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Assessing Radionuclide Solubility Limits for Cement-Based, Near-Surface Disposal

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In shallow, cement-based nuclear waste disposal systems, solubility limits may be used in radionuclide release models assessing long-term performance. These limits should reflect the evolving chemical environment during degradation of the cement-based host. Solubility limit models, using Geochemist's Workbench®, were developed for waters associated with a waste form or host grout containing ordinary Portland cement, fly ash, and blast furnace slag. Initial pH may be as high as 13.5, with Eh_{SHE} as low as −350 mV, but pH will likely be around 12.5 for an extended period. Continuing degradation under the influence of infiltrating waters will lower pH and raise Eh. Interaction with unsaturated zone waters would drive chemistry toward pH ~8 and Eh >600 mV. The solubility models spanned this pH–Eh region; uncertainties due to selection of thermodynamic data and controlling solids were considered, and the models were evaluated in the light of experimental data. Selenium concentrations are unlimited or limited to high concentrations at high Eh, and limited to low concentrations at lowest Eh for pH 11.9–12.6. Technetium solubility is similarly unlimited except at lowest Eh and pH ≥ 12. Uranium solubility limit patterns are less regular, showing the influences of major ion concentrations. Most predicted uranium solubility limits are in the range of 10^{-8} to 10^{-6} M, with deviations to lower values at lowest Eh and to higher values if sodium is especially low. Under reducing conditions, neptunium concentration is limited to around 10^{-9} M, but rises to 10^{-5} to 10^{-4} M as Eh increases. Under initial conditions, plutonium solubility is limited to around 10^{-9} M; this limit will rise as pH and Eh evolve, reaching as high as 10^{-7} M, depending on carbonate content. Colloids, if stable, may sustain plutonium concentrations of ≥ 10^{-8} M.