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Use of Gadolinium as a Primary Criticality Control in Disposing Waste Containing Plutonium at SRS

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INTRODUCTION

Use of gadolinium as a neutron poison has been proposed for disposing of several metric tons of excess plutonium at the Savannah River Site (SRS). The plutonium will first be dissolved in nitric acid in small batches. Gadolinium nitrate will then be added to the plutonium nitrate solution. The Gd-poisoned plutonium solution will be neutralized and transferred to large under-ground tanks. The pH of small batches of neutralized plutonium solution will be adjusted prior to addition of the glass frit for eventual production as glass logs. The use of gadolinium as the neutron poison would minimize the number of glass logs generated from this disposition.

The primary criticality safety concerns regarding the disposal process are: 1) maintaining neutron moderation under all processing conditions since gadolinium has a very large absorption cross section for thermal neutrons, 2) characteristics of plutonium and gadolinium precipitation during the neutralization process, 3) mixing characteristics of the precipitate to ensure that plutonium would remain homogeneously mixed with gadolinium, and 4) potential separation of plutonium and gadolinium during nitric and formic acids addition. A number of experiments [1,2,3] were conducted at the Savannah River National Laboratory to study the behavior of plutonium and gadolinium at various stages of the disposition process.

HISTORICAL PLUTONIUM DISPOSITION

In the past, SRS has dispositioned small quantities of plutonium using iron and manganese as neutron poison. The minimum subcritical absorber-to-plutonium weight ratios (A:Pu) in an infinite system for these neutron absorbers are 160 and 32, respectively [4]. Use of these neutron absorbers results in large quantities of waste that must be processed into glass logs.

EXPERIMENTAL RESULTS

One of the criticality safety concerns is to retain adequate water in waste under all processing conditions to maintain neutron thermalization as gadolinium is extremely effective in capturing thermal neutrons, but its absorption cross section decreases greatly as neutron energy increases.

Bronikowski [1] concluded that plutonium and gadolinium would precipitate together during the neutralization process with 50% NaOH when Gd-poisoned plutonium solutions at Gd:Pu weight ratios of 0.5:1, 1:1, 1.5:1 and 2:1 were neutralized. Concerns were focused on the transient condition during neutralization between a pH of 3, when the plutonium polymerization begins and only 5% of gadolinium precipitates, and a pH of 7, when complete precipitation of plutonium and gadolinium is expected. The experiment demonstrated that this transient is very short (on the order of seconds) under normal operation (i.e., complete precipitation of plutonium and gadolinium at a pH of 7 or greater and no agitator failure). Bronikowski demonstrated that the H:Pu ratio for a precipitate slurry at a pH of 3 is greater than 1900. To determine the H:Pu ratio of a precipitate slurry at a pH of 7 in the transfer line or pump tank where the precipitate is subject to air drying, samples of a precipitate slurry were centrifuged. The H:Pu ratio for the centrifuged precipitate was determined to be 152. According to Figure 1, the Gd:Pu weight ratios of 0.05:1 and 1:1 are sufficient to maintain the precipitate slurry with respective H:Pu ratios of 1900 and 152 subcritical.

During processing, the Gd-poisoned caustic sludge slurry is pumped into a small (~10,000 gallons) sludge receipt and adjustment tank (SRAT) for further processing prior to transfer to the slurry mix evaporator (SME) where the glass frit is added to form a sludge-frit slurry to be sent to the melter feed tank and the melter. In the SRAT, nitric and formic acids are added to the caustic sludge to adjust its chemistry. A series of tests were performed in the presence of major sludge components (e.g., iron, calcium, aluminum, nickel, manganese, and magnesium) and one test in the absence of these components to investigate the possibility of the separation of the plutonium and gadolinium during processing in the SRAT [2,3]. The results indicated that, in the absence of major sludge components, the gadolinium completely dissolved at a pH of 3.5 while 90% of the plutonium remained insoluble. This behavior is presumably due to the higher solubility of gadolinium than plutonium at low pH values. In the presence of the sludge components, Bibler [3] demonstrated that for the pH range from 3.5 to 4.5, 22 to 47% of the gadolinium dissolved upon acid addition while only 0.9 to 6.4% of the plutonium dissolved.

A possible explanation for this is that the dissolution of gadolinium is impeded by the presence of the greater relative amount of the more soluble sludge components. The results from these studies [2,3] indicate that the gadolinium solubility is dependent on the relative amount of the more soluble sludge components, and hence sufficient separation of plutonium and gadolinium in the precipitate sludge will not occur.

MINIMUM SUBCRITICAL Gd-To-Pu MASS RATIO

A criticality safety analysis was performed to determine the minimum safe Gd:Pu ratio in an infinite system [5]. The calculations were performed using MCNP 4B with the ENDF/B-V cross section library running on a Linux workstation. The analysis investigated the reactivity of the system at H:Pu atom ratios ranging from 30 to 3700. The minimum H:Pu of 30 was judged conservative based on the minimum H:Pu of 152 obtained for the centrifuged precipitate slurry [1]. The results are presented in Figure 1. The results indicate that an initial 1:1 weight ratio of Gd:^{239}Pu in the aqueous plutonium solution is sufficient to protect the plutonium at various process stages.

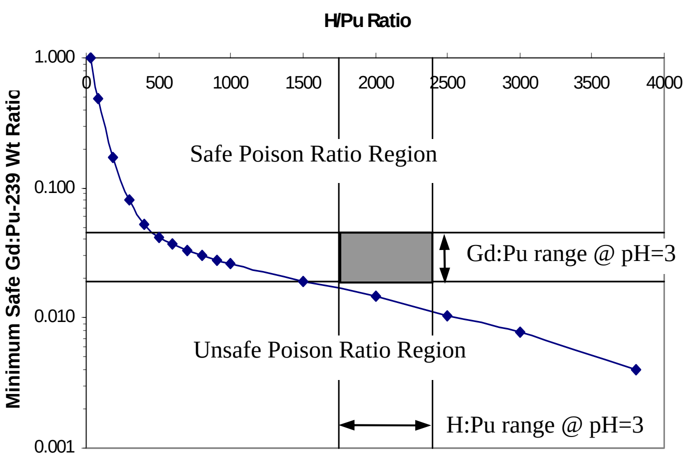

Fig. 1. Minimum Gd:Pu-239 Weight Ratio for Safe Operation

CONCLUSIONS

Use of gadolinium as a neutron poison for disposing of plutonium in an aqueous form is acceptable from criticality perspective. The criticality safety analysis [5] concludes that an infinite mixture of gadolinium and plutonium with a Gd:Pu weight ratio of 1:1 remains subcritical for a system with H:Pu ratio in excess of 30. Thus, a 1:1 weight ratio of Gd:^{239}Pu in the aqueous plutonium solution is sufficient to protect the plutonium at various stages of the disposition process.

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