Secondary Minerals and Radionuclide Transport in a Yucca Mountain Repository: Key Observations Motivated by Studies at Peña Blanca

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Key Observations: Secondary Minerals

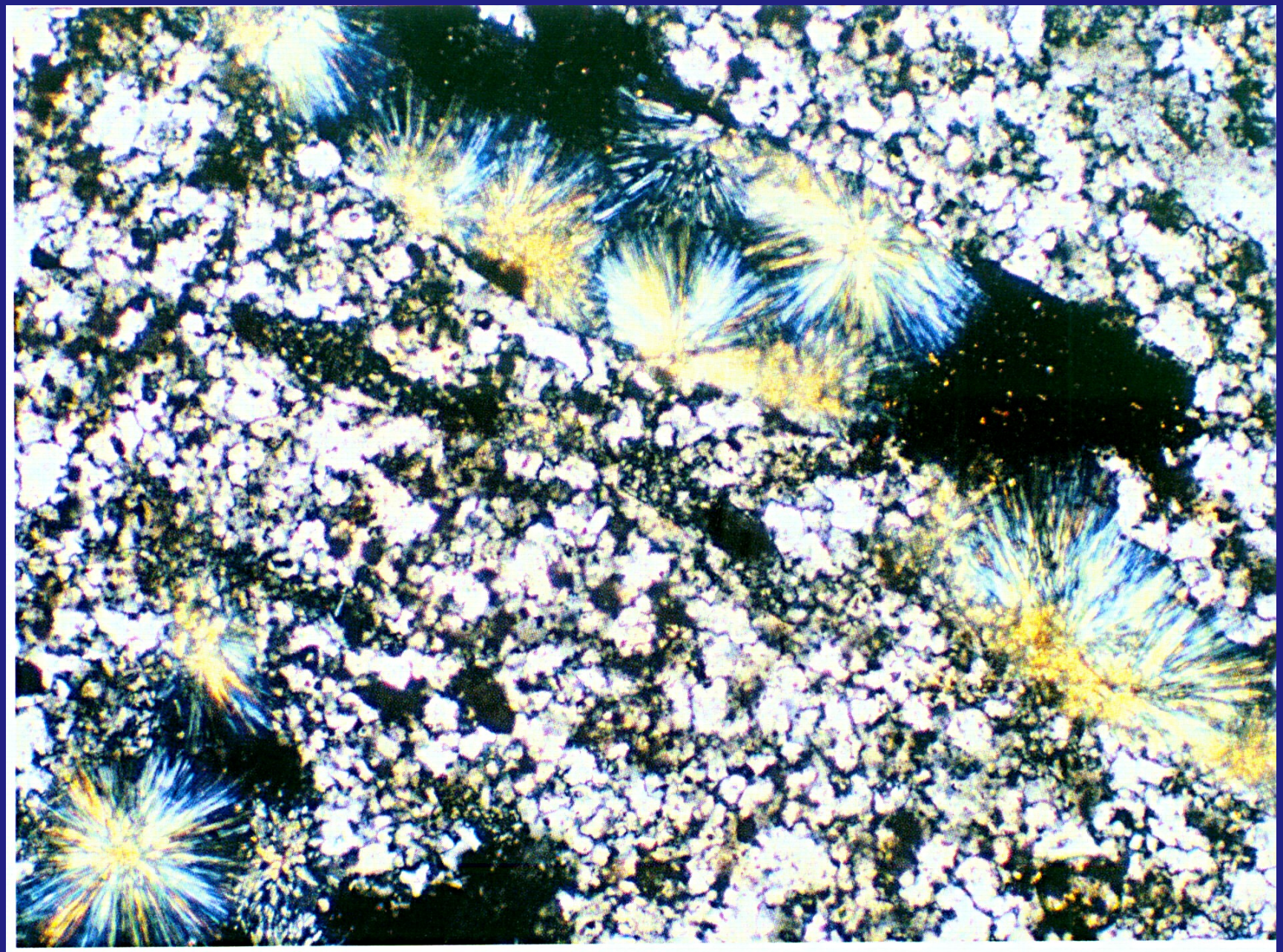
Releases of radionuclides at Yucca Mountain will be controlled by dissolution of thermodynamically unstable spent nuclear fuel, and by formation and alteration of more stable secondary minerals.

