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January 16, 2012
Contract No. NRC–02–07–006
Account No. 20.14005.02.001

Attn: Document Control Desk
U.S. Nuclear Regulatory Commission
Dr. James Rubenstone
Office of Nuclear Material Safety and Safeguards
Division of High-Level Waste Repository Safety
Mail Stop EBB-2-B02
Washington, DC 20555

SUBJECT: Draft Administrative Item 14005.02.001.202, Redox Processes in the Safety Case for the Proposed U.S. Repository

Dear Dr. Rubenstone:

This letter transmits the subject presentation, which is intended to be delivered by Dr. Budhi Sagar at the Final ReCosy (Redox Phenomena Controlling Systems) Workshop, January 23–26, 2012, in Karlsruhe, Germany. Dr. Sagar is one of the peer reviewers for this European Commission-sponsored project related to assessing performance of geologic repositories. We are requesting only U.S. Nuclear Regulatory Commission (NRC) approval of the presentation; Dr. Sagar's travel and time for attendance at the workshop are not being funded by NRC projects. The project organizers invited Dr. Sagar to discuss the role of oxidation-reduction processes in the safety case for the proposed Yucca Mountain repository. The presentation uses only publicly-available information sources and does not include any regulatory conclusions. The only conclusion presented is factual, that is, redox conditions did not play a major role in the U.S. Department of Energy's performance assessment because oxidizing conditions were assumed throughout the system.

We apologize for the short time frame request for NRC approval. Dr. Sagar was only recently invited to make the presentation. We will work closely with you to make any needed revisions.

If you have any questions regarding the presentation, please contact Dr. Sagar at (210) 522-5252 or me at (210) 522-5582.

Sincerely,

David Pickett, Ph.D.
Senior Program Manager
Integrated Spent Fuel Regulatory
Program-Geologic Disposal

DP: ar
Enclosure: Redox Processes in the Safety Case_Presentation

cc:

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Redox Processes in the Safety Case for the Proposed U.S. Repository

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Budhi Sagar

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4th Annual ReCosy Workshop, Karlsruhe, Germany, January 23–26, 2012

Disclaimer

- The views expressed herein are those of the authors and do not constitute a final judgment or determination of the matters addressed or of the acceptability of any licensing action that may be under consideration by the U.S. Nuclear Regulatory Commission (USNRC).

Overview

- Yucca Mountain postclosure safety case relied chiefly on performance assessment (PA) and barrier analysis
- Applicant assumption in all cases: oxidizing conditions
- Confirmatory calculations by regulator
- NRC technical review documented in postclosure technical evaluation report, NUREG–2107
  - Not a licensing review

