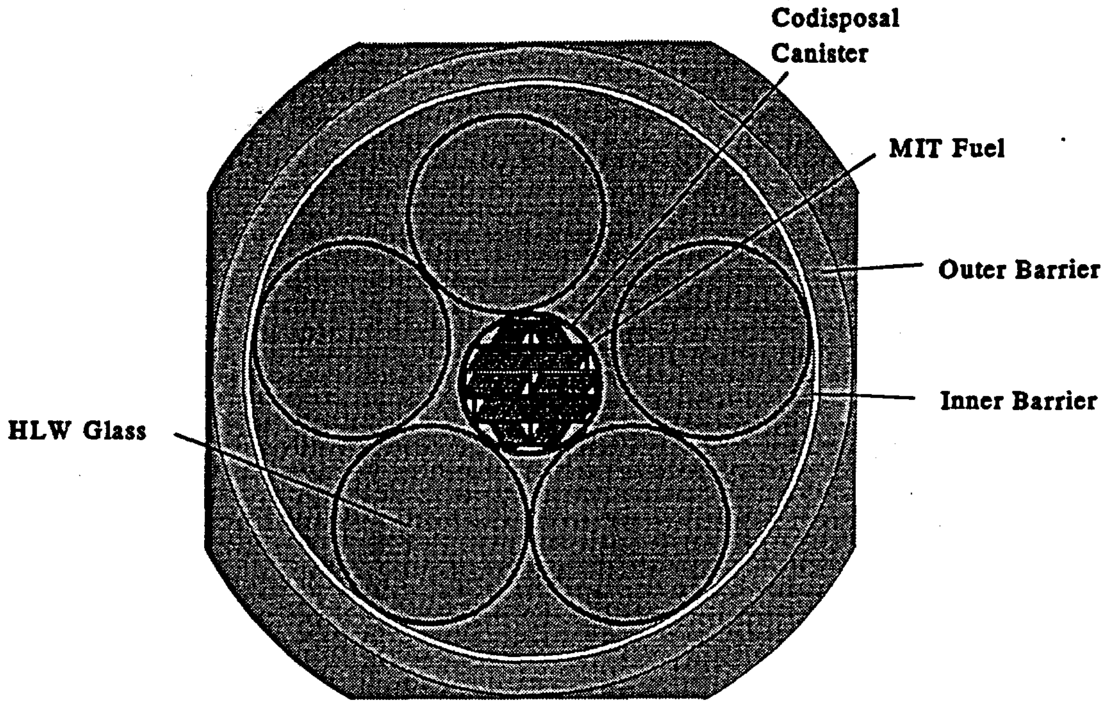

Figure 1. Codisposal Waste Package with MIT SNF Canister

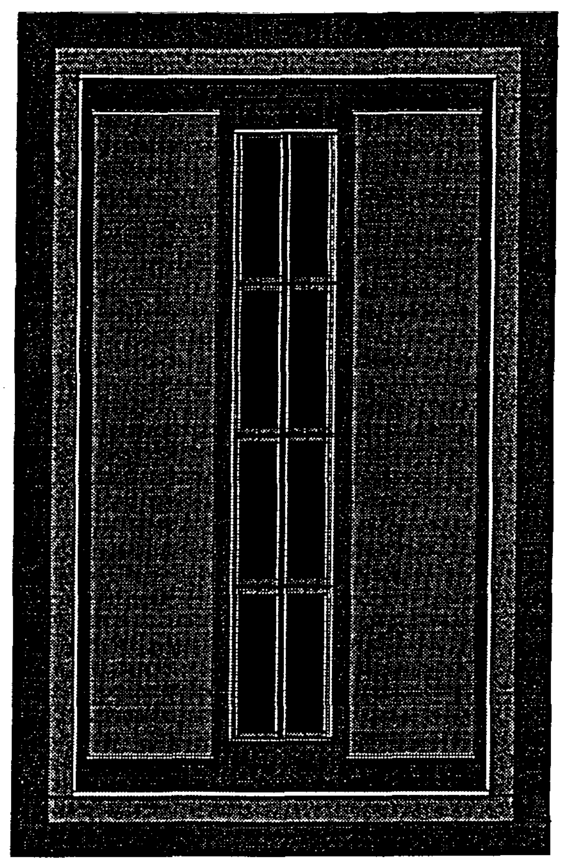

Figure 2. Axial View of the Codisposal Waste Package with MIT SNF Canister