Theoretical, experimental, and natural analog studies provide converging evidence for the role of hydrous uranyl minerals in controlling radionuclide releases.

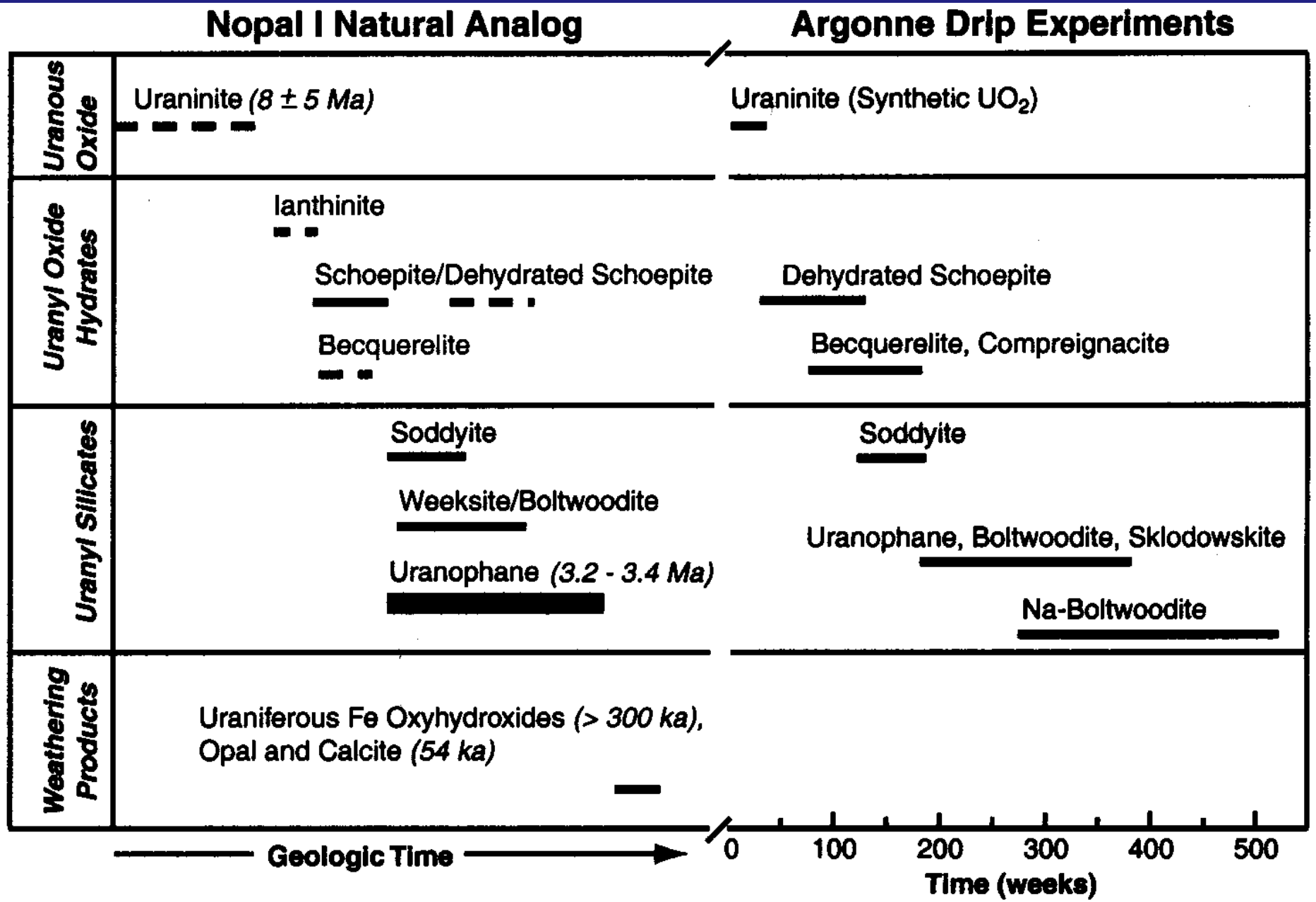
Nopal I, 0 level adit

Alteration of uraninite to hydrous uranyl minerals in silicified tuff
4 centimeters across

Accicular weeksite crystals in fractures (1 mm across)



Uranium mineral paragenesis (compiled by Murphy, 2000)