Yucca Mountain

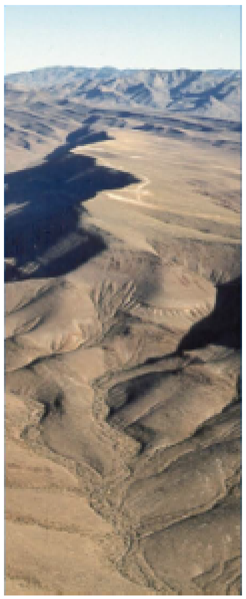


Figure 1-1. Location of Yucca Mountain in the Great Basin

From Yucca Mountain Safety Analysis Report, DOE/RW–0573 Rev. 1

DOE Representation of Yucca Mountain Upper Natural Barrier

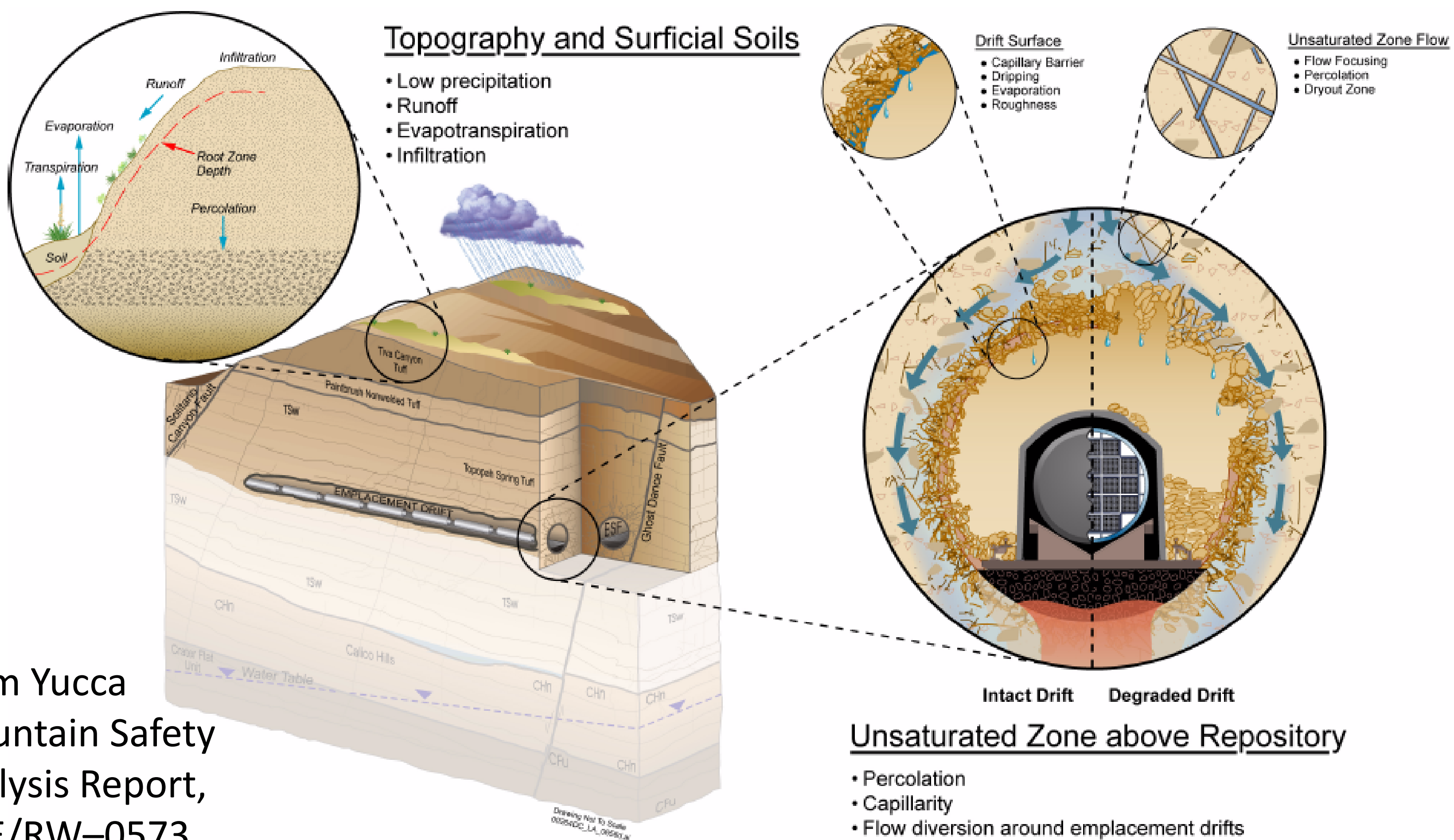
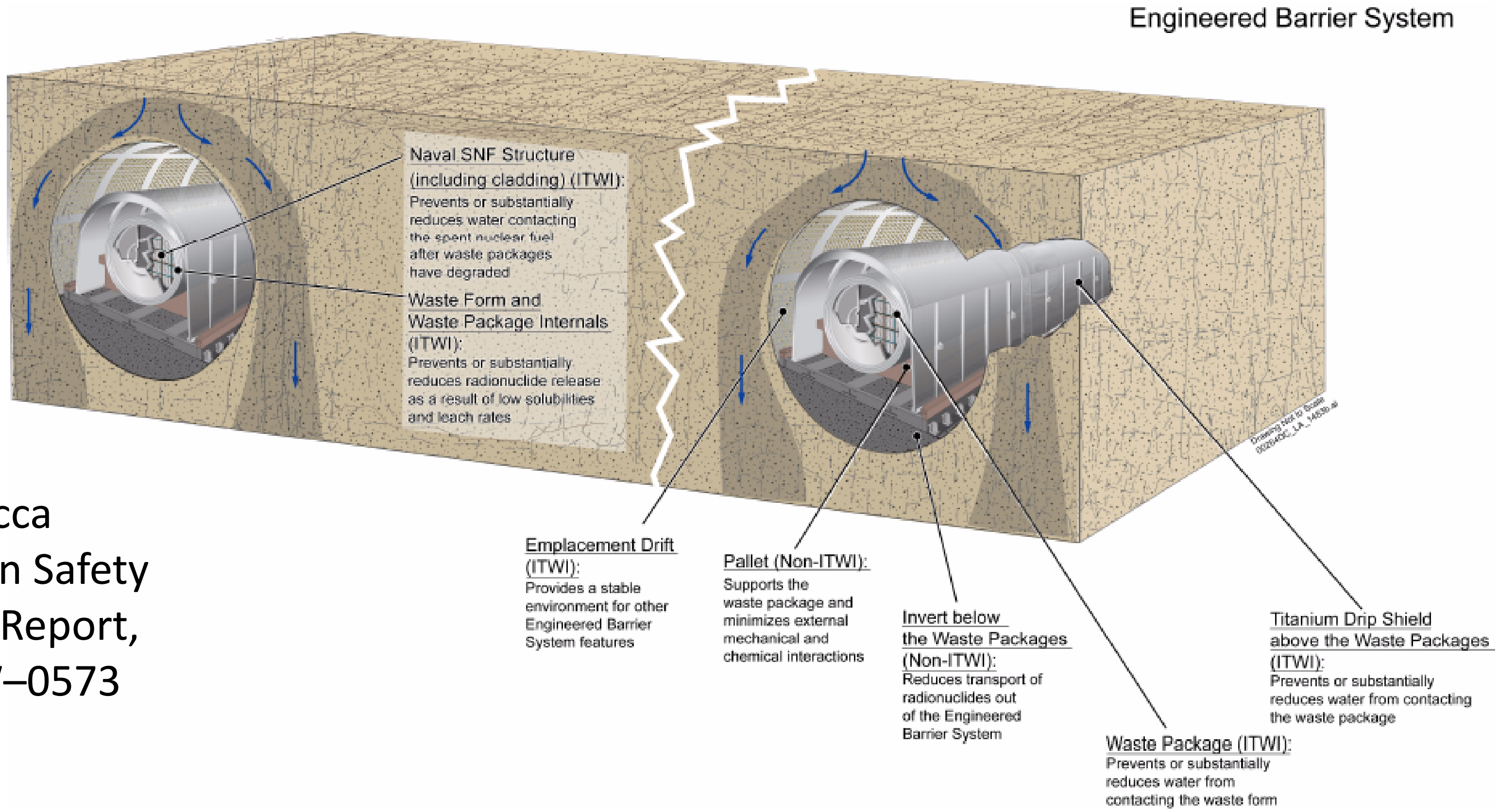


