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ALTERNATIVE RELEASE MODELS

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May 25, 1999
San Antonio, Texas

DOE/NRC Total System Performance Assessment
Technical Exchange

THREE OF SEVERAL ALTERNATE SOURCE TERM MODELS IN NRC/CNWRA TOTAL-SYSTEM PERFORMANCE ASSESSMENT

- Base Case: Regression of Data (Gray et al.,) for the Dissolution Rate of Spent Fuel in a Multicomponent Solution with an Arrhenius Temperature Extrapolation

- Natural Analog Oxidation Rate: Release Rate Based on Maximum Average Oxidation Rate Estimated for the Nopal I Uranium Deposit at Peña Blanca, Mexico

- Schoepite Solubility: Release Rate Based on Schoepite Solubility as a Function of Temperature, pH, and Uranyl Carbonate Speciation, and Release of Matrix Radioelements in Proportion to Uranium

(Gap and Grain Boundary Species Are Released Rapidly in All Cases; Solubility Limits for All Radioelements are Respected in All Cases)

NOPAL I URANIUM DEPOSIT AT PEÑA BLANCA, MEXICO: NATURAL ANALOG FOR THE PROPOSED REPOSITORY AT YUCCA MOUNTAIN

- \+ Fractured Silicic Volcanic Host Rocks
- \+ Unsaturated Zone Hydrology in Semi-Arid Climate
- \+ Primary Uraninite (Analog of Spent Fuel) Almost Completely Oxidized to Uranyl Phases, e.g., Schoepite, Soddyite, and Uranophane
- \+ Relevant Time Scales: Uraninite Deposition ~ 8 million years ago; Oxidizing Conditions > 3 million years.
- – Evidence for Hydrothermalism Involving Acid-Sulfate Conditions
- – Evidence for an Episode of (Rapid?) Oxidative Alteration

MAXIMUM AVERAGE OXIDATIVE ALTERATION RATE OF URANINITE AT NOPAL I

$$R_o = (U_e / t) + F\,C$$

- U_e: Amount of Oxidized Uranium Remaining (320 metric tons)
- t: Minimum Time Period of Oxidative Alteration (3 million years = U-Pb Age of Late Forming Uranophane)
- F: Maximum Volumetric Water Flow (Average Precipitation × Oversized Cross Section = 1200 $m^3\ yr^{-1}$)
- C: Uranium Concentration in Exiting Water (10^{-7} M Based on Uranyl Mineral Solubility Calculations)
- R_o: Maximum Average Oxidation Rate = 140 $g\ yr^{-1}$

NOPAL MAXIMUM AVERAGE OXIDATIVE ALTERATION RATE SCALED TO THE PROPOSED REPOSITORY AT YUCCA MOUNTAIN

$140\ g\ yr^{-1} \times 63{,}000 / (320 + 88)$ (ratio of uranium masses) $= 22\ kg\ yr^{-1}$

- Oxidative Alteration Rate is a Maximum Limit on Release Rate from the Spent Fuel Matrix
- Peña Blanca Data Demonstrate that Oxidative Alteration is Rapid Relative to Removal of Uranium from the System

SCHOEPITE SOLUBILITY

- Uranyl Minerals are Stable in the Yucca Mountain Environment Relative to Spent Nuclear Fuel
- Natural, Laboratory, and Crystallographic Evidence Points to Incorporation of Minor Radioelements in the Structures of Secondary Uranyl Minerals
- Thermodynamic Data Permit Calculation of Schoepite Solubility as a Function of Temperature, pH, and Aqueous Speciation
- Uranyl Minerals Have Retrograde Solubilities with Temperature
- Solutions Passing Through the Waste Package Are Assumed to Be Saturated with Respect to Schoepite and to Contain Other Spent Fuel Matrix Species in Proportion to Uranium may be conservative may be non-conservative