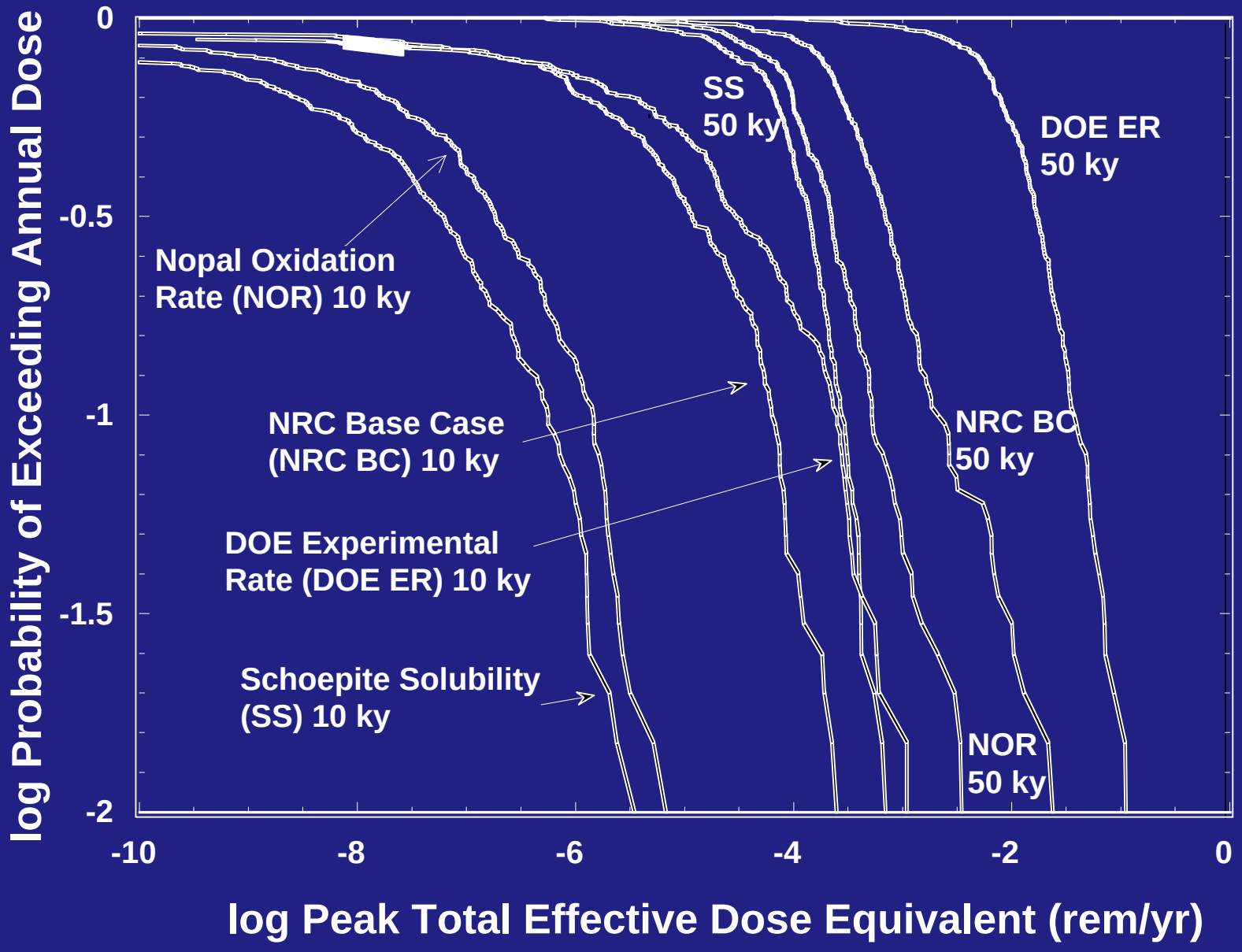
Key Observations: Alternate PA Models

Alternate models show potentially improved repository performance considering:

- Dissolution rate of spent fuel based on estimates of oxidation rates of uraninite at the Peña Blanca analog site
- Coprecipitation coupled with solubility-limited dissolution of schoepite, a secondary uranyl mineral.

Alternate Performance Assessment Models for Yucca Mountain