Figure 2.1-2. Schematic of the Upper Natural Barrier

From Yucca Mountain Safety Analysis Report, DOE/RW–0573 Rev. 1

DOE Representation of Yucca Engineered Barrier System



From Yucca Mountain Safety Analysis Report, DOE/RW–0573 Rev. 1

NOTE: ITWI = Important to waste isolation, Non-ITWI = Not important to waste isolation.

Figure 2.1-7. Schematic of the Engineered Barrier System

DOE Representation of Lower Natural Barrier

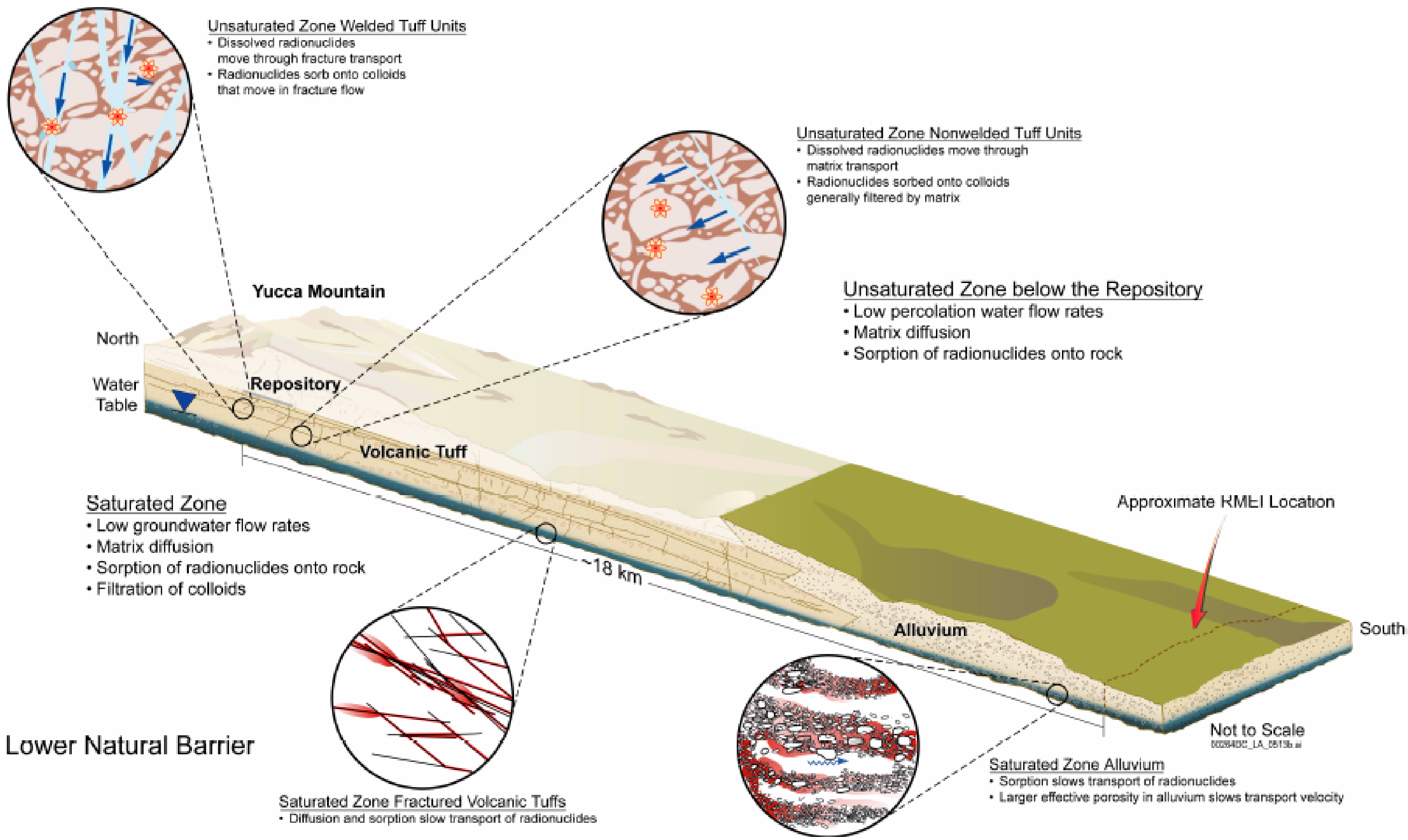


Figure 2.1-30. Schematic of Lower Natural Barrier

From Yucca Mountain Safety Analysis Report, DOE/RW–0573 Rev. 1

DOE TSPA

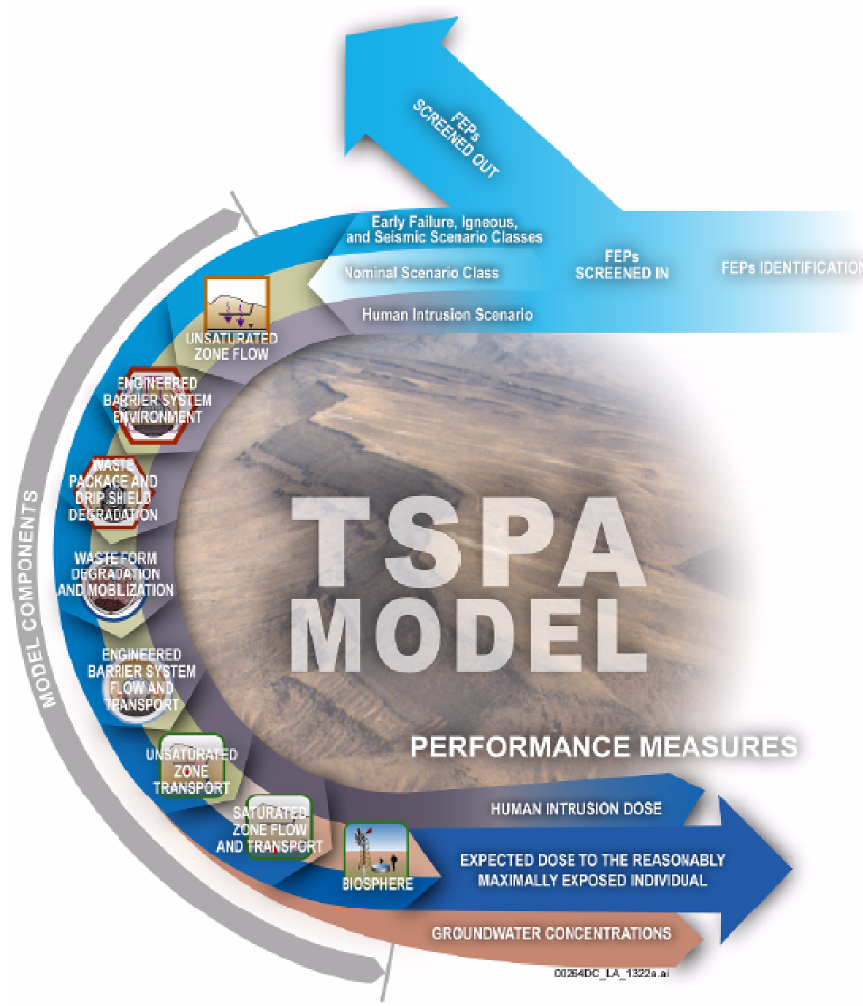


Figure 2.4-1. Schematic Representation of the Development of the TSPA Model, Including the Nominal, Early Failure, Igneous, and Seismic Scenario Classes, as Well as the Human Intrusion Scenario

