Neptunium Solubility Effect in the Unsaturated Zone at Yucca Mountain

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RESEARCH OBJECTIVES

The mobility of radionuclides (such as neptunium) in natural waters is affected by various factors—acidity, oxidation and reduction (redox) states, temperature, and aqueous composition. At Yucca Mountain, where high-level nuclear waste is to be stored in an underground geological repository, it is especially important to know the solubility of radionuclide minerals, because their solubility determines whether they will precipitate at this site. This study is aimed at evaluating the use of solubility models, in conjunction with aqueous chemical data, to quantify the mobility of radionuclides in the Yucca Mountain unsaturated zone.

APPROACH

The solubility of neptunium (Np) minerals was investigated using two representative controlling solids, the moderately soluble $Np_2O_{5(s)}$ and the highly insoluble $NpO_{2(c)}$ among a total of nine known Np minerals. Aqueous speciation is calculated in terms of each of the controlling solids, assuming equilibrium and employing the thermodynamic databases generated from the Yucca Mountain Project and the pertinent literature. A fixed concentration of total carbonate was used for speciation calculations.

ACCOMPLISHMENTS

Solubility calculations provide new insights into the behavior of Np nuclides in the unsaturated zone at Yucca Mountain. Results (Figure 1) emphasize the effects of pH and the redox potential, Eh. The effect of temperature was also investigated, and the solubilities of all the Np minerals were compared. Np solubility drops about 1 to 2.5 orders of magnitude in response to the pH conditions in the flow paths, which are slightly acidic in waters contacting waste package surfaces, and slightly alkaline in pore water within the tuffs of the Topopah Spring (TSw) and Calico Hills (CHn) units. The solubility of Np decreases with rising temperature. When the temperature rises from 25°C to 100°C, the solubility drops about 1 to 1.5 orders of magnitude.

SIGNIFICANCE OF FINDINGS

Solubility data provide chemical evidence of radionuclide precipitation as a likely mechanism for naturally enhanced radionuclide retardation in the Yucca Mountain unsaturated zone. The trends for Np_2O_5 solubility also hold for the highly insoluble NpO_2, and the solubility of the latter is in a comparable order of magnitude to that of the former at higher redox conditions. The results indicate that solubility investigations could provide a method by which to evaluate the mobility of radionuclides in natural systems.

RELATED PUBLICATION

Lu, G., Impact of neptunium solubility on radionuclide retardation in the unsaturated zone at Yucca Mountain. Applied Geochemistry (submitted), 2005.

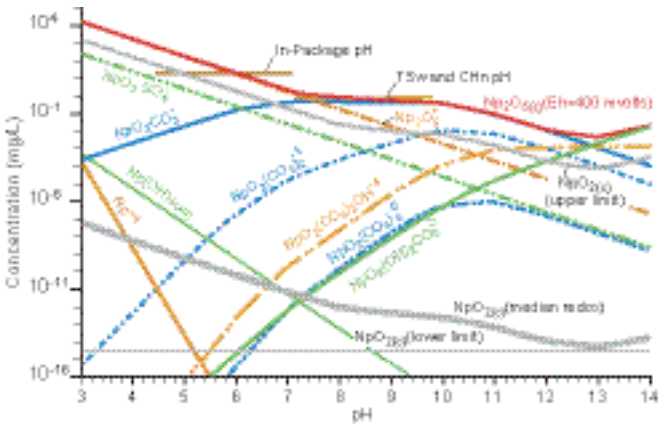

Figure 1. Calculated $Np_2O_{5(s)}$ solubility of neptunium (total aqueous carbonate = 115 mg/L, $[SO_4^{-}]$ = 60 mg/L, temperature at 25°C, and Eh at 400 mV for $Np_2O_{5(s)}$), contributing species are in color. In the background are total solubility curves (in solid gray) for $NpO_{2(c)}$, with Eh covering the water redox stability field ranges.

ACKNOWLEDGMENTS

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