Murphy and Codell (1999)



Key Observations: Coprecipitation

Coprecipitation of radionuclides in secondary uranyl minerals is an attractive mechanism of sequestration, but experimental data that could provide a quantitative basis for this process are inconclusive.

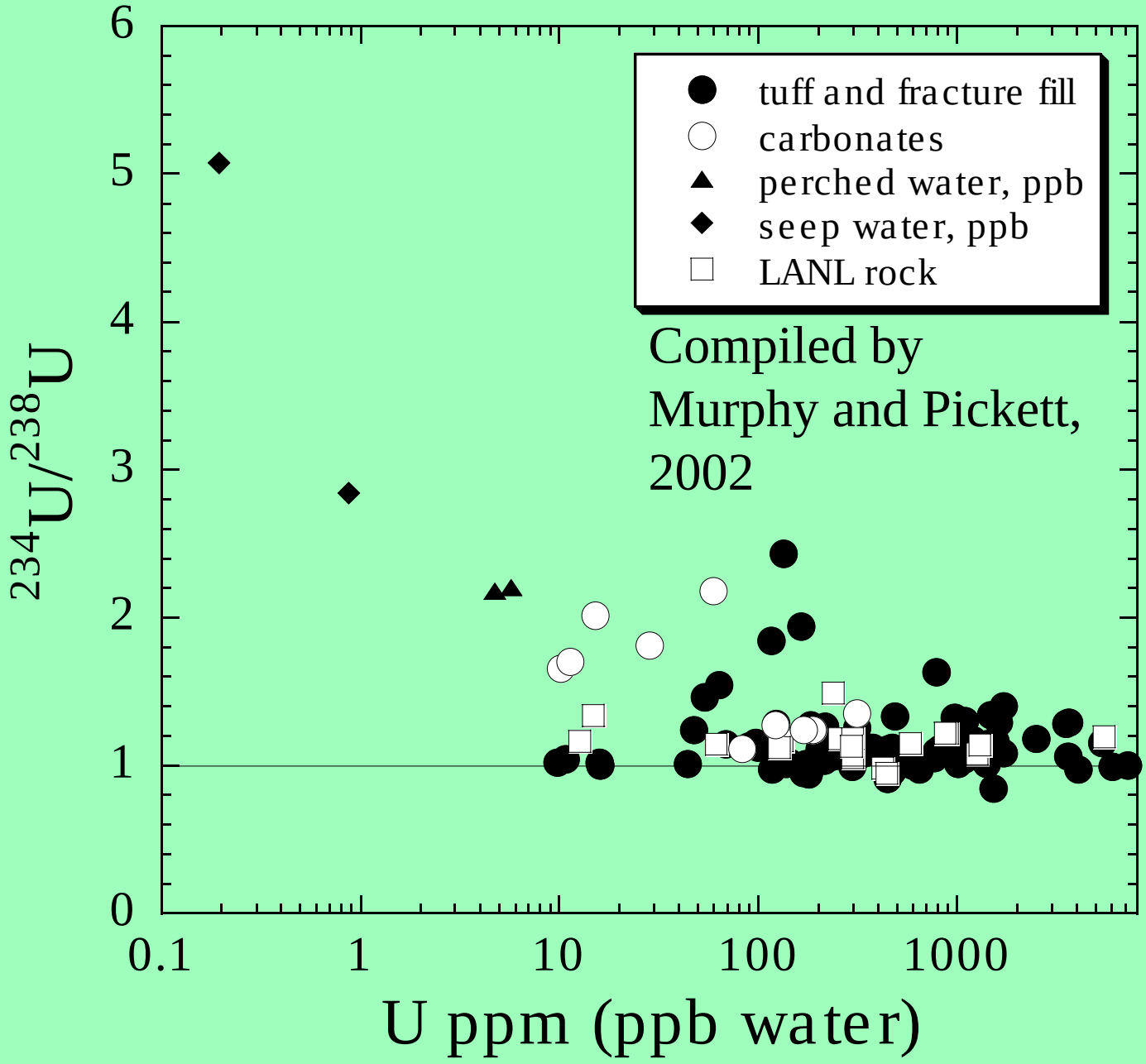
Equilibrium solubilities and distribution coefficients are uncertain, and kinetic effects may outweigh equilibrium controls.