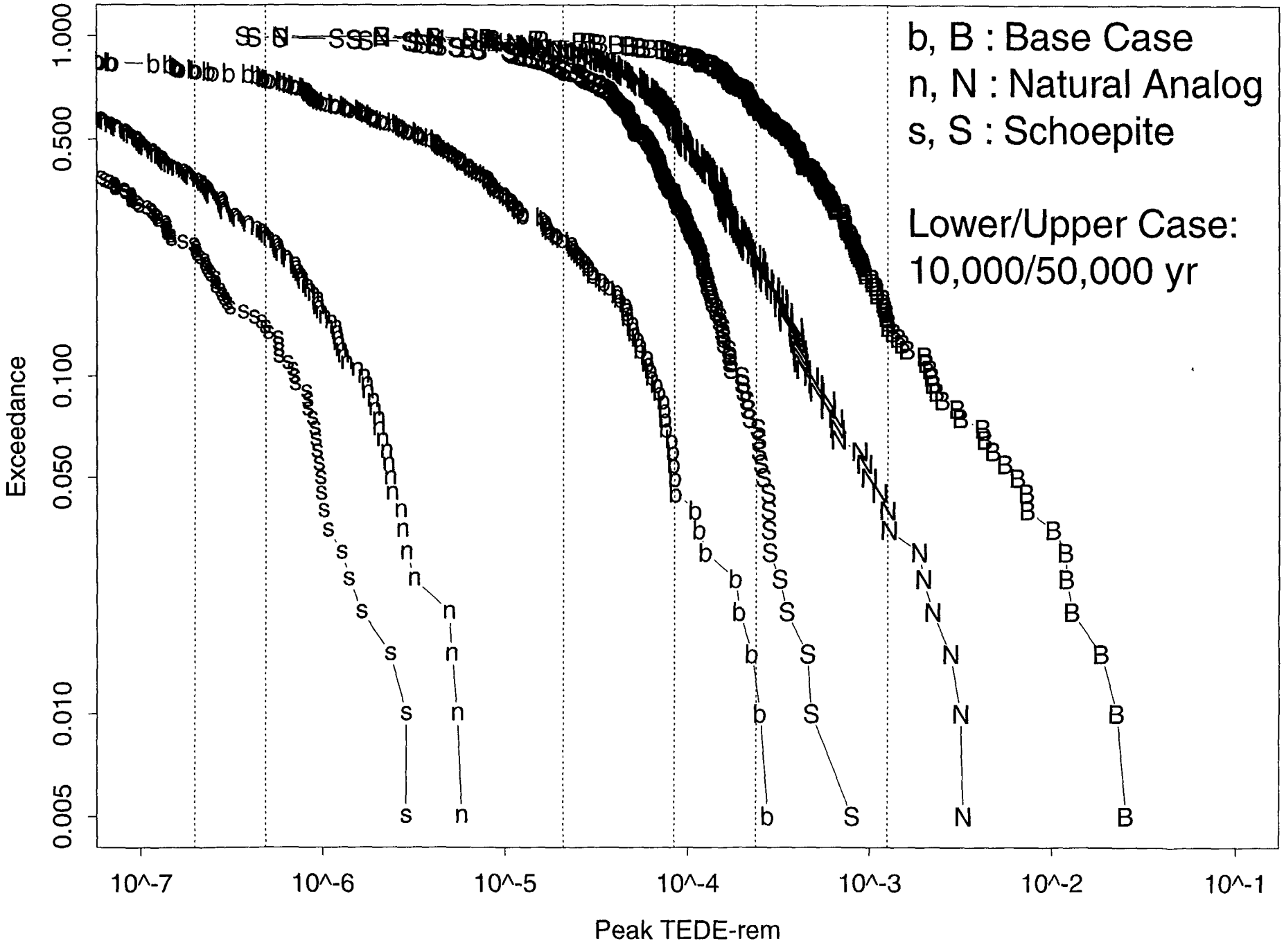
REACTIONS AND MASS ACTION RELATIONS FOR SCHOEPITE SOLUBILITY

Number	Reaction	Mass Action Relation
0	$UO_2(OH)_2 + 2H^+ \rightleftarrows UO_2^{2+} + 2H_2O$	$[UO_2(OH)_2] = [UO_2^{2+}] / K_0[H^+]^2$
1	$UO_2CO_3 + H^+ \rightleftarrows UO_2^{2+} + HCO_3^-$	$[UO_2CO_3] = [UO_2^{2+}][HCO_3^-] / K_1[H^+]$
2	$UO_2(CO_3)_2^{2-} + 2H^+ \rightleftarrows UO_2^{2+} + 2HCO_3^-$	$[UO_2(CO_3)_2^{2-}] = [UO_2^{2+}][HCO_3^-]^2 / K_2[H^+]^2$
3	$UO_2(CO_3)_3^{4-} + 3H^+ \rightleftarrows UO_2^{2+} + 3HCO_3^-$	$[UO_2(CO_3)_3^{4-}] = [UO_2^{2+}][HCO_3^-]^3 / K_3[H^+]^3$
4	$UO_3 \cdot 2H_2O + 2H^+ \rightleftarrows UO_2^{2+} + 3H_2O$	$[UO_2^{2+}] = K_4[H^+]^2$

The temperature dependence of the equilibrium constants is given by the Van't Hoff equation

$$\ln K_i = \ln K_i^0 + \frac{\Delta H_i^0}{R}\left[\frac{1}{T^0} - \frac{1}{T}\right] \qquad (3)$$

Yucca Mountain CCDF for Peak Annual Dose at 20 km Distance



QUALIFICATIONS FOR THE ANALOG SOURCE TERM MODEL

- **Possible Nonconservatism**
  - — **Evidence for episodic secondary mineral formation at 3 million years and episodic uranium mobilization at 400,000 years and at 50,000 years**
- **Conservatisms Adopted in the Model**
  - — **Minimum period of oxidation based on ages of late forming uranophane**
  - — **Upper bound on infiltration equal to precipitation over a large area**
  - — **Model release rate equal to oxidation rate**

QUALIFICATIONS FOR THE SCHOEPITE SOLUBILITY SOURCE TERM MODEL

- Possible Nonconservatisms
  - — All radionuclides (except gap and grain boundary inventories) are incorporated in schoepite in proportion to their concentrations in the spent fuel matrix
  - — More stable tertiary phases (e.g., uranophane) may form and release initially coprecipitated radionuclides.
- Conservatism adopted in the model
  - — A role for secondary phases for control of the radionuclide source term is realistic.

OBSERVATIONS

- Both natural analog and schoepite source term models yield lower doses than the NRC TPA3.2 base case model.
- Both natural analog and base case uranium releases depend on sampled uranium solubility limits. Schoepite solubility model releases depend on calculations of uranium solubility as a function of temperature and solution chemistry.
- Cumulative release of ^{237}Np from the EBS at 50,000 years for the natural analog model is 14 percent of the base case release, and for the schoepite solubility model it is 0.07 percent of the base case release.
- Consideration of the role of secondary phases could reduce conservatism in performance assessment models for Yucca Mountain.
- Natural analog information can contribute to performance assessment.