A fully coupled thermal-hydrologic-chemical (THC) reactive transport model is used to investigate possible mechanisms for plutonium (Pu) migration from an underground nuclear test. Pu was detected 1.3 km from the BENHAM nuclear test in two wells, a lava formation located near the depth of the blast, and a welded tuff aquifer located some 500 m above the detonation point. In both aquifers, Pu was observed in the presence of colloids. The BENHAM test, with an official yield of 1.15 megatons (DOE, 1994), produced a spherical cavity roughly 200 m in diameter. A cylindrical, rubblized, chimney extending from the working point of the test into the upper welded tuff aquifer formed as rock above the cavity collapsed. At the bottom of the cavity, a puddle of melted rock formed to an estimated depth of 35 m. A puzzling question arises as to how Pu could have migrated from the test site to two distant wells located in aquifers that apparently are out of communication with one another. To investigate this question, the computer code FLOTRAN (Lichtner, 2000), which couples transient, non-isothermal flow and reactive transport, was used to model Pu migration from the BENHAM underground test. Both dissolved and colloidal forms of Pu were included in the model. A mechanistic model for glass dissolution is incorporated in the model to describe the release of Pu from the melt glass. The glass dissolution rate is dependent on temperature of the melt glass and solution composition within the interstitial pore spaces. FLOTRAN accounts for changes in redox state, pH, carbonate and other species concentration over time and distance as the ambient fluid reacts with the melt glass and minerals in the chimney and surrounding host rock. The model can also account for sorption of radionuclides on rock surfaces and colloids through both ion exchange and surface complexation reactions. Two-dimensional calculations predict strong coupling between thermal, hydrologic, and chemical processes at the BENHAM test. The thermal perturbation produced by the explosion and subsequent cooling of the melt glass results in the formation of convection cells within the chimney. As convection becomes more vigorous, fluid is pushed vertically by buoyancy forces into the overlying tuff aquifer carrying along with it dissolved and colloidal form Pu. Once Pu reaches the upper aquifer it rapidly migrates horizontally through the fractured tuff aquifer as a consequence of the ambient flow field. As the melt glass cools, the convection cells collapse, resulting in a pulse release of Pu to both the upper and lower aquifers.

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