Key Observations: Radioisotope Constraints and Effects

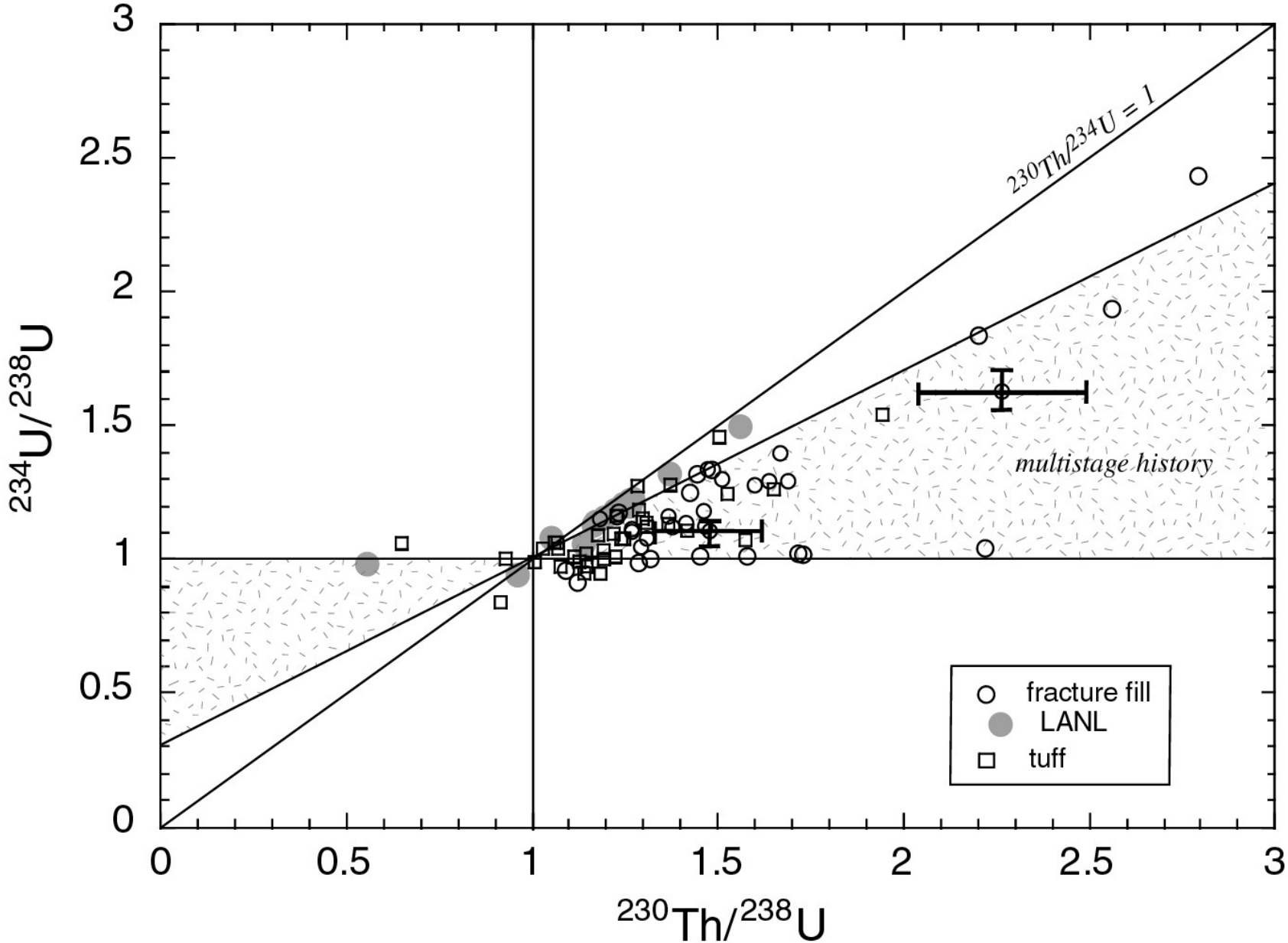
Uranium and thorium decay series isotopes reveal open system characteristics at Peña Blanca.

