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Desorption of Plutonium from Altered Nuclear Melt Glass Colloids

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Radionuclides have been introduced into the environment and transported by groundwater at nuclear weapons testing sites, such as Nevada National Security Site (NNSS), USA. In the case of underground nuclear testing, the majority of the actinides that remain after the detonation are sequesterd in glass formed during the cooling of the plasma. Thus, the migration of residual plutonium (Pu) will be controlled by the rate of melt glass alteration. In particular, the nature of the Pu associated with secondary colloids produced as a result of glass alteration will have a significant impact on the transport behavior of Pu. To evaluate the nature of Pu association with glass alteration products, nuclear melt glass from NNSS was hydrothermally altered at 25, 80, 140 and 200 °C for about 1000 days. The degree of secondary colloid formation increased with increasing temperature, from no observable alteration—to almost complete. Smectite and zeolite colloids were identified by XRD in the 140 and 200°C samples. In follow-on experiments, long- term, flow-cell desorption experiments were conducted using the glass alteration colloids formed at 140°C and 200°C. The Pu desorption for the 140°C and 200°C experiments ranged from 7-15% and 1-8%, respectively. The Pu desorption behavior was simulated using a numerical model previously developed for Pu-montmorillonite adsorption/desorption (Begg et al., 2017). Pu desorption from the 140°C colloids can be predicted using our numerical model of Pu desorption from montmorillonite. However, Pu desorption from 200°C colloids is overestimated. This suggests that our numerical model of Pu desorption from montmorillonite may not accurately describe the behavior of colloids formed at higher temperatures. The results suggest that Pu desorption from melt glass colloids formed at 200°C is kinetically limited and/or irreversible. A fraction of the Pu in these materials may be permanently associated with colloids, possibly via structural incorporation, and may, as a result, have much greater migration potential. This apparent irreversibility helps to explain why trace levels of Pu are found in certain downgradient wells many decades after underground nuclear testing has ended.

Begg, J.D., Zavarin, M. and Kersting, A.B. (2017) Desorption of plutonium from montmorillonite: An experimental and modeling study. Geochimica et Cosmochimica Acta
197, 278–293.