From Yucca Mountain Safety Analysis Report, DOE/RW–0573 Rev. 1

Applicant Approach

- Assumed oxidizing conditions in drift, inside package (with some exceptions), and below drift
  - Unsaturated zone
  - Lack of basis for asserting reducing conditions
- Important performance implications
  - Metal component degradation faster
  - Spent fuel degradation faster
  - Higher radionuclide solubilities
  - Lower sorption
  - Can, in general, neglect radiolytic effects
- Beyond this assumption, redox was therefore generally not considered with few exceptions

Example Redox-Sensitive Radioelements

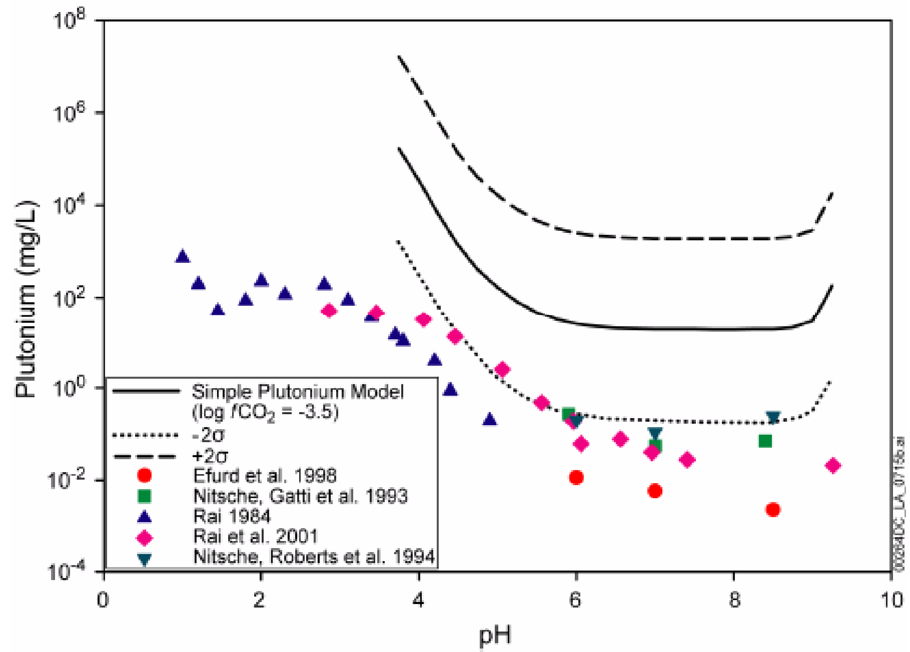
Mean parameter values used in DOE performance assessment (Yucca Mountain Repository Safety Analysis Report, DOE/RW–0573, Rev. 1)

