

## PLUTONIUM BEHAVIOR IN BRINES AFTER EQUILIBRATION WITH PERICLASE (MgO) BACKFILL

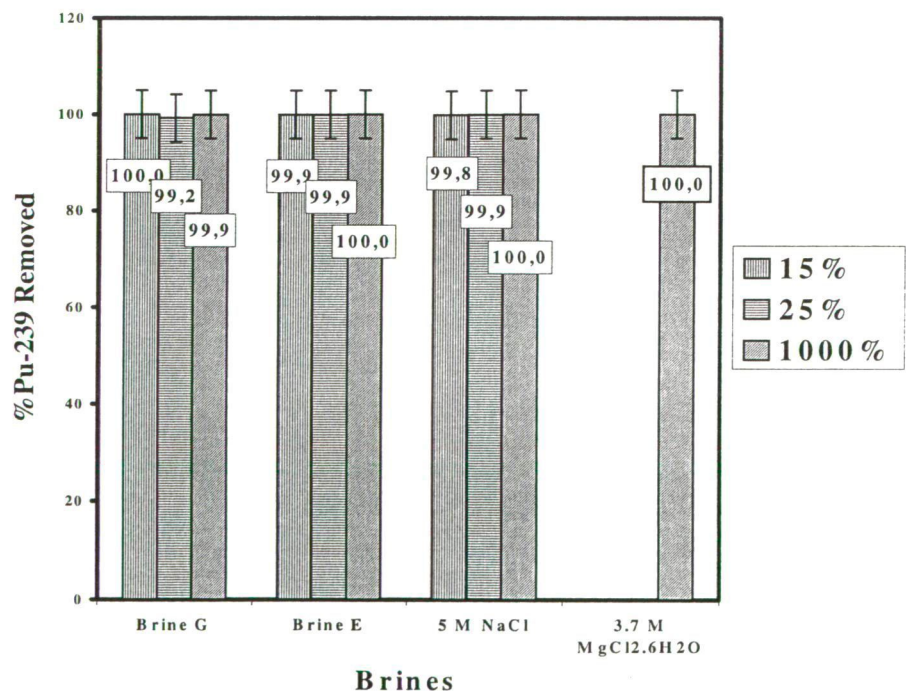
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Most radioactive waste disposal programs are considering the use of repository backfill materials to enhance the containment of radioactive waste. The concepts include the use of backfill to provide well-defined chemical conditions, favorable hydraulic conductivity, and desirable physical characteristics within the disposal facility. The backfill materials have been classified into two groups according to their primary properties: 1) chemical backfills such as cement, iron, phosphates, and MgO, and 2) hydrological/physical backfills such as clay, salt and cement. Magnesium oxide was proposed, and is being used, as a backfill material in a salt repository for transuranic waste in Carlsbad, New Mexico. It was chosen for its capacity to control pH and carbon dioxide (CO<sub>2</sub>) concentrations through specific reactions, maintaining the pH of the repository between 8.5 and 10, the range in which many radionuclides exhibit their lowest solubilities. In past years, most research has focused on the hydration and carbonation of MgO. A separate, but related issue concerns the heterogeneity of the repository in terms of rewetting after closure and the formation of new minerals in the backfill. Plutonium, with its multiplicity of oxidation states, is one of the primary actinides of concern for long-term disposal and storage of nuclear waste. The chemical behavior of plutonium is influenced strongly by its oxidation state, which determines the strength of its complexation reactions, solubility, formation of colloids and sorption processes. In this study, laboratory experiments were performed

to evaluate 1) stability of <sup>239</sup>Pu(VI) in synthetic Brine G, Brine E, 5M NaCl and 3.7M MgCl<sub>2</sub> • 6H<sub>2</sub>O as a function of time, 2) behavior of <sup>239</sup>Pu(VI) in the four brines after equilibrated with MgO backfill for 68 days at various water content, 3) release of <sup>239</sup>Pu from of Pu-MgO-brine agglomerates as a function of time, 4) characterization of <sup>239</sup>Pu-loaded – MgO-brine agglomerates. Equilibration experiments were conducted at MgO-to-water ratios of 1:0.15, 1:0.25 and 1:10. Release of <sup>239</sup>Pu from <sup>239</sup>Pu-loaded MgO agglomerates was determined in the presence or absence of hypochlorite (OCl<sup>-</sup>), under agitated and non-agitated conditions. After the <sup>239</sup>Pu(VI)-brines were equilibrated with MgO backfill for 68 days, the solution pH and alkalinity changed dramatically, with 99% to 100% of the <sup>239</sup>Pu(VI) being removed from the brines (Figure 1). Only a small amount of <sup>239</sup>Pu was subsequently released from the <sup>239</sup>Pu-loaded MgO-Brine G agglomerates after 110 days, but there was no <sup>239</sup>Pu released from the <sup>239</sup>Pu-loaded MgO-Brine E agglomerates. Our findings suggest that in NaCl-base brines such as Brine E, the studied MgO material is an effective backfill to buffer the pH to 9-10, which retards the release and migration of plutonium. In MgCl<sub>2</sub>-base brines such as Brine G, the MgO is slightly less effective because of buffering to a lower pH (8.0-8.5). The MgO showed great affinity for Pu under repository conditions and should perform well beyond its performance predictions.



**Fig. 1:** Percentage of <sup>239</sup>Pu removed from brines after equilibrated with MgO backfill for 68 days.