Disposal Criticality Analysis for Immobilized Plutonium: Internal Configurations

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Introduction

Initial concepts of plutonium (Pu) immobilization considered both glass and ceramic waste forms containing 10% ^{239}Pu with an equimolar amount of the neutron absorber Gd as Gd_2O_3 for criticality control in 3 mm thick steel cans, with 20 such cans placed inside a 1 cm thick steel canister of the same size and material planned for high level waste (HLW) glass disposal. The remaining 75% of the canister will be filled with HLW glass. One, or more, such Pu containing canisters is then placed inside a disposal container together with canisters containing only HLW glass. Such a co-disposal waste package (WP) would have a total of 4 or 5 canisters.

Since the disposal criticality control technique has the neutron absorbing elements contained in the waste form itself, both the fissile and the absorber are available for removal from the waste package as the waste form degrades and releases them.

Therefore, the balance between the amounts of fissile and absorber remaining in the waste package must be tracked.

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Approach

The analysis for immobilized Pu follows the disposal criticality analysis methodology¹.

In this study² the focus is on determining the range of chemical compositions of the configurations which can occur following the aqueous degradation processes, particularly with respect to the concentrations of uranium, plutonium, and the principal neutron absorber, gadolinium. The principal analysis tool is a mass balance program that computes the amounts of plutonium, uranium, gadolinium, and chromium in solution as a function of time with inputs from a range of possible waste form dissolution rates, stainless steel corrosion rates, and compound solubilities for the neutronicallly significant elements.

The configurations having fissile material separated from neutron absorber fall into two general categories: (1) Insoluble waste form alteration products precipitated at the bottom of the waste package in a clayey mass (where the major clay ingredient would be the silica from the HLW glass or contained in the water dripping into the waste package), and (2) Fissile material (plutonium and uranium) precipitated on metal surfaces. The second configuration would put the fissile material into a very thin geometry; scoping calculations have showed it to require an unrealistically large critical mass. Since the degradation processes may be slow compared to the 24,000 year half-life for the decay of ^{239}Pu into ^{235}U , the analysis has been conducted for the range of fissile isotope mixtures as a function of time.

Results

The key parameter in this analysis is found to be the pH of the solution in the waste package. For high pH, which can occur while the highly alkaline HLW glass is degrading, the uranium will be sufficiently soluble that most of it could be flushed from the waste package within a few thousand years. If this occurred after 24,000 years, when most of the Pu has decayed to U, it would preclude internal criticality. On the other hand, at $\text{pH} < 5.8$ the Gd_2O_3 may have more than 1000 times the solubility of uranium or plutonium. This could result in so much more of the gadolinium being removed (by comparison with any fissile removal) that there is a significant potential for a criticality.

The low pH condition can arise during the corrosion of the steel canisters, which can span the time period from 0 to 60,000 years following breach of the waste package; after that time period, the gadolinium will no longer be soluble. Therefore, if the dissolution of the waste form requires significantly more than 60,000 years, there will be no chance of criticality.

There are two additional mechanisms for limiting the potential for criticality:

- Tracking the balance between removal of fissile and the removal of Gd shows that if the total Pu loading is kept below some limit, the amount of Pu (or U) remaining in the waste package will be less than a critical mass by the time enough Gd has been removed from the waste package to permit criticality. Even the relatively high Gd solubility at low pH cannot prevent this effect.

- The Pu loading limit (to preclude criticality) is larger for the ceramic waste form than for the glass because of the hafnium naturally present in the chosen ceramic formulation, which serves as a backup neutron absorber under those conditions in which the primary absorber, gadolinium, becomes soluble and is removed from the waste package.

Conclusions

For the waste forms and degradation modes considered here, it is possible to preclude the possibility of criticality by maintaining a plutonium loading limit. Since the presence of hafnium is shown to increase this loading limit, the defense-in-depth policy would suggest the maximization of the amount of Hf as a backup criticality control material.

At the end of 1997, after this study was completed, the ceramic waste form was downselected and a new formulation was developed, with the amount of Hf increased to the point where internal criticality may no longer be possible. In addition, recent calculations³ indicate that GdPO_4 is insoluble over a much broader range of pH than is Gd_2O_3 , so that its use as the Gd carrier in the waste form would provide an extra margin of defense-in-depth.

Acknowledgement

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