	Mean Solubility Limit*	Mean Alluvium Sorption Coefficient (K_d)
Technetium	No solubility control	No retardation
Neptunium	$NpO_2 = 5 \times 10^{-8}$ M $Np_2O_5 = 8 \times 10^{-7}$ M	6 L/kg
Plutonium	1×10^{-8} M	100 L/kg

*At pH 8 and $\log f(CO_2) = -3.5$.

Exception: Plutonium

- Plutonium (Pu) solubility limit
  - Applicant used an “adjusted-Eh” model that reduced Eh from what would be imposed by atmospheric oxygen
  - Resulted in lower Pu solubilities



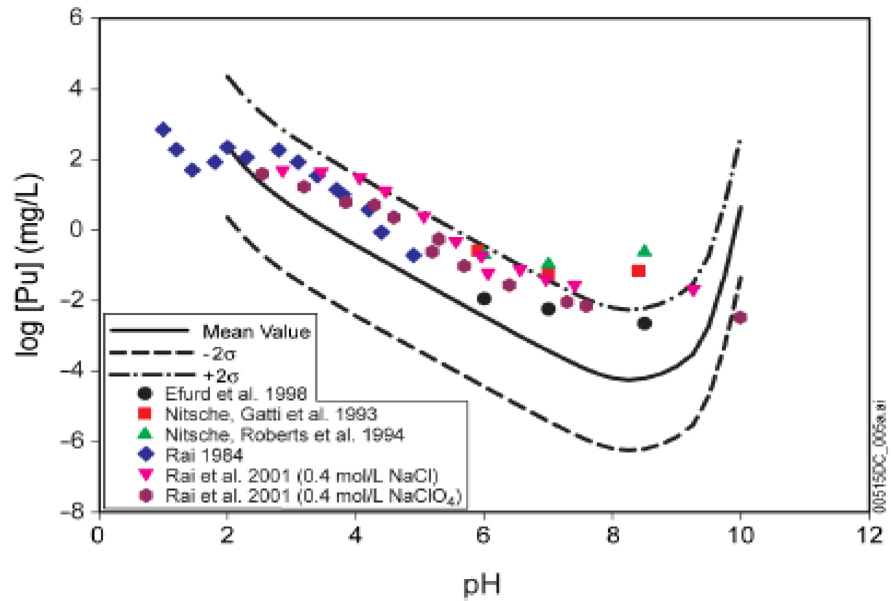
Applicant argued that Pu solubility model using equilibrium with atmospheric O_2 (solid line above) gave excessively high Pu concentrations compared to lab data.

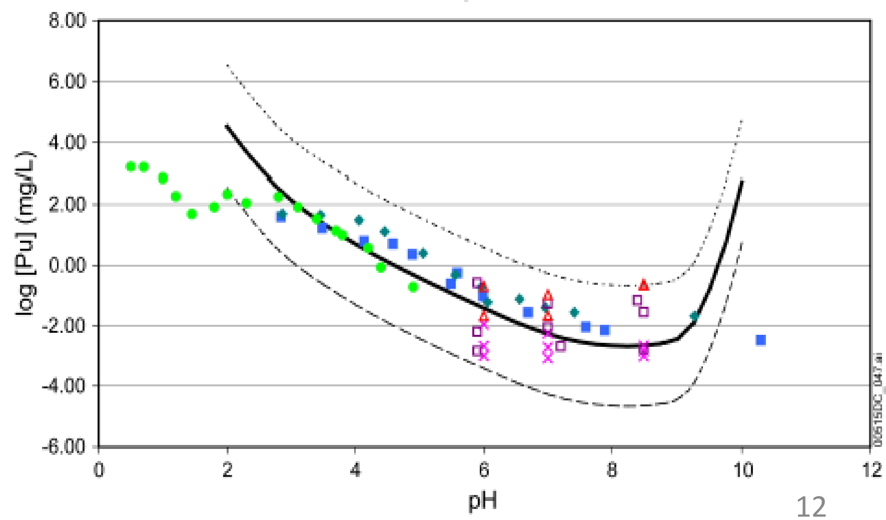
Figure 2.3.7-35 of Yucca Mountain Safety Analysis Report, DOE/RW–0573, Rev. 1

