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NEPTUNIUM SOLUBILITY IN THE NEAR-FIELD ENVIRONMENT OF A PROPOSED YUCCA MOUNTAIN REPOSITORY, David C. Sassani, Management and Technical Support Contractor/Golder Associates Inc., Abe Van Luik, Office of Repository Development, U.S. Department of Energy, Jane Summerson, Office of Repository Development, U.S. Department of Energy

For representing the source-term of a proposed repository at Yucca Mountain, NV, total system performance assessment models evaluate the disequilibrium degradation of the waste forms to capture a bounding rate for radionuclide source-term availability and use solubility constraints that are more representative of longer-term, equilibrium processes to limit radionuclide mass transport from the source-term. These solubility limits capture precipitation processes occurring either as the waste forms alter, or in the near-field environment as chemical conditions evolve. A number of alternative models for solubility controls on dissolved neptunium concentrations have been evaluated. These include idealized models based on precipitation of neptunium as separate oxide minerals and more complex considerations of trace amounts of neptunium being incorporated into the secondary uranyl phases from waste form alteration. Thermodynamic models for neptunium under oxidizing conditions indicate that tetravalent neptunium (NpO_2) solids are more stable relative to pentavalent (Np_2O_5) phases, and thereby set lower dissolved concentrations of neptunyl species. Data on solids and solutions from slow flow through (dripping) tests on spent fuel grains indicate that neptunium is tetravalent in the spent fuel and that over ~9 years the neptunium concentrations are near to or below calculated NpO_2 solubility. The possibility of kinetic rate limitations to NpO_2 precipitation has led to temperature-dependent studies of NpO_2 precipitation kinetics and solubility to reduce uncertainties and confirm application of the model.