—ABSTRACT—

MAINTAINING CHEMICALLY REDUCING WASTE PACKAGE CONDITIONS IN AN OXIDIZING GEOCHEMICAL ENVIRONMENT

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ABSTRACT

Maintaining Chemically Reducing Waste Package Conditions in an Oxidizing Geochemical Environment

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Most fission products and actinides in spent nuclear fuel (SNF) are trapped in the uranium dioxide (UO_2) crystal structure and cannot escape until the UO_2 is oxidized. For a repository in an oxidizing geochemical environment, release of radionuclides can be delayed by using depleted uranium dioxide (DUO_2) and iron to locally maintain chemical reducing conditions for extended times after waste package (WP). The WP could be constructed of a cermet with DUO_2 imbedded within the steel (iron) in the cask walls and basket. Alternatively, the DUO_2 could be used in particulate form to fill void spaces in the WP.

Earlier studies (Forsberg, MRS, 2001) showed that the preferential oxidation of iron, and then DUO_2, can maintain chemically reducing conditions for potentially more than 100,000 years in a WP with SNF— if it is assumed that the only method of oxygen transport into the failed WP is as dissolved oxygen in the groundwater (i.e., oxygen flow is limited by groundwater flow). However, if air is present in the repository drifts, oxygen diffusion via the gas phase into the failed WP could also occur. The potential for diffusion of oxygen via the gas phase was analyzed based on diffusion in air and in water. Oxygen diffusion rates depend strongly upon (1) the minerals that are formed (uranium minerals and iron compounds) and their potential to reduce void spaces and (2) the potential for capillary forces to maintain water (an oxygen diffusion barrier) in the pore space of the degraded waste and WP. Under many conditions, the reducing materials in a failed WP can significantly delay SNF oxidation in an oxidizing geochemistry. This same effect that is seen in some uranium ore bodies.