Exception: Plutonium (continued)

- Initial adjusted-Eh model (top) used Eh-pH relationship measured in natural waters
- Judged inconsistent with lab data, so Eh was adjusted upward to match data (bottom)
- Argued to be more consistent with laboratory and field data

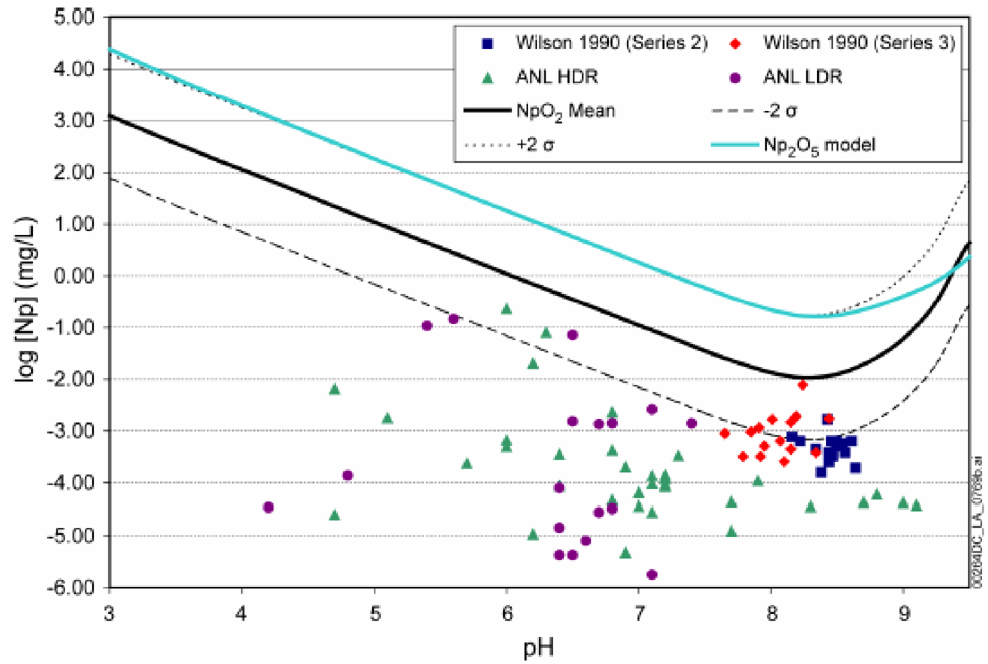
Figures V-8 and V-9 from Sandia National Laboratories, 2007, ANL–WIS–MD–000010, Rev. 6





Exception: Neptunium

- Neptunium (Np) solubility limit
  - Model controlled by Np(IV)O_2 until in-package steel exhausted, then Np(V)$_2O_5$
  - NpO_2 = lower Np
  - NpO_2 may be thermodynamically favored under oxidizing in-package conditions, but evidence is inconclusive
  - Applicant supported choice of NpO_2 by suggesting local conditions would be reducing when steel is corroding



Two Np solubility limit models shown, with comparison to Np concentration data from spent fuel leaching tests