Preferential releases of alpha decay products are widely recognized in natural systems but absent from prior performance assessments for Yucca Mountain.

Nopal I Preferential U-234 Release



Uranium Decay Series Isotopes Open System Behavior
Pickett and Murphy, 1997, plus LANL data



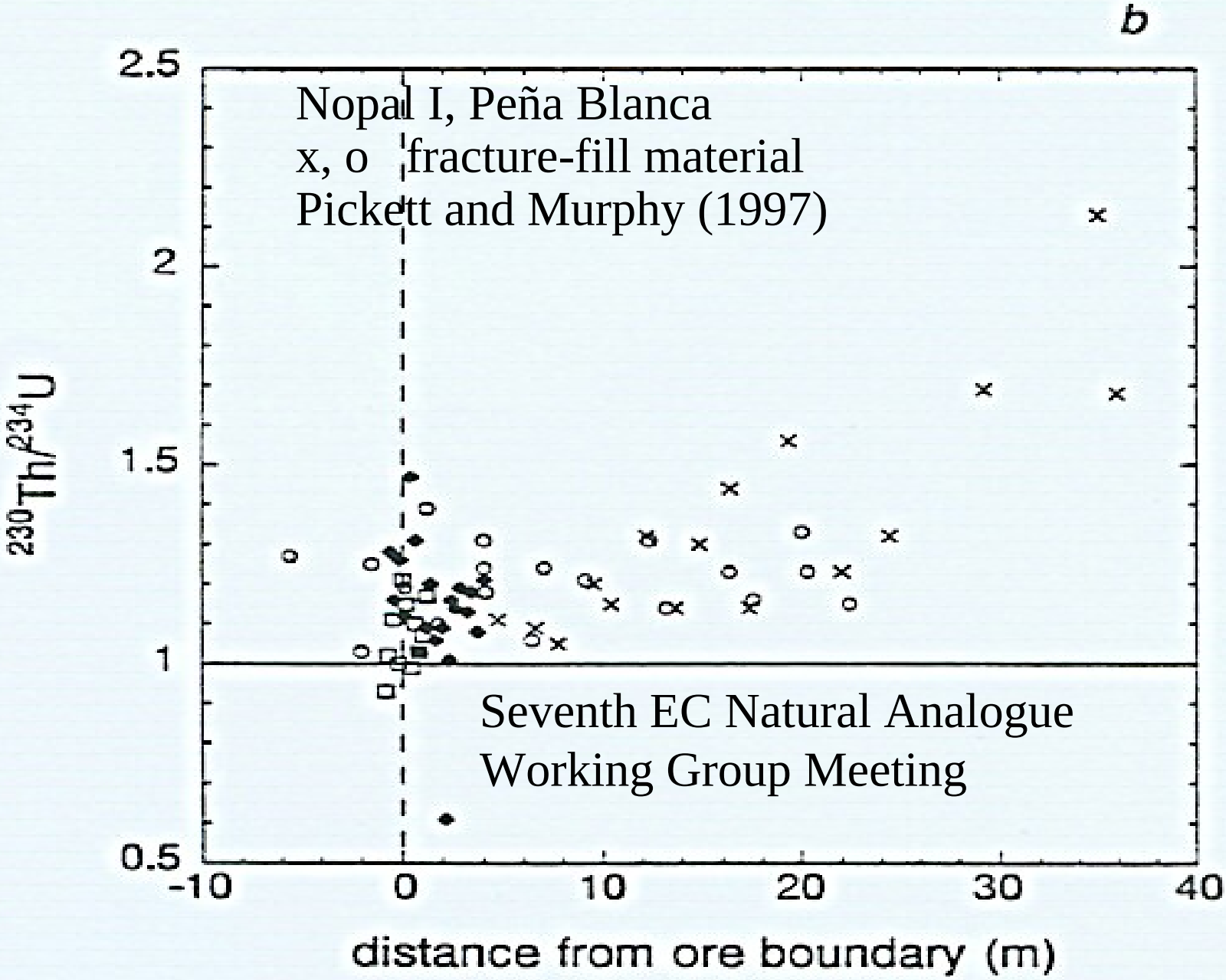

b
Nopal I, Peña Blanca
x, o fracture-fill material
Pickett and Murphy (1997)
Seventh EC Natural Analogue
Working Group Meeting
2.5
2
1.5
1
0.5
-10
0
10
20
30
40
$^{230}Th/^{234}U$
distance from ore boundary (m)

Table I. Radionuclides in commercial spent nuclear fuel at the proposed Yucca Mountain repository that may show enhanced release effects from alpha decay. The first seven listed radionuclides all grow in by several orders of magnitude toward equilibrium with their parents. Inventory from CNWRA [15].

Radionuclide	Maximum factor increase in inventory due to alpha decay	Percent of inventory occupying alpha decay sites at peak	Potential dose effect
^{210}Pb	Large by 10^5 y	100 %	High
^{226}Ra	Large by 10^5 y	100 %	High
^{227}Ac	Large by 10^5 y	100 %	Low
^{229}Th	Large by 10^5 y	100 %	Low
^{230}Th	Large by 10^5 y	100 %	High
^{231}Pa	Large by 10^5 y	100 %	Low
^{233}U	Large by 10^5 y	100 %	High
^{234}U	2.2 by ~400 y	53 %	High
^{235}U	1.7 by 10^5 y	41 %	Low
^{236}U	1.5 by 10^4 y	35 %	Low
^{237}Np	3.4 by ~10^3 y	71 %	High

Murphy and Pickett (2002)

Summary of Key Observations

- Secondary Minerals Control Releases.
- Alternate Models Show Performance Significance.
- Coprecipitation Data Are Inconclusive.
  - -Thermodynamic and kinetic data would help.
- Radioisotopes Demonstrate System Openness.
  - -Strong potential for quantifying transport
- Alpha Daughters Are Released Preferentially.
  - -Performance consequences are being explored.

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