Figure 2.3.7-39 of Yucca Mountain Safety Analysis Report, DOE/RW–0573, Rev. 1

Redox Considered Conservative

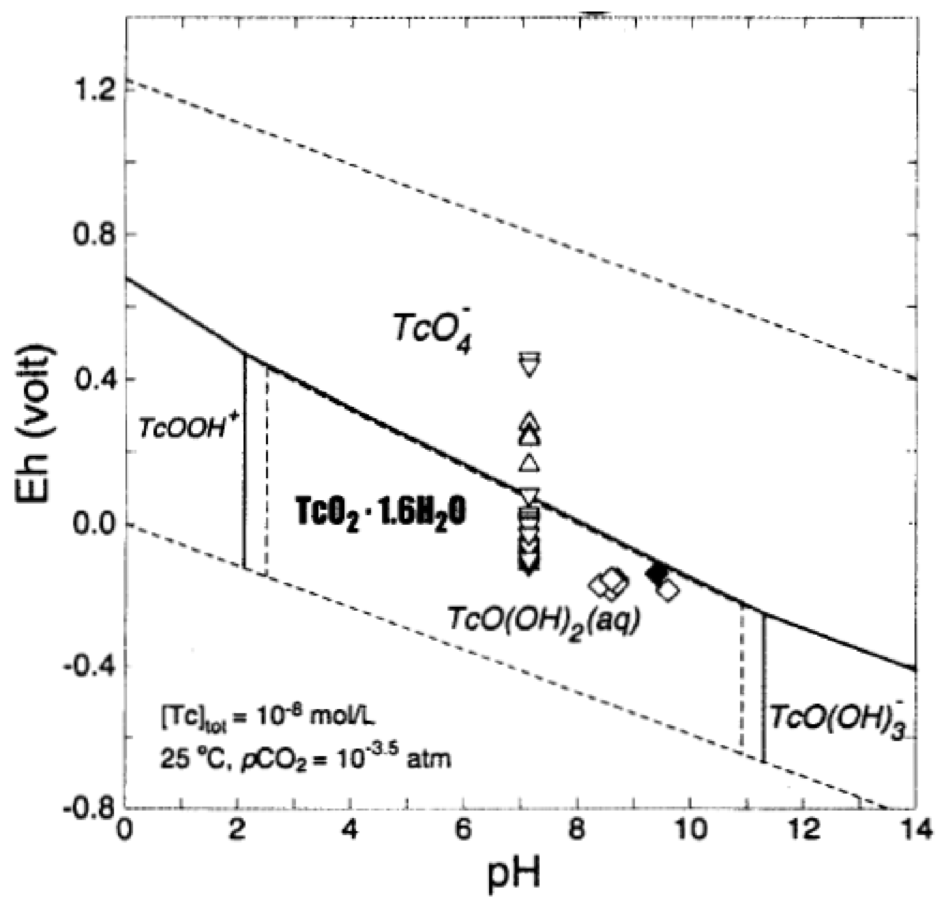
- Applicant frequently bolstered arguments by claiming that oxidizing assumption was conservative and that locally reducing conditions were plausible (drift, package, below drift)
- Applicant cited local low-Eh zones in the saturated zone as beneficial, but did not use in PA
  - For example, redox sensitivity analyses in Arnold & Houseworth, "Impacts of Solubility and Other Geochemical Processes on Radionuclide Retardation in the Natural System," Rev. 1, 2006

Regulator Perspective

- Redox considered in independent and confirmatory process models and PA abstractions
- NRC/CNWRA PA assumed oxidizing conditions along transport path
  – Reasonably conservative

Independent NRC/CNWRA Technetium Calculation

- Pabalan, Turner, and Miklas (2000) Scientific Basis for Nuclear Waste Management XXIII, MRS Symposium Proceedings 608, pp 231–236.
- In locally reducing zones below the water table, technetium (Tc) would have low solubility and high sorption
- Figure from Pabalan, et al. (right) shows calculated Tc speciation and Eh-pH measured in Yucca Mountain vicinity waters



Independent NRC/CNWRA Technetium Calculation (continued)

- Sensitivity analyses conducted using NRC/CNWRA PA (TPA Version 3.2)
  - Decreased Tc solubility if waters are reducing, due to reduction of Tc(VII) to Tc(IV), can decrease calculated Tc-99 dose
  - If applied throughout the transport pathway, or throughout the alluvial aquifer only, increased Tc sorption significantly delays Tc-99 transport
- Not included in base case of NRC/CNWRA PA
  - Technical basis lacking that reducing waters are sufficiently widespread to affect performance
  - Base case considered conservative for transport of Tc

Summary

- PA calculations for the proposed Yucca Mountain repository assumed oxidizing conditions throughout the system
- Applicant solubility limit abstractions, however, took into account somewhat lower Eh (Pu) or locally reducing effects of steel corrosion (Np)
- Sensitivity analyses showed potentially lower Tc solubility under more reducing conditions in transport path, but not credited by applicant or implemented in PA